1. PLENARY SESSION (PLN)

Lectures

PLN.L01 (Id: 122)
RADIOCHEMICAL ANALYSIS FOR THE DETERMINATION OF RADIONUCLIDES DIFFICULT TO MEASURE FOR CHARACTERISTICS AND DECOMMISSIONING OF NUCLEAR FACILITIES

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With increasing numbers of nuclear facilities, especially nuclear power reactors, being closed in recent years and from now on, a considerable work is going to be carried out all over the world for decommissioning these nuclear facilities. For this purpose, characterization of various wastes from decommissioning is required for evaluation of the radioactivity inventory in various materials and decision making for management of the produced waste. This is carried out by quantitative determination of various radionuclides present in the materials. The neutron activation products of components and impurity in the materials used in the nuclear facilities, such as ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{60}Co, ^{55}Fe, ^{63}Ni, ^{133}Ba, ^{152}Eu, ^{154}Eu, and some transuranics, are the main contributors to the total radioactivity, especially in the construction materials. But some long-lived fission products, such as ^{90}Sr, ^{99}Tc, ^{129}I, and ^{137}Cs, are the major concern for materials contaminated by spent nuclear fuel. Of these radionuclides, the gamma emitting radionuclides, such as ^{60}Co, ^{133}Ba, ^{152}Eu, ^{154}Eu, or ^{137}Cs, are easily measured by gamma spectrometry. However, the determination of pure β and α emitters including ^{3}H, ^{14}C, ^{36}Cl, ^{41}Ca, ^{55}Fe, ^{63}Ni, ^{90}Sr, ^{99}Tc, ^{129}I and some transuranics is the major challenges, because they could not be measured without separation from the matrix of the samples and from all other radionuclides, this entitles them as the radionuclides difficult to measure. Radiochemical analysis is the only way to complete the determination of the radionuclides difficult to measure by including a complete separation of individual radionuclides from the matrix and other radionuclides before measurement by β counting, α spectrometry, or mass spectrometry. Although plenty of analytical methods have been reported for the determination of these radionuclides since the discovery of radioactivity, the suitable methods are not always available for the purpose of decommissioning. This is because a large number of samples are required to be analyzed during the decommissioning, which needs simple and rapid methods to provide a good analytical capacity. In addition, the sample matrix varies very much from concrete, graphite, exchange resin, to various metals, which requires different radiochemical methods for different sample matrix and target radionuclides. In the past few years, our laboratory developed various radiochemical analytical methods aiming at characterization of various decommissioning waste by determination of various radionculides difficult to measure. This presentation gives an overview of these analytical methods with some examples including (1) rapid determination of tritium and ^{14}C in solid materials, such as graphite, concrete, steel, aluminium, paint, silica gel, soil, and dust; (2) determination of ^{14}C in high tritium samples, such as heavy water, waste water, and oil; (3) determination of ^{36}Cl and ^{129}I in graphite, steel, concrete, waste water, and dust; (4) determination of ^{41}Ca and ^{90}Sr in concrete; (5) determination of ^{55}Fe and ^{63}Ni in graphite, concrete, steel, aluminium, sediment, sand, waste water, seawater, and lichens.

PLN.L02 (Id: 370)
SPECIATION ANALYSIS OF RADIONUCLIDES IN THE ENVIRONMENT

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Naturally occurring and artificially produced radionuclides in the environment can be present in different physicochemical forms (i. e. radionuclide species) varying in size (nominal molecular mass), charge properties and valence, oxidation state, structure and morphology, density, complexing ability eTc. Low molecular mass (LMM) species are believed to be mobile and potentially bioavailable, while high molecular mass (HMM) species such as colloids, polymers, pseudocolloids and particles are considered inert. Due to time dependent transformation processes such as mobilization of radionuclide species from solid phases or interactions of mobile and reactive radionuclide species with components in soils and sediments. Therefore, the original distribution of radionuclides deposited in ecosystems will change over time and influence the ecosystem behaviour. To assess the environmental impact from radionuclide contamination, information on radionuclide species deposited, interactions within affected ecosystems and the time-dependent distribution of radionuclide species influencing mobility and biological uptake is essential. The development of speciation techniques to characterize radionuclide species in waters, soils and sediments should therefore be essential for improving the prediction power of impact and risk assessment models. The present paper will focus on analytical techniques which should be utilised for characterizing radionuclide species in the environment.

PLN.L03 (Id: 365)
ACCELERATOR MASS SPECTROMETRY - FROM ARCHAEOLOGY TO ASTROPHYSICS AND HUMAN DNA

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The advancement of Accelerator Mass Spectrometry (AMS) some 30 years ago opened the possibility to detect long-lived radionuclides (both natural and man-made) through isotopic abundance measurements down to a level of $\sim 10^{-16}$. It thus became possible to explore our world atom by atom in almost every section of the environment at large. Worldwide there are about 80 facilities in operation covering a large range of applications. The Vienna Environmental Research Accelerator (VERA) is an AMS facility for 'all' isotopes, based on a 3 MV tandem accelerator. The principle of AMS will be described with reference to this facility, and a few selected examples will be discussed. A well-known application of AMS is radiocarbon (^{14}C) dating in archaeology and other fields, where the counting of atoms as compared to the classical way of counting beta decays increased the ^{14}C detection sensitivity by literally a factor of a million. This allowed us to date small samples of a very precious skull of the earliest humans in Europe, excavated some 100 years ago from the Mladec cave near Olomouc in the Czech Republic[1], and preserved at the Museum of Natural History in Vienna. At VERA we also started to explore areas of the nuclear landscape beyond any known isotope, such as the one around $Z = 114$, $N = 186$, where an 'island of stability' for Super Heavy Elements (SHE) has long been predicted. If there are SHE isotopes with half-lives in the hundred-million-year range, it may be possible to find traces of them in natural materials left over from the formation of the solar system. We have conducted such an AMS search and will report on recent results[2] and future experiments. In the previous experiment we pursued experiments to find isotopes, which have been synthesized in stars before our solar system even existed.. The last example deals with events which happened during the last 50 years. Atmospheric nuclear weapons testing during the late 1950s and early 1960s led to a doubling of the ^{14}C content in the atmosphere. After the Nuclear Test Ban Treaty in 1963, the rapid distribution of the excess ^{14}C ('^{14}C bomb peak') into the biosphere including humans allows one to study the formation of new cells in the human body after birth[3]. This can be accomplished by extracting DNA from millions of cell nuclei and measuring the ^{14}C content in them. Brain cells, heart cells, and fat cells have been investigated so far, leading to new insights into cell birth dates and turnover times, respectively. In addition to the biomedical application, the ^{14}C bomb peak provides an important means to study the dynamics of the CO_2 exchange between the atmosphere and the hydrosphere and biosphere, respectively. This is of utmost importance for a deeper understanding of our climate on earth.

Refererences:

1. Wild, et al.: Nature *435* 332 (2005).
2. Dellinger, at al.: NIM B (2009) on line.
3. Spalding, et al.: Cell *122* 133 (2005).

PLN.L04 (Id: 288)
CHEMISTRY OF THE TRANSACTINIDES

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Advanced methods that have been developed and applied to study the chemical properties of the transactinide elements in comparison with their lighter homologs are presented.These include thermochromatography, and isothermal chromatography in the gas phase as well as ion-exchange and reversed-phase liquid chromatography in the aqueous phase, liquid-liquid extraction, and electrodeposition. Latest applications of these methods to studies of the chemistry of the transactinides are shown. From a systematic study of the anion-exchange behavior of Rf, it has been concluded that the properties of Rf in HCl and HNO_3 solutions are similar to those of Zr and Hf (H. Haba, et al. 2002, R. Günter, et al. 1998, H. Haba, et al. 2007). However, in HF solutions, the fluoride complex formation of Rf is significantly different to that of its homologs (E. Strub, et al. 2000, A. Kronenberg, et al. 2004, H. Haba, et al. 2004, Y. Nagame, et al., A. Toyoshima, et al. 2004). In dilute HNO_3/HF, the nitrate ion is the counter ion that removes the Rf hexafluoride di-anion more effectively from the binding sites of the anion-exchange resin than the Zr and Hf fluoride complexes. In strong HF solutions, it is the HF_2^- ion, that removes Rf much earlier from the anion-exchange resin than the much stronger fluoride complexes of Zr and Hf (H. Haba, et al. 2004, Y. Nagame, et al.). Chloride and fluoride complexation of Db has been studied in reversed-phase extraction chromatography with an aliphatic amine (W. Paulus, et al. 1999). Chelating of Db with α-hydroxyisobutyrate was shown to be much stronger than for tetravalent and trivalent metal ions (M. Schädel, et al. 1992). Sg has been eluted from a cation-exchange column as Sg oxyfluoride complex (M. Schädel, et al. 1997). In 0.1 M HNO_3, hydrolysis of Sg is weaker than that of Mo and W (M. Schädel, et al. 1998). Successive deprotonation leads to a cationic species for Sg, while for Mo and W, neutral hydrolysed species are eluted from a cation-exchange column. In the gas phase, Sg was volatilized as SgO_2Cl_2 (A. Türler, et al. 1998) and as $SgO_2(OH)_2$ (S. Hübener, et al. 2001). Elements 107 and 108 were transported in the gas phase as BhO_3Cl (R. Eichler, et al. 2000) and HsO_4 (Ch. E. Düllmann, et al. 2002). In (A. von Zweidorf, et al. 2004), the highly volatile HsO_4 was deposited on a thin layer of NaOH in the presence of water vapor, thus forming a salt in analogy to the formation of an osmate (VIII). This shows that HsO_4, like OsO_4, is an acid anhydride. Most recently, first chemical studies were performed with elements 112 (R. Eichler, et al. 2007) and 114 (R. Eichler, et al. 2009). Element 112 in its atomic state was shown to be very volatile, but unlike radon, reveals a metallic interaction with a Au surface (R. Eichler, et al. 2007). Preliminary results on the volatility of element 114 in its atomic state indicate that it is a very volatile element with a weaker metallic interaction with a Au surface than 112 (R. Eichler, et al. 2009). This surprising result needs to be confirmed.

PLN.L05 (Id: 366)
ELECTROCHEMICAL SEPARATION OF ACTINIDES FROM MOLTEN LiCl-KCl ON SOLID Al CATHODES

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To comply with the sustainability goals defined for innovative reactor systems, a waste minimization through recycling of all actinides is required and can be realised using a so-called Partitioning and Transmutation (P&T) concept inherent to the reactor systems. It is obvious that the corresponding fuel cycles will play a central role because an efficient and selective recovery of the key elements is mandatory. One of the major goals is to develop and implement clean waste technologies using a global actinide management. For this purpose, advanced fuel cycle processes with full actinide fuel technologies and ultimate waste forms with a minimal content in actinides (< 0.1%) are required. This necessitates that Am and Cm can be selectively separated from lanthanide fission products, certainly the most difficult and challenging task in advanced reprocessing of spent nuclear fuel due to the very similar chemical behaviour of these elements. The separation can be derived from aqueous or pyrochemical partitioning processes. Both have advantages and disadvantages and should be applied in a complementary way. In Europe, significant progress was made for both routes in international collaborations in the frame of the European research projects PARTNEW, PYROREP, EUROPART and ACSEPT. Pyrochemical processes rely on refining techniques in high temperature (around 500 °C) molten salt (e.g. LiCl/KCl eutectic). In nuclear technology, these processes are mostly based on electrorefining or on extraction from the molten salt phase into liquid metal. In the electrorefining process the fuel is anodically dissolved, releasing actinides and lanthanides mainly as MCl_3 species into the molten salt electrolyte. The separation is based on electrodeposition of the actinides on a cathode leaving the fission products in the bulk. The choice of cathode material onto which the actinides are deposited during electrolysis is essential. Molten Cd and Bi have been widely used because they are reactive electrodes, i.e they form stable alloys with the actinides. This alloying inhibits the re-dissolution of the electrodeposited trivalent actinides by a comproportionation reaction with not deposited An(III) in the salt melt and thus allows an efficient recovery of An metal. The redox potentials on solid cathodes show a difference in the reduction potential between actinides and lanthanides which is about 3 times higher than for liquid cathodes, improving obviously a separation of these two element groups. Solid Al cathodes unite both advantageous properties, i.e. the large difference in the electrodeposition potential and the reactivity to form alloys. Therefore we use Al electrodes for our electrochemical partitioning studies. In view of a grouped actinide recycling, foreseen in the fuel cycles of new generation reactors, electrorefining experiments using a fuel with the overall composition $U_{61}Pu_{22}Zr_{10}Am_2Ln_5$ were carried out. The metallic alloy was anodically dissolved in a LiCl-KCl eutectic and the actinides were co-deposited on Al cathodes as alloys, leaving more than 98% of the lanthanides in the salt phase. The results of a multiple-run electrorefining experiment without changing the salt bath, are very promising in view of a large-scale development of pyroreprocessing in advanced nuclear fuel cycles. They demonstrate that a grouped actinide recovery better than 99.9% are possible and therefore represent an important step towards achieving the sustainability goals of future reactor systems.

PLN.L06 (Id: 297)
RADIOACTIVE AND GASTRIN RELEASING PEPTIDE RECEPTOR SPECIFIC GOLD NANOPARTICLES IN MOLECULAR IMAGING AND THERAPY OF CANCER

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The most recent study involving 77,000 North American men has shown that regular prostate specific antigen (PSA) screening did not save a significant number of lives over 10 years. Development of cancer receptor specific gold nanoparticles will allow efficient targeting/optimum retention within tumors and thus provide synergistic advantages in oncology as it relates to molecular imaging and therapy of prostate cancer. Bombesin (BBN) peptides have demonstrated high affinity toward Gastrin Releasing Peptide (GRP) receptors in vivo that are over expressed in prostate, breast, and small cell lung carcinoma. We have synthesized a library of GRP receptor-avid nanoplatforms by conjugating gold nanoparticles (AuNPs) with Bombesin (BBN) peptides. Cellular interactions and binding affinities (IC50) of AuNP-BBN conjugates toward GRP receptors on human prostate cancer cells have been investigated in detail. In vivo studies using AuNP-BBN and its radiolabled analog ^{198}AuNP-BBN, exhibiting high binding affinity (IC50 in microgram/nano/pico molar ranges), have provided unequivocal evidence that AuNP-BBN constructs are GRP receptor specific showing accumulation with high selectivity in GRP receptor rich pancreatic acne in normal mice and also in tumors in prostate tumor bearing SCID mice. The selective uptake of AuNP-BBN peptide analogs have demonstrated realistic clinical potential in molecular imaging via X ray CT techniques as the contrast numbers in prostate tumor sites are several fold higher as compared to the pretreatment groups (ΔHU = 150 units). On the therapeutic front, recent results on therapeutic efficacy and clinical translation efforts of GA-^{198}AuNP (NBI-29) - a glyco protein matrix-conjugated radioactive gold nanoparticulate therapeutic agent will be discussed. Intratumoral administration of a single dose of β-emitting GA-^{198}AuNP (70 Gy) resulted in clinically significant tumor regression and effective control in the growth of prostate tumors over 60 days and the overall reduction in tumor volume reached an unprecedented 82%. This presentation will include: (a) details on clinical utility of AuNP-BBN as a tumor specific molecular imaging agent for X ray CT imaging of prostate and other GRP receptor positive cancers; (b) details on clinical translation efforts of GA-^{198}AuNP (NBI-29) with early Phase I clinical trial results involving therapeutic efficacy in treating prostate tumor bearing dogs. The overall oncological implications on how GA-^{198}AuNP and cancer specific peptide conjugated gold nanoparticles will provide significant benefits to prostate tu, pancreatic, and breast tumor patient community will be discussed.

2. CHEMISTRY OF NUCLEAR FUEL CYCLE, RADIOCHEMICAL PROBLEMS IN NUCLEAR WASTE MANAGEMENT (NFC)

Lectures

NFC.L01 (Id: 92)
KINETICS OF REDUCTION OF HEXAVALENT NEPTUNIUM BY NITROUS ACID IN SOLUTIONS OF NITRIC ACID

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Extraction of hexavalent and tetravalent actinides with tributylphospate (TBP) from their solutions in nitric acid is the most industrially mature process for reprocessing spent nuclear fuel and is the basis for the development of advanced reprocessing schemes like UREX and TRUEX. One of the less resolved issues is the management of neptunium in the extraction system, because of not clearly defined redox speciation. Neptunium is present in both pentavalent and hexavalent oxidation states that differ greatly in their extractability to the organic phase (hexavalent state being very well extracted by TBP, whereas pentavalent neptunium is almost not extracted). As a result, neptunium is distributed in both organic and aqueous streams of the separation process. Apparently, the most important factor determining the redox speciation of neptunium is the presence of nitrous acid that is formed during the dissolution of spent fuel and also by radiolysis. Nitrous acid HNO_2 is relatively well extracted by TBP ($\log D \approx 1$) and is therefore distributed from the source raffinate solution in further stages of the separation system. If present in small quantities, it autocatalyzes the oxidation of Np(V) by nitric acid, whereas large concentration of HNO_2 leads to reduction of Np(VI) back to the pentavalent state - the kinetics of this reaction have been covered in detail by many authors. The kinetics of reduction of Np(VI) by HNO_2 have so far attracted only little interest and are the subject of this study. UV-VIS-NIR absorption spectrophotometry (OceanOpticsTM) in a stirred cuvette was employed in order to study the reaction: $2Np(VI) + HNO_2 + H_2O \leftrightarrow 2Np(V) + NO_3^- + 3H^+$ The reaction progress was recorded by following the absorbance at characteristic peak of Np(V) at 981 nm and simultaneous observation of the broad absorption peak of HNO_2 between 350-400 nm. The reaction orders with respect to concentrations of Np(VI) and HNO_2 were found to be close to one. Increasing the concentration of nitric acid at constant ionic strength ($\mu = 4M$ $H/LiNO_3$) had a decreasing effect on the reaction rate with an order of -0.8. The dependence of the rate constant on the ionic strength and temperature will be also reported.

NFC.L02 (Id: 164)
ELECTRO-FLUID ANALYSIS OF A MOLTEN-SALT ELECTROREFINER FOR PYROCHEMICAL NUCLEAR WASTE TREATMENT

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Pyrochemical technologies offer the capability to treat spent nuclear fuels and were acknowledged to meet the proliferation-resistant principles. These processes could play an important role in reducing the long-term hazards of a spent nuclear fuel by separating uranium and transuranic actinides, which in turn may be transmuted in a fast breeder reactor. The most effective way to accelerate the development of these technologies is to formulate physical models of the underlying electrochemical and transport processes. The multi-physics computational models can be essential for design and operational analysis of advanced processors and offer an efficient approach to implementing these processes. In this study, the electrochemical principles and forced convection of molten eutectic (LiCl-KCl) electrolyte in a uranium electrorefining cell are considered to establish an appropriate electro-fluid model within the 3-dimensional framework of a conventional computational fluid dynamic model. Diffusional mass transport as a controlling step is modeled for the surface resistance of charge transfer between the electrode and molten-salt phase in which a constant composition is provided by an intense forced mixing of the bulk solution. The approach in this study is focused on the mass transport and current arising due to the concentration and the surface overpotential based on a cell configuration. This computational platform includes the electrochemical reaction rate of charge transfer kinetics which is described by a Hitler-Volmer equation, while mass transport is considered using an ionic transport equation. The coupling of the local overpotential distribution and uranium concentration gradient makes it possible to predict the local current density distribution at the electrode surfaces.

NFC.L03 (Id: 295)
ELECTROCHEMICAL SEPARATION OF ACTINIDES FROM MOLTEN LiCl-KCl ON SOLID Al CATHODES

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An electrorefining process in molten chloride salts using solid aluminium cathodes is being developed in the Institute for Transuranium Elements to recover all actinides from metallic spent nuclear fuel. In this process, actinides are group-selectively electrodeposited on the cathode in a form of solid actinide-aluminium alloys. Fission products are anodically co-oxidised from the fuel together with actinides to the electrolyte. Without purification of this carrier salt, the process would have to be stopped after concentration of the dissolved fission products would become too high to prevent selective deposition of actinides on the cathode. A multiple-steps procedure is considered for cleaning of the salt and a process refered as 'exhaustive electrolysis' is proposed for the first purification step. Similarly to the electrorefining process, this technique is based on the group-selective electrodeposition of actinides on solid aluminium cathodes forming actinide-aluminium alloys. On the anodic side, chlorine gas is produced by electrochemical decomposition of the carrier salt. The presented work was carried out in order to prove feasibility of the method. Two galvanostatic electrolyses were realised and the potentials of both electrodes were constantly followed. Uranium was successfully recovered from LiCl-KCl melts containing UCl_3 and a mixture of $UCl_3^{-}NdCl_3$ and its concentration decreased from 1.7 to 0.1 wt. % with no co-deposition of neodymium. Although the maximum applicable current densities were relatively low, the results are promising, showing high current efficiency and selectivity of the proposed method. A design and application of a special chlorine gas producing inert anode is also discussed.

NFC.L04 (Id: 279)
ADVANCED ORIENT CYCLE - PROGRESS ON RESEARCH, WITH FOCUSING ON SAFETY AND ENGINEERING

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A multi-functional separation process is proposed on the Advanced-ORIENT Cycle (Advanced Optimization by Recycling Instructive Elements) concept [1]. The tertiary pyridine-type anion-exchange resin embedded in silica beads was proposed for the separation process using spent fuel. In this process, hydrochloric acid (HCl), mixture of nitric acid (HNO_3) and methanol (MeOH) are used as eluents. In order to apply this process to engineering scale, two important subjects should be solved so as to prove the availability. One is the environmental aspect for the use of HCl solution, because of its corrosive property to the material. The other is explication of the reactive safety between IER (ion exchange resin) and solvent (HNO_3 - MeOH). The four candidate metals of Ta, Zr, Nb, Hastelloy-B (28%Mo-Ni) and SUS316L as a reference were tested. The conditions of immersion and acid mist-exposure tests were conducted in 0.5~12 M pure and simulated high level liquid waste (SHLLW) HCl at room temperature for maximum 7200 h and at 90 °C for 72~1440 h. In addition, corrosion potential was evaluated by electrochemistry measurement under the same conditions. From the result of the corrosion experiment, Ta was anti-corrosive in HCl media, and Hastelloy-B seemed to be acceptable at RT, with evidence of low corrosion rate (<0.1 mm/y) and general type corrosion. Thermal analysis by DSC was performed to investigate the thermal stability of TPR itself and the mixtures of TPR/MeOH/HCl or HNO_3. Moreover, the thermal stability of TPR mixtures of TPR and SHLLW-NO_3 was investigated and the stability of presence or absence of coexistent elements was compared. Sudden exothermic heat and release of fumes were observed with a mixture of TPR-NO_3/MeOH/HNO_3 when the temperature of the heater reached 220 °C regardless of the HNO_3 concentration. The temperature increased slowly to 175 °C followed by a sudden increase in temperature. Violent reaction occurred in TPR under high temperature in the HNO_3 system. The reaction occurred regardless of HNO_3 concentration and presence or absence of MeOH. However, it was confirmed that the reaction did not occur when the heating temperature decreased. In this report, outline of current progress of Adv-ORIENT research as well as detailed evaluation results of this safety and engineering study are described.

Reference:

1. Ozawa M., Suzuki T., Koyama S., Akatsuka H., Mimura H., Fuji Y.: Progr. Nucl. Energy *50*, 476 (2008).

NFC.L05 (Id: 191)
COBALT BIS(DICARBOLLIDE) IONS WITH COVALENTLY BONDED TODGA-LIKE SUBSTITUENTS FOR ACTINIDE AND LANTHANIDE EXTRACTIONS

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The partitioning and transmutation of long-lived nuclides such as minor actinides from high level waste (HLW) issued from nuclear fuel reprocessing is a method how to reduce the long-term environmental burder of HLW. In recent years, several HLW partitioning processes using different extractant agents have been developed. Among them, the tridentate ligands like N,N,N´,N´-tetraoctyl diglycolamide (TODGA) are one of the most promising extractants for the recovery of actinides and lanthanides in the nuclear fuel treatment. In this work, the recently developed extractants based on cobalt bis(dicarbollide) ion(1-), [(1,2-$C_2B_9H_{11}$)2-3-Co)]- (COSAN) functionalized with TODGA like substituents were evaluated for extraction of trivalent actinides and lanthanides from acidic waste solutions. The extractants under study differed in substitution of the amide nitrogen (e.g., by butyl-, octyl-, tert-octyl-, dodecyl-, benzyl- groups) and the mode of attachment of two cobalt bis(dicarbollide) anions (with or without spacer) to diglycolyl acid platform by amidic bonds. It was found that the compounds with longer diethyleneglycol connectors between COSAN cage and diglycolamide group extracted trivalent lanthanides and actinides very effectively even from highly acidic nitric acid solutions. Additionally, the extraction efficiency of such derivates was significantly higher than that of comparable organic TODGA molecules without COSANs. The most perspective compound from this series corresponds to the formula X-[(8-CH_2-$CH_2O)_2$-1,2-$C_2B_9H_{10}$)(1',2'-$C_2B_9H_{11}$)-3,3'-Co]$_2$, where central amidic unit X corresponds to [(n-C_8H_{17}-$NCOCH_2)_2O$]. We shall demonstrate that this extractant enables good extraction of trivalent lanthanides and actinides from majority of fission products presented in the simulated PUREX feed. Trivalent radionuclides can be effectively stripped using complexants. This extractant will be further studied for possible technological applications.

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NFC.L06 (Id: 35)
EXTRACTION OF RADIONUCLIDES WITH SOLUTIONS OF COMPLEXING AGENTS IN SUPERCRITICAL FLUIDS AND COMPRESSED GASES

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One of the main advantages of extraction with supercritical fluids (SCF) or liquid gases (LG) is a possibility of the reagent-free regeneration of "solvent", which sharply decreases the secondary waste volume. As a result, in the last 20 years the researchers in many countries focused their attention on the supercritical fluid extraction (SFE) of the trace amounts of radioactive and heavy metals for treatment to remove and decontamination of various solid objects (including soils) and on SFE of actinide macroamounts for reprocessing the spent nuclear fuel of the nuclear power plants (SNF NPP). In this work we analyze the similarities and differences between the liquid-liquid extraction of radionuclides and extraction with SCF and LG and the prospects of extraction using supercritical and liquid gases in radiochemistry. In the last 20 years the data on SFE of a series of metals were published in more than 100 papers. The detailed data was presented on SFE of U, Th, rare-earth elements (REE), Co, Cu, Mn, Pu, Np, and Am. The SFE of U, Th, and REE with tri-n-butyl phosphate (TBP) and its mixture with β diketones were also extensively studied. Rather detailed data were presented on SFE of transuranium elements and fission products using carbon dioxide solutions of TBP, β diketones and their mixtures and even on recovery of these elements from non-irradiated nuclear fuel and from SNF NPP. SFE of Cs and Sr with solutions of organic acids and polyethers was also well studied. The detailed data are published of the extraction of U, REE, Cu, Pb, Zn, Cd, Cr, and As using carbon dioxide solutions of various acids such as di-2-ethylhexyl phosphoric, bis(2,4,4-trimethylphenyl) phosphoric (cyanex 272), bis(2,4,4-trimethylphenyl)monothio phosphoric (cyanex-302), bis(2,4,4-trimethylphenyl)dithio phosphoric (cyanex-301), and di-2-ethylhexylthio phosphoric acids. Our analysis showed only slight differences between the extraction of macro- and microamounts of metal cations with solutions of complexing agents in liquid and supercritical CO_2 and liquid-liquid extraction. At the same time it should be noted that:

- low viscosity and high diffusion rate allows performance of extraction in CO_2 without mixing of the phases;
- CO_2 can affect pH of the liquid phase being in equilibrium with it;
- main difference between the extraction of metal cations with solutions in liquid and supercritical CO_2 and liquid-liquid extraction are observed for microamounts of metals.

Moreover, it should be mentioned that the technologies of SNF reprocessing and procedures of deactivation in supercritical and compressed CO_2 and Freons are most promising for radiochemistry. The advantages of these processes are sharp decrease in the volume of secondary liquid radioactive wastes; absence of organic solvents and toxic compounds, and rather low operation temperature (25-80°C). It should be noted that the use of Freons instead of CO_2 allows to decrease the working pressure to 1- 4 MPa.

NFC.L07 (Id: 81)
REMOVAL OF ANTIMONY-124 FROM PWR COOLANT WATER

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Selective ion media, e.g. inorganic adsorbents and ion exchangers, are increasingly used for the removal of key radionuclides such as ^{60}Co, ^{90}Sr and ^{137}Cs from nuclear waste effluents due to their radiation stability, high processing capacity and high decontamination efficiency. The materials used that are commercially available (e.g. zeolites, titanates, silicotitanates, hexacyanoferrates) are cation exchangers or adsorbents, which can remove efficiently cationic and neutral radioactive species. Inorganic anion exchange materials are quite rare and do not possess high selectivity. Considering radiation doses to personnel and environment, ^{58,60}Co and ^{137}Cs are the most critical radionuclides in nuclear power plant (NPP) waste liquids and water streams. Improved processing systems have been able to reduce markedly the discharges of these radionuclides at many utility sites and further efforts have been directed to remove other radionuclides such as ^{51}Cr, ^{110m}Ag and ^{125}Sb that dominate in solution after cesium and cobalt elimination. Much attention has been paid recently to ^{125}Sb. It may exist completely in soluble form in the Floor Drain Waters. In solution, antimony can exist in two oxidation states (+3,+5) and in several hydroxyl species (e.g. $Sb(OH)_6^-$, $Sb(OH)_3$ (aq), $Sb(OH)_4^+$), depending on the pH and redox conditions. This indicates that antimony is difficult to remove from solution. Recent tests showed that standard demineralizer resins and ion selective media are ineffective for the removal of Sb from liquid radwaste. However, some commercially available inorganic cation exchangers, such as CoTreat, can remove cationic species of ^{125}Sb from NPP Floor Drain water with good efficiency in some cases. Chemical additives coupled with ultra filtration have been shown to be effective method for ^{125}Sb removal in a test program conducted at Duke Power Company's Oconee plant. Study of other methods such as electro-deionization and hollow-fibre filtration is underway e.g. in the EPRI Low-Level Waste program. In Loviisa NPP (PWR, Finland) about 50 % of the radiation dose received by personnel during the service shut-down period is caused by ^{124}Sb. Immediately after shut-down, ^{124}Sb is released from fuel into the primary coolant water at levels greater than 100,000 Bq/L (>3x10^{-3} μCi/mL). The mixed-bed organic resin demineralizer system used for activity removal under routine reactor operation is inefficient in removing the released ^{125}Sb. Loviisa NPP has thus established a project to study and assess various ^{124}Sb abatement technologies. As a part of this project, Laboratory of Radiochemistry has just initiated a laboratory study to remove ^{124}Sb from the primary coolant water of Loviisa NPP using novel zirconium oxide (ZrO_2) sorbents. Test samples were obtained from service shut-down of Loviisa Unit 1. Water samples contained 600,000 Bq/L of soluble ^{124}Sb after filtration.

NFC.L08 (Id: 69)

ANALYSIS OF Th, U, Pu, AND Am IN RADIOACTIVE METAL WASTE USING EXTRACTION CHROMATOGRAPHY

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Lots of metal waste the surface of which is contaminated with radionuclides are generated in the decommissioning of nuclear facilities. For the waste management, characterization of radionuclides inventory in the waste is required. In this work, determination method of α nuclides, Th, U, Pu, and Am, contained in the metal waste was developed. Taking into account the half-lives of these α nuclides and their expected concentration in the waste, inductively coupled plasma mass spectroscopy (ICP-MS) and α-ray spectrometry were selected as analytical tools. The determination using these techniques requires that these α nuclides in the metal waste were separated from the large quantities of stable matrix and interfering radionuclides. In our laboratory, we have applied commercially available extraction chromatographic resin to the analysis of α nuclides in a dissolved solution of molten products of low-level radioactive waste; UTEVA resin (for uranium and tetravalent actinide resin) for the analysis of Th and U, and TRU resin (for transuranium resin) for the analysis of Pu and Am. In a similar manner as above, application of these extraction chromatography methods to the metal waste was planned. In the case of the metal waste, we have to take into account the existence of large amounts of Fe, because Fe(III) has significant negative impact on the sorption of trivalent actinides on TRU resin. Hence, the effects of Fe(III) amount and concentration in the sample solution on the uptake of Am, representative of trivalent actinide, on TRU resin were studied in detail with the solution prepared from $Fe(NO_3)_3$. Fortunately, when Fe(III) is reduced to Fe(II) with ascorbic acid, its effect on the sorption of trivalent actinide is practically negligible. The amount of ascorbic acid required to improve the extraction of Am was also studied. Based on this study of the effect of Fe(III) on the extraction of α nuclides on the extraction resin, the schemes to analyze α nuclides in metal waste was optimized. The optimized methods were validated with dissolved solution of stainless steel, SUS304 with added α nuclides. The recoveries of Th and U from dissolved solution of SUS304 were 89±4% and 102±5%, respectively, with optimized scheme using UTEVA resin. The recoveries of Pu and Am were 96±4% and 89±7%, respectively, using TRU resin. Furthermore, Th, U, Pu, and Am in simulated waste prepared by dissolving pipe wastes (the surface ^{60}Co concentration was about 70 kBq) sampled from a coolant system of nuclear reactor and by adding α nuclides were analyzed with the optimized method. The ^{60}Co was sufficiently separated from α nuclides. The recoveries of Th, U, Pu, and Am were 91±3%, 103±4%, 97±4%, and 91±3%, respectively. These recoveries are comparable to that analyzed dissolved solution of SUS304. These results support the usefulness of the optimized schemes.

NFC.L09 (Id: 144)

MICRO-SCALE INVESTIGATIONS OF U(VI) IMMOBILIZATION BY CEMENTITIOUS MATERIALS

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Cement-based materials play an important role in multi-barrier concepts developed worldwide for the safe disposal of low- and intermediate-level radioactive waste. Cement is used to condition the waste materials and for the construction of the engineered barrier system (container, backfill and liner materials) in deep geological repositories. The cementitious near field is subject to chemical alteration processes due to the interaction of cement with groundwater infiltrating from the host rock. A molecular-level understanding of the interaction of radionuclides with cement improves long-term predictions of radionuclide retention in cement-based repositories with regards to influences of the heterogeneity of the cement matrix and the chemical evolution of the cementitious near field with time caused by cement degradation. In this study the speciation of U(VI), which is the dominant oxidation state under oxidizing and slightly reducing conditions in cementitious environments, was investigated by a combination of wet chemistry and synchrotron-based (micro-)spectroscopic and micro-diffraction techniques. Wet chemistry experiments were carried out to quantify U(VI) uptake by cement. Sorption isotherm measurements enabled us to distinguish U(VI) retention by sorption from solubility-controlled effects. Complementary to wet chemistry experiments, we carried out synchrotron-based X-ray absorption spectroscopy (XAS) studies on U(VI) doped crushed cementitious materials (bulk-XAS) and micro-X-ray fluorescence (micro-XRF), micro-XAS and micro-diffraction studies (micro-XRD) on U(VI) doped compact hardened cement pastes (HCP) to obtain information on the U(VI) speciation on the molecular level. Micro-scale studies provided structural information on U(VI) binding mechanisms which are essential for the development of thermodynamic models of U(VI) uptake by cementitious materials. To the best of our knowledge, the combined use of bulk-XAS, micro-XAS and micro-XRD techniques for speciation studies on U(VI) doped HCP materials is novel. The synchrotron-based studies showed that in the cement samples with low U(VI) concentration, i.e., in the linear range of U(VI) sorption, U(VI) was predominantly bound onto calcium silicate hydrates (C-S-H). The latter phases are among the most important cement phases governing immobilization processes. The coordination environment of U(VI) was found to resemble that of U(VI) in uranyl silicate minerals under these conditions. At high U(VI) loadings, however, where U(VI) immobilization is controlled by a solubility-limiting process, a second U(VI) species was observed in addition to uranyl-silicate-like U(VI) coordination. The coordination environment of this species was similar to that of U(VI) in Ca-uranate. This study demonstrates that C-S-H are the uptake-controlling phase for U(VI) in HCP. This finding, in combination with the observed long-term existence of C-S-H phases in an evolving cementitious near field, implies that safe disposal of U(VI) in a cement-based repository should be possible over a very long period of time.

NFC.L10 (Id: 80)

NEW f-ELEMENT PHOSPHATES WITH LANGBEINITE-TYPE STRUCTURE

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Crystallochemical principles make possible to create new compounds with expected structure and properties. This approach was used for "constructing" new f-element phosphates with langbeinite-type structure (Lb). Formerly, the same approach was used for the calculation of compositions of tetrahedral-octahedral frameworks for NZP and Lb forms [1], later some new phosphates with Lb structure were prepared [2-4], namely: $K_2RZr(PO_4)_3$, R = Pr-Lu,Y; $K_2R_{3/2}Ta_{1/2}Zr(PO_4)_3$, R = Gd-Yb,Y; $A_2RZr(PO_4)_3$, A = Rb,Cs; R = Pr,Er; $KBaR_2(PO_4)_3$, R = Yb,Er; $CsBaR_2(PO_4)_3$, R = Dy-Yb; $ABaFe_{2-x}Pr_x(PO_4)_3$, A = K,Cs, x = 0.25; 0.5; 0.75. As a development of this research we have calculated the formula phosphate compositions with expected Lb-type structure with tri- and tetra-valent elements in the interframework sites: $[A_{3/2}R_{1/2}]^{3+}$; $[A_{5/3}M_{1/3}]^{3+}$; $[AR]^{4+}$; $[A_{4/3}M_{2/3}]^{4+}$, A,R,M - uni-, tri-, tetra-valent cations, also f-elements. We were going to prepare the phosphates: $A_{3/2}Ln_{1/2}MgZr(PO_4)_3$, $A_{3/2}Ln_{1/2}Fe_2(PO_4)_3$ and $ALnMg_{3/2}Zr_{1/2}(PO_4)_3$, where A = K, Rb, Cs; Ln = Pr, Sm, Yb. Synthesis was carried out by sol-gel technology. The IR, X-Ray and DSC analyses have been used for samples characterization. Optimum conditions were as follows: pH = 7-9; T = 600 and 800°C during 24 h on every stages, quick addition of H_3PO_4 solution; ultrasonic activation was ineffective. The phosphates $A_{3/2}Ln_{1/2}MgZr(PO_4)_3$ and $A_{3/2}Ln_{1/2}Fe_2(PO_4)_3$ higher presented with cubic structure, sp. gr. P213 were formed. An attempts to prepare phosphates with bigger concentration of Ln-cations in the interframework positions such as ALn or $A_{1.2}Ln_{0.8}$ was unsuccessful. The additional X-ray reflections of the monazite type phases presented on XRD patterns. Monazite phase as an admixture was also present in all phosphate samples prepared. The temperature limits of existence of Lb type phases were established: from 750 - 800 up to 900 - 1150 °C. The thermal decomposition products were identified as phosphates with NZP- or monazite-type structures and as phosphate $Cs_7Fe_7(PO_4)_8O_2$ in the case of iron-containing compounds. The lanthanide ions in LnO_n polyhedra in the structure of new phosphates are characterized by ninefold coordination in cavities. In known phosphates with such structure with Ln-cations located in framework sites the coordination is $n = 6$ [2,3]. Crystal chemical principle allows to predict new phosphate compositions with expected Lb-type structure containing lanthanide cations in sites of both types simultaneously. This may be realized as the subject of future investigations.

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NFC.L11 (Id: 23)
DETERMINATION OF ^{237}Np, ^{93}Zr AND OTHER LONG-LIVED RADIONUCLIDES IN MEDIUM AND LOW-LEVEL RADIOACTIVE WASTE SAMPLES

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The majority of long-lived radionuclides produced in the nuclear fuel cycle can be regarded as “difficult-to-determine nuclides” due to the low activities and/or the absence of γ-radiation of medium or high energies in their decay schemes. Most isotopes of actinoides are α-emitters, ^{90}Sr (fission product) and ^{93}Zr (activation product) emit almost exclusively β-particles, ^{93m}Nb (activation product) decays by isomer transition or electron capture and emits only X-rays, ^{94}Nb (activation product) emits low intensity γ-radiation due to its low activity. Chemical separation is needed before the nuclear measurement of all the isotopes mentioned above. A combined radiochemical separation method had been developed that enables the simultaneous determination of ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, $^{239-240}$Pu, ^{238}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm, ^{89}Sr, ^{90}Sr, ^{93m}Nb, and ^{94}Nb. Recently, this method has been extended for the determination of ^{237}Np and ^{93}Zr. The main steps of the method are addition of tracers and carriers, sample destruction, co-precipitation on iron(II) hydroxide and calcium oxalate, separation by extraction chromatography using supported dipentyl-pentylphosphonate (UTEVA), supported N,N-octylphenyl-di-i-butylcarbamoylmethyl phosphine oxide (TRU) and supported bis-(t-butylcyclohexano)-crown(18,6)ether (Sr.Resin), separation on anion exchange resin, α- and β-source preparation, α-spectrometry, liquid scintillation counting (LSC), γ-spectrometry, measurement by inductively coupled plasma mass spectrometry (ICP-MS). ^{237}Np and ^{93}Zr-93 are pre-concentrated by co-precipitation on iron(II) hydroxide and zirconium oxide, and separated by extraction chromatography using UTEVA. The key parameter of the method is the adjustment of the oxidation state of the actinoides before adding the sample onto the UTEVA column. This can be done using many redox agents (for example potassium bromate, sodium nitrite or ammonium peroxo disulfate). Highest yields were achieved when ammonium peroxo disulfate was used with silver nitrate as catalyst. As even traces of isotopes with mass number near 237 or 93 cause considerable interferences during ICP-MS detection, a purification step by extraction chromatography was inserted. It was determined by model experiments, that even a small amount of fluoride anions inhibits the retention of Zr on UTEVA, but this problem can be eliminated by addition of boric acid. Analyzing real samples (evaporation concentrates of a nuclear power plant) 66-97% and 31-66% chemical yields were achieved for Np and Zr, respectively.

NFC.L12 (Id: 60)
SORPTION OF Ni AND Eu IN A MULTI-ELEMENT SYSTEM

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At present the immobilisation of High Level Waste (HLW) in vitreous matrices followed by their burial in deep geological repository systems, composed of natural and engineered barriers to isolate the long lived radionuclides from the biosphere, is the most preferred procedure. Granitic rock formations are being considered as host rocks for such geological repositories [1]. The study of the sorption of radionuclides onto geological media is, therefore, an important part of the safety assessment of deep geological disposal of radioactive waste. Due to the many combinations of adsorbents, data collection in multicomponent systems (MCS) is complex; therefore mathematical models have been developed to predict multicomponent (MC) sorption based on the adsorption properties of each element [2]. The problem of predicting adsorption based on the information of single component isotherms is still a challenge in adsorption studies. Multi-element sorption systems were examined in the last century, however, none of these studies dealt with competitive adsorption and only a few dealt with the selectivity of the sorption processes [3]. Several isotherm models have been used to model experimental data obtained from mixed radionuclide systems. One of the commonest of these models is shown below.

- $Q_{mix}/Q_0 > 1$, the sorption is promoted by the presence of other metal ions,
- $Q_{mix}/Q_0 = 1$, there appears to be no observable effect and,
- $Q_{mix}/Q_0 < 1$, the sorption is suppressed by the presence of other metal ions in solution.

Static baTch sorption experiments with 0.2 g of granitic rocks and different granitic minerals with 40 cm^3 of non active Eu and Ni solutions have been performed in systems of single and multiple elements. Solutions were doped with ^{63}Ni and ^{152}Eu, acting as analogues for di- and tri-valent elements. Equilibration periods were between 7 and 10 days, after which radiometric methods (Liquid scintillation counting and γ spectroscopy were used to determine the sorption patterns. The results obtained showed that generally Eu sorption to the 6 granitic materials studied is affected by Ni competition, except in the case of Adamellite granite. Ni sorption to granitic materials was not affected only in the presence of Eu in solution in the case of biotite mica.

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NFC.L13 (Id: 63)
SORPTION OF Tc(IV) TO GEOLOGIC MATERIALS ASSOCIATED WITH A GEOLOGICAL DISPOSAL FACILITY

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Technetium-99 is one of the most important isotopes likely to be disposed of in the proposed UK geological disposal facility (GDF) for higher-activity radioactive wastes. This is due to its long half life (2.13 x 10^5 y), fission yield (6%), and its ability to migrate through soils and other environmental components when in its oxidised, pertechnetate, TcO_4^- form. However, much of the technetium in the GDF is likely to be in the lower oxidation state of Tc(IV) as TcO_2(am) or $TcO(OH)_2$(aq). Therefore, an important aspect of the behaviour of technetium in the near- and far-fields of a GDF is its sorption to geologic and cementitious materials in its reduced (Tc(IV)) oxidation state. BaTch sorption experiments on technetium in both oxidised and reduced forms have been conducted in the presence of various materials which are associated with a deep geological disposal facility. These solids can be placed in the following categories: Single minerals; Sedimentary rocks; Crystalline rocks; Carbonates; Clays and clay minerals; Concretes and cements. Tc(IV) was produced by electrochemically reducing a solution of ^{95m}Tc, ($t_{1/2} = 60$ days) pertechnetate which was used as a spike added to a carrier solution of reduced ^{99}Tc pertechnetate solution. Tc(IV) solutions were used at concentrations from 10^{-9} to 10^{-11} mol dm^{-3}. The results for these studies show R_d values ranging from 0 - 39 000 mL g^{-1} depending on pH and solid material. The results for both oxidised and reduced technetium in the presence of solids associated with a geological disposal facility are presented here. It is noticeable that the R_d is very dependent on the pH. For example, for a 10^{-9} mol.dm^{-3} technetium solution in contact with bentonite at pH 7, $R_d = 21$ mL.g^{-1}, however, at pH 10 for the same concentration of technetium $R_d = 2974$ mL g^{-1}. Surface complexation modelling of the data generated has been performed, as well as characterisation of the solids. In general using bidentate constants give best agreement between model and data, e.g. for bentonite: Bent-OH + H^+ ↔ Bent-OH_2^+ (Log K = 4.5 [1]) Bent-OH ↔ Bent-O^- + H^+ (Log K = -7.9 [1]) Bent-OH + Na^+ ↔ Bent-ONa + H^+ (Log K = -0.1) Bent-OH + TcO_2^+ + H_2O ↔ (Bent-O)$_2$TcO(OH)$^-$ + 3H^+ (Log K = -0.75)

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NFC.L14 (Id: 71)
EFFECT OF Ca^{2+} ON THE SORPTION OF URANIUM(VI) AND HUMIC ACID ON NATURAL CLAY

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Actinide migration in clay formations has been studied by sorption experiments. Also the influence of humic acid (HA) was investigated as it is ubiquitously found in natural environments and can interact with actinides of the nuclear waste. Natural clay contains HA and fulvic acid like organic matter, which can be released from the clay [1]. HA also forms stable colloids. Due to this behavior HA can influence the mobility of actinides, e.g. U(VI), in a nuclear waste repository. Opalinus clay (OPA) from Mont Terri, Switzerland, was studied. BaTch sorption experiments were performed to determine the sorption ability of U(VI) ([U] = 1x10^{-6} M) onto OPA in the absence and presence of HA ([HA] = 10 mg/L, 50 mg/L) and of HA in the absence and presence of U(VI) using synthetic OPA pore water (OPAWA, [2], pH 7.6, I = 0.39 M) and $NaClO_4$ (pH 3-10, I = 0.1 M) as background electrolyte. During these studies a large influence of calcite on the sorption behavior of U(VI) and HA was found. Calcite represents only a fraction of 13% in OPA and its ions occur also in the OPAWA ($[Ca^{2+}] = 0.03$ M, $[CO_3^{2-}] = 5\times10^{-4}$ M). In OPAWA, U(VI) forms with the ions in solution the neutral aquatic complex $Ca_2UO_2(CO_3)_3$ [3]. This complex dominates the speciation with a fraction of 99%. It was shown that this complex adsorbs weakly onto OPA. Normalized to the specific surface area of OPA (BET = 41.6 m^2/g) the U(VI) sorption was determined to be 0.05 ± 0.002 µg U/m^2 clay. It was also shown that HA has no influence on the U(VI) sorption onto OPA. This was proved by speciation calculations, which showed that also in the presence of HA the $Ca_2UO_2(CO_3)_3$(aq) complex is the dominating species in solution. In order to interpret the sorption data onto OPA, additional ζ potential measurements were performed, where between pH 0 and 7.5 a negative ζ potential for OPA was determined. The HA sorption onto OPA in OPAWA was determined to be 3.57±0.01 µg HA/m^2 (10 mg/L) and 17.28±0.15 µg HA/m^2 (50 mg/L). In the presence of U(VI), a slight increase of HA sorption (D = 0.3 µg HA/m^2) was observed for [HA] = 50 mg/L. Species calculations showed that Ca^{2+} ions affect also the HA speciation, because Ca^{2+} is complexed by HA. Ca^{2+} is present in such a high concentration that it saturates the binding sites of HA. Thus, only few binding sites, about 0.1% according to speciation calculations, are available for the complexation of U(VI). Consequently, U(VI) and HA have no effect on each other during the sorption studies. These experiments show the large effect of the calcite fraction of the OPA and thus the resulting composition of the OPAWA on the U(VI) and HA sorption. Thus, calcite should be taken into account for the safety case analysis of a nuclear waste repository.

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NFC.L15 (Id: 103)
SORPTION OF Pd, ONE LONG LIVED FISSION PRODUCT, ONTO SYNTHETIC HYDROXYAPATITE

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The chemistry of several B-metals in the +II oxidation state is of concern for nuclear waste management. Isotopes of ruthenium (Ru), rhodium (Rh) and palladium (Pd) fission products formed in nuclear spent fuels can have an important contribution to the long-term radiotoxicity of high level wastes. The sorption of one long lived fission product, Pd on hydroxyapatite ($Ca_{10}(PO_4)_6(OH)_2$) has been studied at 25 °C as a function of pH, from 0.025 M $Ca(ClO_4)_2$ and 0.025 M NaH_2PO_4 aqueous background electrolytes, trying to minimize some types of reactions, such as dissolution of solid and precipitation of metal. The radiotracer palladium, ^{109}Pd, obtained by the neutron irradiation of $Pd(NO_3)_2$ salt in Triga Mark III research reactor of the ININ (Mexico), has been used to measure their partition coefficients between aqueous phase and hydroxyapatite. In the interpretation of the sorption measurement, we take into account the existence of active sites at the hydroxyapatite surface and the aqueous solution chemistry of palladium as well as the effect of phosphate anions from solid dissolution. The results can be interpreted as evidence of sorption of the species $PdOH^+$, and of a mixed hydroxo complex of Pd^{2+} fixed onto Ca-OH surface sites of the hydroxyapatite.

NFC.L16 (Id: 100)
THE ADSORPTIVE BEHAVIOUR OF CADMIUM ON CLAYS

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Heavy metals present in the environment such as cadmium are considered harmful due to their toxicity. Adsorption-desorption reactions of metals from aqueous solutions to the soil play an important role in influencing the accumulation and transport of metal contaminants. These are affected by the surface and chemical properties of the soil components. Sorption and desorption of Cd^{2+} has been examined on a series of clays bentonite, illite, illite-smectite, kaolin and montmorillonite through γ-spectroscopy as a function of pH and ionic strength. Experimental data acquired for the systems studied have been used to determine the isotherms. The order of increasing cadmium adsorption onto the clays in the absence of organic matter has been found to be:

illite<≈ kaolin<montmorillonite<illite-smectite< bentonite

Further, to gain mechanistic insights to the sorption and desorption processes NMR and XRD have been used to probe these systems.

NFC.L17 (Id: 221)
SPECIATION AND SURFACE COMPLEXATION MODELLING OF Np(V) SORPTION ON MONTMORILLONITE

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The sorption of Np(V) on Na-montmorillonite (STx-1) has been studied by baTch experiments, spectroscopic measurements, and surface complexation modelling with the aim to contribute toward a better understanding of the sorption of ^{237}Np ($t_{1/2} = 2.1 \times 10^6$ a) in the near field (bentonite backfill material) and far field (argillaceous rocks) of high-level nuclear waste repositories. BaTch experiments were performed in the absence of inorganic carbon and under air-equilibrated conditions with 0.1 and 0.01 M $NaClO_4$ as background electrolyte, 8×10^{-12} and 9×10^{-6} M Np(V), and $3 \leq pH \leq 10$. At pH > 8 the presence of inorganic carbon has a strong influence on the sorption behavior of Np(V) due to the formation of aqueous Np(V) complexes with carbonate. Neptunium LIII-edge extended X-ray absorption fine structure (EXAFS) measurements on Np(V)/montmorillonite samples with Np(V) loadings in the range of 0.3-3.5 μmol/g have been performed to determine the speciation of Np at the solid-liquid interface. The EXAFS spectra of samples prepared under ambient air conditions (pCO_2 = 10-3.5 atm) revealed the formation of Np(V)-carbonate complexes at the montmorillonite surface. The results of the baTch experiments obtained under CO_2-free conditions could be modeled using the two site protolysis non-electrostatic surface complexation and cation exchange (2SPNE SC/CE) model described in [1]. For modeling the sorption behavior of Np(V) on montmorillonite in the air-equilibrated system, the aqueous complexation of Np(V) with carbonate [2] was included and the following additional surface complexation reaction was required: $\equiv SOH + NpO_2^+ + CO_3^{2-} \leftrightarrow \equiv SONpO_2CO_3^{2-} + H^+$.

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NFC.L18 (Id: 41)

AN APPROACH FOR ACQUIRING DATA FOR DESCRIPTION OF DIFFUSION IN PERFORMANCE ASSESSMENT OF RADIOACTIVE WASTE REPOSITORIES

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Repositories for radioactive wastes are sited in the environment with very low permeability. One of the most important processes leading to the release of radionuclides to the environment is diffusion and therefore data for its evaluation are very important for the results of performance assessment of these repositories. These data are obtained usually from the evaluation of laboratory through-, in- or out-diffusion experiments, which are evaluated by various mathematical methods that have to take into account also deficiencies of real experimental equipments. E.g., a very long experimental time is needed for sorbing radionuclides to achieve stationary conditions under which the standard evaluation of the through-diffusion experiment is applicable. In such cases only values of apparent diffusion coefficients are usually obtainable from evaluation of in or out-diffusion experiments. A robust performance assessment codes, however, require knowledge of effective diffusion coefficients and equilibrium sorption coefficients, which cannot be easily obtained by the evaluation of in- and/or out-diffusion experiments. This fact can lead to the propagation of uncertainties in performance assessment of radioactive waste repositories. In this paper a new approach is proposed for the evaluation of diffusion data for performance assessment. This approach consist in the following steps: i) experimental measurements of material diffusion parameters (tortuosity, transport porosity) under various conditions (e.g. different density, different ionic strength of water) using non-sorbing radionuclides such as tritium and chlorine for which it is easy to reach conditions under which mathematical solution of diffusion equation is applicable, ii) to measure sorption isotherms for sorbing radionuclides by baTch methodology, iii) to calculate diffusion coefficients for sorbing radionuclides from well-defined diffusion coefficients in free water and determined tortuosity, transport porosity and sorption coefficients, iv) to carry out the relatively short in-diffusion experiments with sorbing radionuclides and v) to compare experimental results with simulated curves using the performance assessment computer code. We see the advantage of this approach in the use of the same computer code both for verification of the laboratory diffusion results and for the performance assessment. The diffusion description uncertainty in performance assessment is therefore decreased. The relationships between measured data and parameters used in performance assessment are discussed on the basis of baTch sorption and diffusion experiments performed for HTO, ^{36}Cl and ^{137}Cs with different types of bentonite.

NFC.L19 (Id: 54)

THE COMPLEXATION OF Tc(IV) WITH ORGANIC LIGANDS IN RADIOACTIVE WASTE

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The preferred option in the UK for the management of higher-activity radioactive wastes is to store it in a deep Geological Disposal Facility (GDF). This may then be backfilled with a cementitious material. Once closed, the GDF will become saturated with groundwater and highly alkaline porewater will develop with an initial pH of ca. 13.4. This will decrease to 12.5 as groundwater flow removes the NaOH and KOH present. Mineral phases in the cement will then act as a buffer and maintain the pH at 12.5 for ca. 105 years. Corrosion of waste-containing steel canisters will lead to the gradual formation of reducing conditions. Thus, the behaviour of radionuclides in the waste must be understood in the context of this chemistry. Organic complexing agents will be present as inherent components of the waste, especially isosaccharinic acid (ISA) and other polyhydroxylated carboxylic acids which will be formed by the degradation of cellulose. These are highly complexing and can cause significant increases in radionuclide solubility at high pH. The GDF will not be homogenous, there are likely to be areas of reducing and oxidising potential. This heterogeneity could mean that both Tc(VII) and Tc(IV) are present within the GDF. If TcO_4^- migrates into an area in which reducing conditions exist, the organics may complex with technetium during reduction to form water-soluble complexes. Also of relevance is the possibility of increased solubility when organics are in contact with reduced technetium (TcO_2(am)). In other words, do organics affect the reduction of Tc(VII) to Tc(IV)? Therefore, studies have been undertaken in which TcO_4^- was reduced electrochemically, and by Sn(II) and Fe(II), in the presence and absence of ISA, gluconic acid, EDTA, NTA and picolinic acid, to determine whether they caused an increase in Tc solubility when TcO_2(am) was contacted with them. In the presence of ISA and gluconic acid a lowering of [Tc(aq)] took place on reduction, showing such ligands did not prevent some reduction occurring. If this reduction was to Tc(IV), then the final aqueous concentration should be the same as that produced by the addition of the same ligands to Tc(IV) solution at steady state, i.e. the Tc(IV) complexes would again be formed, but by 2 different routes. However, the final [Tc](aq) in the system where reduction took place in the presence of ISA and gluconate was higher than when starting from TcO_2(am). This indicates that Tc(VII) may not have been reduced to Tc(IV) but an intermediate oxidation state complex such as Tc(V) may have formed. This concept is well known in ^{99m}Tc radiopharmaceuticals, where polyhydric complexes of Tc(V) can be formed by the reduction of pertechnetate in aqueous solution of excess O-donor ligand, although in pharmaceuticals this is not carried out at high pH.

NFC.L20 (Id: 95)
CHARACTERIZATION OF PORTUGUESE GEOMATERIALS, THE CLAY COMPONENT OF RAÑAS, AS POTENTIAL LINERS FOR LOW AND INTERMEDIATE RADIOACTIVE DISPOSAL SITES

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Safety assessment of low and intermediate level waste repositories requires the understanding of radionuclides sorption-desorption mechanisms, mainly the degree of interaction between radionuclides and mineral surfaces with the aim to analyse processes that can affect the safety of the repository at both near- and far-field zones. Among the different radionuclides that are part of the radwastes' inventory produced by industrial, medical, teaching and research activities carried out in Portugal, ^{137}Cs is one of the most important nuclides from the radiological point of view due to its half-live (30 years) and high radiotoxicity. The clay component of natural geomaterials named rañas, originated from an area located in the NE Mainland of Portugal, were studied for their potential as effective barriers to avoid or reduce the impact of radionuclides migration. Characterization of these geomaterials is the first step towards identification of the adsorption/desorption mechanisms involved. Cationic exchange capacity (CEC), specific adsorption capacity on frayed edge sites (FES), and radiocesium interception potential (RIP) for potassium ion, using γ spectrometry were determined for three selected grains-size fractions (<63 μm, <36 μm and <20 μm). Organic content and pH were also determined. The selected fractions were mineralogically analysed by X-ray diffraction and by Instrumental Neutron Activation Analysis for chemical elemental composition. Smectite, illite and chlorite-smectite mixed-layers were identified in the clay fractions analysed. The overall values obtained for CEC ranged from 184.9 ± 11.7 to 349.5 ± 22.1 mmol kg^{-1}, while FES values were scattered between 0.55 ± 0.06 and 1.55 ± 0.16 mmol kg^{-1}. The RIP values, defined by a clear plateau at higher potassium concentrations, varied from 1.77 ± 0.05 mmol kg^{-1} to 2.90 ± 0.11 mmol kg^{-1}. However, these values are clearly overestimated since they are 3 to 8 times higher than the correspondent exchangeable RIP values. A possible explanation for this deviation could be the fixation of cesium in non-specific sites during the procedure. The ionic capacity of FES is related to the sample granulometry. The values obtained for the FES were shown to decrease for higher grain-size fractions. Also, the presence or absence of an expanded mixed-layer (hydrous) phyllosilicates (i.e., one-or two-layers smectite) in the fractions analysed was found to increase or decrease the radiocesium adsorption. Preliminary results concerning the characterization of these specific geomaterials with regard to the behaviour of radiocesium are very promising in terms of a possible application as lining materials in a hypothetic LILW repository. Further studies involving the sorption, fixation and selective coefficients onto clay minerals related to rãna deposits, are currently under progress.

NFC.L21 (Id: 114)
LONG TERM DIFFUSION EXPERIMENT (LTD) IN GRIMSEL URL: COMPARISON OF MODELING AND IN-SITU RESULTS

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Phase 1 (2005 - 2008) of the long term diffusion experiment (LTD) project has taken place in the Grimsel underground research laboratory (GTS, Switzerland) in a joint effort between NAGRA (Switzerland), University of Bern (Switzerland), NRI Řež (Czech Republic), HYRL (Finland), JAEA (Japan) and AIST (Japan). The project aim was to study matrix diffusion of radionuclides relevant to performance assessments of deep geological repositories (DGR) of nuclear waste, and confirm the role of matrix diffusion for radionuclide retardation within the repository. Work Package 1 of Phase 1 focused on the study of radionuclide diffusion from a single borehole into the undisturbed rock matrix. Within this framework a radionuclide cocktail was injected into a defined borehole interval sealed by packers (8 m depth from Grimsel URL tunnel) in June 2007. The cocktail consisted of a conservative tracer (^{3}H), a weakly sorbing cation (^{22}Na), a non-sorbing anion (^{131}I), and a strongly sorbing cation (^{134}Cs). The injection was preceded by series of preliminary modeling studies. The predictive studies were performed using different codes, namely; a FORTRAN based code, created in NRI, a code based on SW GoldSim with radionuclide migration module integrated (NRI, JAEA), CrunchFlow (UniBern), Nflow (CSCI, Spain). Compiled results predicted ^{3}H migration up to 30 cm depth from the borehole wall, ^{22}Na and ^{131}I migration up to several cms and ^{134}Cs sorption within the first centimeter of the rock. A significant decrease of radionuclide activity level in the circulation water was predicted only for ^{134}Cs. In reality, the radionuclide cocktail was left in contact with the undisturbed rock matrix under real crystalline rock conditions in GTS for 26 months (June 2007 to August 2009). The radionuclide activity level in the circulation water was checked by regular sampling of small aliquots of contact solution and by activity measurements at PSI. As expected, significant activity decrease in in-situ circulation water was determined only for ^{134}Cs. The activity decreased rapidly within 100 days down to the 35% of original level, which was far faster than predicted. Meanwhile short lived ^{131}I ($T_{1/2}$ 8 days) decayed during the first few weeks, ^{3}H and ^{22}Na activity levels decreased down to 80% and 85%, respectively, of the original activity over the 2 year period. Only the results of in-situ reservoir sampling can be compared and re-evaluated with the simulated results. The real extent of radionuclide migration into the rock matrix will be detected after borehole overcoring, followed by core sampling and activity measurements by the end of October 2009. The NRI FORTRAN based code, developed specially for LTD, was used for re-evaluation calculations. This is a 1-D model with cylindric coordinates, that is solved with a finite difference method. Translator G77/GFORTRAN was used for calculations, implementing the results of laboratory and analytical results.

NFC.L22 (Id: 124)
THE EFFECT OF COMPETITION FROM OTHER METALS ON NICKEL COMPLEXATION BY A-ISOSACCHARINIC, GLUCONIC AND PICOLINIC ACIDS

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The UK has an underground repository concept with a combination of engineered and natural barriers. Cementitious materials will produce high pH conditions for at least 1 Ma and surfaces for sorption which will greatly reduce the solubility of many radionuclides. Likely to be in the repository are many organic ligands, as inherent components of wastes or due to the degradation of organics, e.g. cellulose. It has been shown that cellulose degradation products, e.g. α-isosaccharinic acid (ISA), can substantially increase the solubility of radionuclides. Picolinates are in some waste-streams due to the use of picolinic acid in decontamination. Gluconic acid is structurally similar to ISA, a strong ligand useful for comparison purposes and may occur in a repository as a cement additive. From pH 11.5 to 13.5, Ni is fairly insoluble. The 3 ligands increase its solubility by forming aqueous complexes - a process that is fairly well understood. The purpose was to investigate the effects on Ni complexation, of competition from other metals. A repository will also contain Fe metal so experiments were performed in its presence and absence. Predictions of Ni solubility were made for each system to aid the interpretation of competitive effects. Measured [Ni] in the absence of other metals showed good correlation with calculated values. There was evidence for slight sorption of Ni to Th and Eu phases formed. There was no significant effect due to other metals and/or solid phases being present. In general, the calculated [Ni] trend was followed with picolinate. Th and Hf had little effect on Ni solubility in the presence of picolinate, Co reduced [Ni], which might be expected as it complexes in a similar fashion, above pH 13 all metals caused a significant reduction in [Ni]. This may have been due to sorption to solid phases, or coprecipitation, with Ni being scavenged out as the lowest inventory metal. All the Ni was complexed by gluconate. Eu had no significant effect at the highest Ni concentration. Hf, Co and Th reduced [Ni] across the pH range, probably due to complexation with gluconate. Eu had a marginally greater effect at a $[Ni] = 10^{-3}$ M, suggesting a very slight amount of complexation. Hf again had little effect. The Th results were the most difficult to explain. At [Ni] = 0.01 and 0.0001 M, Th affected [Ni] as though forming strong gluconate complexes, but not at 0.001 M. At the lowest [Ni], all competing metals significantly reduced [Ni]. Th had complexed strongly with gluconate, causing Ni to precipitate. Hf had a similar effect to. Co significantly reduced [Ni] in line with its predicted effect. Eu had a significant effect on [Ni] reducing it from the predicted 10^{-4} M to ca. 2×10^{-6} M. Fe was predicted to have no effect on [Ni] by complexation with ISA. The measured [Ni] was lower than the calculated in virtually all cases, including those with no competing metal. This may have been caused by sorption to Fe(s). No significant competition effects from other metals were observed.

NFC.L23 (Id: 249)
MIGRATION CHARACTERISTICS OF ROCK SAMPLES STUDIED BY ELECTROMIGRATION METHOD: METHODOLOGY: PROCEDURE MODIFICATION

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In several concepts of deep geological repository (DGR) development granites are considered as potential host rocks (Sweden, Finland, Czech Republic). Safety calculations, evaluating safety functions of DGR barriers, require information about radionuclide migration within fractured rock formations, where advection and matrix diffusion are considered as the most important processes for activity decrease. Radionuclide diffusion into rock matrix can be studied both in laboratory and in-situ (e.g., Long term diffusion project, Grimsel URL, Switzerland). However, due to diffusion speed the lab experiments usually take longer time than, e.g., baTch experiments, even in year perspective. Moreover, some parameters, e.g., formation factor F_f, are difficult to determine using conventional methods. Hereby, the through electromigration methods (TEM; [1]) can be used for diffusion parameter studies, especially due to speeding up the experimental work in comparison with standard through-diffusion methods. The TEM experiments gain both formation factor (F_f) and effective diffusion coefficient (D_e) values. In NRI the experimental cells for TEM method, based on the work of Löfgren [1], were assembled, modified in order to increase its function and tested. Iodide anion as a tracer in sodium chloride background electrolyte of different concentration was used. Blank experiments with Plexiglas piece, substituting rock sample, were performed for in order to test the experimental apparatus for leakage failures, sample sealing and non-conductive cell materials. Subsequently, TEM experiments with rock samples were accomplished, including samples of crystalline rocks (granite, granodiorite) from the Czech Republic, Sweden and Switzerland. The results (F_f and D_e) obtained were compared and evaluated, taking into account rock sample properties. The research on samples from Sweden (Forsmark and Laksemark) was funded by SKB Sweden that also provided the rock material.

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NFC.L24 (Id: 61)
JOINT DETERMINATION OF ^{99}Tc AND ^{108m}Ag IN L/ILW LIQUID WASTES

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The low- and intermediate-level liquid wastes produced by the Paks Nuclear Power Plant (NPP) contain routinely measureable γ-emitting nuclides (e.g. ^{54}Mn, ^{60}Co, ^{110m}Ag, and ^{137}Cs) as well as many so-called "difficult-to-measure" isotopes. Despite of their low specific activity compared to the total, the reliable determination of these isotopes is an important issue of nuclear waste management. The increasing amount of waste samples to be qualified yearly by our laboratory put a pressure on revising the existing procedure of ^{99}Tc separation applied. We have managed to halve the initial amount of the sample required to achieve the same level of MDA of technetium. Furthermore, one of the new purifying steps introduced have proved to be able to separate more than 99% of ^{108m}Ag (and ^{110m}Ag) keeping the ^{99}Tc content of the product almost intact. As intended, this new procedure has a major impact on the chemical reagent as well as the electricity requirement of the separation making it more cost-effective.

NFC.L25 (Id: 113)
TRITIUM (HTO) AS A CONSERVATIVE TRACER USED FOR CHARACTERIZATION OF CONTAMINANT MIGRATION IN POROUS ROCK ENVIRONMENT

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Chemical leaching of uranium in Northern Bohemia (Stráž pod Ralskem) took place in between 1972 to 1996. Acid technology solutions, mainly consisting of sulphure acid (more than 4 mil tons), nitric acid, ammonia and fluoric acid, were pumped into uranium bearing layers and caused contamination of Cenomanian and Turonian water reservoirs. Cenomanian aquifer containing U was contaminated by more than 5 mil tons of dissolved species, including mainly H_2SO_4, sulfates, Fe, Al, NH_4. However, trace metals are also presented in a broad range of mobile species - e.g., Th, As, V, Mn, Tl, and be both released and migrate within contaminated aquifer and neighbouring layers. Contaminant migration within different layers must be considered during evaluation of measures and methodologies to be used during remediation process. Target rock layers include both porous „friable“ sandstones with permeability coefficient up to 10^{-5} m/s (majority of contamination) and upper „fucoid“ sandstones, containing more impermeable clay and organic fraction with permeability coefficient from 10^{-8} to 10^{-7} m/s. Diffusion transport of potential contaminants is expected to dominate mainly within fucoid sandstone layers. In the presented study ^{3}H (HTO) was used as a conservative tracer in order to study diffusion of non-sorbing, i.e. the most mobile contaminants, within porous environment saturated with groundwater. The through-diffusion technique was used for different samples, including both friable and fucoid sandstones. Breakthrough curves were evaluated using GOLDSIM diffusion module (NRI Řež/CTU), enabling to take into account the unsteady boundary condition in the inlet reservoir. Although the determined values of effective diffusion coefficient D_e fell into relatively narrow interval of ($2.0x10^{-10}$ - $6.34x10^{-10}$) $m^2.s^{-1}$, the dependence of tracer diffusion through sandstone sample on rock properties could be distinguished. It was found that ^{3}H diffusion rate was dependent on pore size distribution, even though the total porosity of different samples did not differ significantly (22-27 % range), and furthermore on mineral content. Kaolinite was finally identified as the main influencing factor for species diffusion rate within sandstone as its content in pores results in different pore size distribution. In samples with higher content of kaolinite (max. 16 %) mainly pores with small size were observed in which tracer diffusion movement was slowed down, and vice versa. Moreover, this phenomenon influences also out-diffusion process of species/contaminants, retained in sandstone samples: faster out-leaching was observed for samples with lower kaolinite content and larger pores.

NFC.L26 (Id: 234)
EXTRACTION OF THORIUM FROM THE FEN DEPOSIT IN NORWAY

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After the renaissance of nuclear power and thorium breeding in particular, the interest for the Norwegian thorium deposits has revived. However, the economic potential for exploitation is depending on several parameters, i.e. heterogeneity of ore, content of carbonaceous minerals, other valuable elements present, eTc. Fen is the site where the largest Norwegian thorium deposits are found. The thorium minerals at Fen are reported to be oxide and silicate, but experience obtained indicates that some thorium must also be present in carbonate rock. Based on experience from extraction of rare earth elements from red rock ("rødberg") at Fen the possibilities for thorium extraction are assessed. The Fen complex in Telemark, Norway is a geological region noted for an unusual suite of igneous rocks. Several varieties of carbonatite are present in the area as well as highly alkaline rocks. The Fen complex is a roughly circular area about three kilometers in diameter. It is located just west of the Oslo graben, on the south side of Lake Norsjø, from Ulefoss in NW some three km eastbound. The area covers some 6 km^2 and has been known for its complex mineralogical composition for almost a century. Rødberg rock in the local area called GruveÅsen is covered with soil and vegetation.

GruveÅsen has the highest concentration of thorium in the area. The thorium concentration in GruveÅsen is reported to reach 0.4%, but ordinarily it is 0.1 - 0.2%. Only in GruveÅsen the amount of Th is expected to be of the order of 5 000 tonnes. The whole Fen area is populated and used as residential and recreational areas. Not only is the heterogeneity large, the mineral grains are small, less than 0.1 mm. The content of carbonaceous rock requires a high consumption of acid to leach it. In a pilot plant operation to extract rare earth elements (REE) HCl was chosen as the dissolving species. 700 kg HCl per tonne rødberg was consumed to leach a yield of 100% Ca and Mg and 80% of REE. Thorium was also leached indicating that Th is not solely present as silicate or oxide. However, small amounts of silicic acid were also present in the leachate causing severe problems. To extract thorium also other valuable elements must be cost carriers and the rare earths are an obvious choice. Another possible element present is niobium, but it is not present in the same minerals as REE. Thorium from Fen will hardly ever be recovered as the main element, but may be a valuable by-product. However, the main obstacle in developing an economic feasible process is the carbonaceous rocks consuming too much acid or energy.

NFC.L27 (Id: 262)

TECHNETIUM CHEMISTRY AT THE UNIVERSITY OF NEVADA LAS VEGAS

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The chemistry of technetium is being explored at the University of Nevada Las Vegas. Our goal is to investigate both the applied and fundamental aspects of technetium chemistry, with a special emphasis on synthesis, separations, and materials science. The synthetic chemistry focuses on metal-metal bonding and binary halides. Recently, two new multiply metal-metal bonded dimers and technetium tribromide and tetrabromide were prepared and characterized. These compounds were used as precursor for synthesis of low valent technetium complexes. The structure of $(n\text{-}Bu_4N)_2Tc_2Br_8$ was solved by single crystal XRD and its electronic structure analyzed by first principles calculations. Separation and materials chemistry is related to the nuclear industry. The separation of uranium/technetium from acidic solution, and synthesis of Tc containing waste forms have been investigated. The facilities at UNLV include modern radiochemistry laboratories where investigators can work with multi-milligram quantities of ^{99}Tc, and analytical instrumentation dedicated for radioelement characterization and analysis.

NFC.L28 (Id: 299)

RECENT DEVELOPMENTS OF NUCLEAR FORENSIC SIGNATURES OF YELLOW CAKES

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Natural uranium is the starting material for the production of nuclear fuels. Uranium of natural isotopic composition is mined from uranium containing ores in different geological formations. The uranium is extracted, chemically purified and pre-concentrated. As an industrial scale material, uranium ore concentrates will carry signatures that provide information on the history and on the origin of the uranium. These signatures may be source material inherited or process inherited. In the present work we investigated the significance and potential application of parameters such as the rare earth elemental patterns or strontium and lead stable isotope ratios. The methodology developed and its application to uranium ore concentrates from different mines around the world will be presented.

Posters

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EVALUATION OF DIHEXYLOCTANAMIDE AS EXTRACTANT UNDER PLUTONIUM RICH FEED CONDITIONS

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Tributyl phosphate (TBP) has been the work horse of Nuclear Fuel Reprocessing Technologists for more than five decades. However, based on five decades of experience on spent fuel reprocessing, certain problems have also been identified with the use of TBP as extractant. Prominent amongst them are: (a) high aqueous solubility, (b) poor radiation stability and interference of degradation products during stripping of Pu/U, (c) poor decontamination factor (DF) values of Pu/U with respect to fission products, (d) low limiting organic concentration (LOC) of Pu(IV), and (e) a large volume of secondary (phosphate) waste. In addition, as a consequence of the radiolytic degradation of TBP, the extraction and stripping behavior of U/Pu, and hydrodynamic properties such as viscosity, density, and phase disengagement time, are adversely affected. These problems are of particular concern to the separation scientists and technologists during the reprocessing of short-cooled thermal reactor fuels as well as of fast reactor fuels. In this context, completely incinerable N,N-dialkyl amides have been evaluated extensively as alternative extractants to TBP. Studies carried out at Radiochemistry Division, BARC, India, on the development of new extractants for the reprocessing of spent fuel suggested that a straight chain N,N-dihexyloctanamide (DHOA) was promising alternative to TBP for the reprocessing of irradiated uranium based fuels. This paper deals with the evaluation of DHOA vis a vis TBP as an extractant for plutonium rich feed solutions encountered in fast reactor spent fuel reprocessing. Solvent extraction studies were carried out to evaluate the two extractants DHOA and TBP using pure 20 g/L Pu as well as simulated Pu rich feed solution containing 20 g/L Pu, 7 g/L U, fission products (FPs) and structural materials (SMs) at 4 M HNO_3. D_{Pu} values were higher for 1.1 M DHOA (26.4±1.2) for pure Pu as well as for simulated feed solution as compared to those for 1.1 M TBP (16.6±2.2) under identical conditions. Under simulated feed conditions, two successive stages were sufficient for quantitative extraction (>99.9 %) of Pu(IV) employing 1.1 M DHOA as extractant and maintaining organic-to-aqueous phase ratio (O/A) as 1. On the other hand, three stages were required in the case of 1.1 M TBP as extractant. Plutonium stripping experiments using 0.5 M HNO_3 as strippant (without any reductant) showed that only six stripping stages were sufficient for quantitative stripping of Pu from loaded DHOA phase. On the other hand, >10 stages were required for Pu stripping from loaded TBP phase and it became further difficult with the aging of the organic phase. By contrast, no problem related to Pu retention was observed for aged DHOA solution. These studies also suggested that DHOA is a promising extractant for coprocessing of U/Pu from spent fuels. In addition, DHOA was found distinctly better than TBP with respect to FPs and SMs decontamination.

NFC.P02 (Id: 283)
COMPREHENSIVE INVESTIGATION OF THE CORROSION STATE AND SURFACE PROPERTIES OF THE STAINLESS STEEL TUBES OF STEAM GENERATORS

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Evaluating the water chemistry in the primary circuit and the effect of chemical decontamination of the heat exchanger tubes performed by the AP-CITROX (AP: alkaline permanganate; CITROX: citric and oxalic acid) procedure at Paks NPP (Hungary), a project dealing with the comprehensive investigation of the general corrosion state of the steam generators (SGs) has been initiated. Owing to the fact that there is no investigation method available for the in-situ monitoring of the inner surfaces of heat exchanger tubes, a research program based on sampling as well as on ex-situ electrochemical (voltammetric) and surface analytical measurements (SEM-EDX, CEMS, XRD, XPS) was developed and elaborated. In the time period of 2000-2008 - within the frame of the above project - 45 stainless steel specimens, cut out from various locations of the steam generators of the Paks NPP were investigated. Based on the measured corrosion characteristics (corrosion rate, thickness and chemical composition of the protective oxide-layer) it was found that these parameters are strongly dependent on the decontamination history of steam generators. The present work gives a brief overview on the general corrosion state of the heat exchanger tubes of SGs, concerning the long-term effects of the AP-CITROX procedure on the chemical composition and structure of the protective oxide-layer.

NFC.P03 (Id: 142)
PYROCHEMICAL AND ELECTROCHEMICAL SEPARATIONS STUDIES ON PLUTONIUM (PART 2)

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Pyrochemical separations, involving molten salt and metal media, by liquid/liquid extraction or electrorefining are studied with nuclear defense and civil applications. The electrochemical properties of plutonium have been studied in molten salt - ternary eutectic mixture NaCl-KCl-$BaCl_2$, equimolar mixture NaCl-KCl and pure $CaCl_2$, and in liquid gallium at 1073 K. These processes concern actinide separations. However, lanthanides, such as cerium, are often used as surrogates. The first steps of a pyrochemical process development consist in the solvent media. Activity coefficients of the solutes in the two phases, that describe the solvent-solute interaction, are important thermochemical parameters to predict separations efficiency and to assess the solvents influence. As nuclear defense scientist, I discuss the advanced developments to separate plutonium by electrochemical method that has been supporting the developments of pyrochemical processes involving plutonium as main goal and actinides separations.

NFC.P04 (Id: 145)
EXTRACTION OF Ln/An FROM HIGHLY ACIDIC SOLUTIONS USING COBALT BIS(DICARBOLLIDE) FUNCTIONALIZED WITH COMPLEXING CMPO GROUP.

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Nuclear fuel reprocessing issues an important problem with respect to the family of actinides representing the main source of radiotoxity during long-term storage. The separation of long-lived radionuclides from liquid radioactive waste enables these hazardous elements to be either conditioned more safely in specific matrices or destroyed by transmutation. Thus, elimination of minor actinides would lead to significant reduction of volume and radiotoxicity of the waste for the final storage and should consequently minimize the possible risks to biosphere. Several extraction concepts were proposed for separation of Ln and An from fission product mixture during the last ten years. In this paper, extraction of trivalent lanthanides and actinides using a compound based on covalent combination of cobalt bis(dicarbollide) (1-) anion (COSAN) and CMPO (N,N- dialkyl carbamoyl methyl diphenyl phosphine oxide) complexing group of the formula [8-$Ph_2P(O)$-$CH_2C(O)N$-t-C_8H_{17}-(CH_2-$CH_2O)_2$-1',2'-$C_2B_9H_{11}$)-3,3'-Co]- will be presented. This compound was selected for detailed tests from a broad panel of other derivatives differing in the substitution at the CMPO function and its bonding to the COSAN cluster [1,2] due to very effective extraction of Ln(III) and An(III) from highly acidic solutions (3M HNO_3), good solubility characteristics and an easy synthetic accessibility in a large scale. Extraction efficiency under different conditions (acidity, reagent concentration, macro amounts of metals, eTc.), solubility in different solvents and chemical stability were tested to evaluate a possible technological use. The composition of extracted complex was studied. Extraction from fission product mixture revealed very effective separation from the majority of fission products with separation factor exceeding 1000. For some fission products, the presence of complexing agents was necessary.

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NFC.P05 (Id: 167)
STUDY OF PROPERTIES OF EXTRACTION-CHROMATOGRAPHIC MATERIAL TBP-PAN

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This project is focused on studying properties of extraction-chromathographic material prepared by embedding tributylphosphate (TBP) into the matrix of polyakrylonitrile (PAN). After elementary characterization of the prepared materials, the kinetics of uranium extraction from 3 mol L^{-1} HNO_3 was examined. The influence of nitrates and nitric acid concentration on the values of weight distribution coeficient D_g as well as "extraction isoterm" was specified. For determination of ^{233}U in aqueous solution, liquid scintillation spectrometry was used. After evaluation of all experiments it can be concluded that TBP-PAN material behaves like TBP in liquid-liquid extraction.

NFC.P06 (Id: 222)
ELECTROCHEMICAL BEHAVIOUR OF SELECTED ACTINIDES AND LANTHANIDES IN MOLTEN FLUORIDE SALTS FLINAK AND FLIBE

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The presented work is focused on research of basic electrochemical properties of several actinides and lanthanides representing the fissile material and fission products in suitable molten fluoride melt. The general framework of this work is to study the electrochemical behaviour of systems relevant for possible future use in the Generation IV reactor concepts. Results of cyclic voltammetry, chronopotentiometric and electrolytic experiments with uranium and several lanthanides in the LiF-NaF-KF (acronym FLINAK) are presented. Mechanisms of electrochemical reduction were investigated. Recorded reduction steps were investigated and described in terms of reversibility, number of exchanged electrons, diffusion coefficients eTc. For the lanthanides, it seems impossible to reach solid deposit of studied lanthanide on the electrode. For uranium, deposition depending on electrode material and shape was studied with special attention given to its deposition on reactive (Ni) working electrode. Beryllium based molten salts $7LiF-BeF_2-ZrF_4$ and $7LiF-BeF_2$ were used as a carrier melts during Molten Salt Reactor Experiment and Molten Salt Breeder Reactor projects [1] and are considered as key systems also for the future use. In the $LiF-BeF_2$ melt (acronym FLIBE), the work was focused on research of uranium behaviour in the melt and the influence of its composition between two eutectic points of FLIBE (eutectic compositions of FLIBE melt are defined by molar ratio of BeF_2 $x = 0.328$ and $x = 0.531$). Also the general consequences of the obtained data for the development of separation process are concluded.

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NFC.P07 (Id: 291)
EPR MEASUREMENTS ON N-BEARING LIGANDS USED IN SPENT NUCLEAR FUEL REPROCESSING FOR An(III)/Ln(III) PARTITIONING

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A future goal for the reprocessing of spent nuclear fuel is the transmutation of long-lived minor actinides, such as americium and curium, into short-lived isotopes by means of neutron irradiation. In order to develop an effective transmutation process, it is necessary to separate the trivalent minor actinides, An(III), from the trivalent lanthanides, Ln(III), by means of two subsequent solvent extraction processes: i) DIAMEX to extract both Ln(III) and An(III) from PUREX raffinate and ii) SANEX to obtain the selective extraction of An(III) from the output of DIAMEX process. The present work concerns the degradation due to ionizing radiation released by radionuclides to the selected solvent system involved in the SANEX process. The high-energy radiations are able to modify the ligands molecules structures changing their extractant capabilities. In particular, several aspects must be considered when selecting a molecule to be used in the mentioned process: chemical stability; solubility in organic diluents; extraction capability of metal ions with appropriate distribution ratios; and resistance towards irradiation. Some aza-heterocyclic extractant molecules C2-BT (5,6 diethyl (1,2,4 bis-triazine), C2-BTP (2,6-diethyl(1,2,4-triazine-3-yl)pyridine), C5-BTBP (6,6'-bis-(5,6-dipentyl-[1,2,4]triazine-3-yl[2,2']bipyridinyl) and CyMe4BTBP (2,6-bis-(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo[1,2,4]triazin-3-yl)-[2,2']bipyridinyl), constituted by pyridine and triazine aromatic rings with chelating nitrogen donor atoms, have been studied during the European Research Project EUROPART. Furthermore, the mentioned molecules present aliphatic side groups as n-carbon alkyl substituents (methyl, ethyl or penthyl chains) or cyclohexyl moieties to ensure their lipophilic behavior after coordination of trivalent metal ions during liquid-liquid extraction processes. To carry out studies concerning radical mechanisms, the abovementioned molecules were irradiated in a ^{60}Co γ irradiation facility (up to 113 kGy with a dose rate of 0.5 kGy/h), at 77 K to reduce radicals reactivity before investigations by EPR spectrometry. A preliminary EPR screening on the four ligand molecules shows the presence of radiation-induced radicals on both the aromatic and aliphatic sides of the ligands. In particular, irradiation of C2-BTP in alcoholic solutions highlighted the formation of azacyclohexadienyl radical as intermediated species. The azacyclohexadienyl radical has been recognized by simulated spectra using appropriate hyperfine constants, and similar signals have been collected also for irradiated solution of C5-BTBP. The EPR spectra collected on samples of C5-BTBP powder, irradiated in air, disclosed the formation of peroxide radical species $ROO^{\bullet}$. Further simulations on the EPR spectrum collected at 208 K revealed definitively the presence of peroxide species on C5-BTBP. In conclusion, taking into account the peroxide radical formation proven on C5-BTBP powder, it was possible to propose several radical mechanisms able to modified the extractant capabilities of the investigated molecules.

NFC.P08 (Id: 342)
REMOVAL OF Cs, Sr, Pu AND Am FROM CONTAMINATED SOLUTIONS BY INORGANIC SORBENTS

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Recently growing concern about contamination of the environment with radioactive and non-radioactive pollutants resulted in intensive studies related to the development of new technologies for separation of radionuclides from liquid waste. These new technologies should be based on highly selective materials (e.g., crystalline titanium silicates) which are hard to decompose over a wide range of pH, which remain stable at high temperatures, which are resistant to ionizing radiation and which are able to operate in the presence of a great excess of competitive ions, organic solvents and oxidants. However, complicated technologies, high capital and regeneration costs stimulated studies to develop low-cost and efficient technologies based on naturally occurring minerals such as zeolites and clay minerals. Another option could be an application of amorphous porous mixed oxides - a rapidly developing class of materials prepared by sol-gel procedures, the main benefit of which are very simple procedures conducted under mild reaction conditions in the ambient atmosphere. Ferrites and a variety of iron-containing minerals such as akaganeite, feroxyhyte, ferrihydrite, goethite, hematite, lepidocrocite, maghemite and magnetite are also a promising class for the treatment of liquid wastes containing radioactive and hazardous metals. The aim of this study was to prepare amorphous TiSi by sol-gel procedures, to synthesize various iron oxides, to characterize them using IR, XRD, Mössbauer spectroscopy and to carry out a comparative assessment of possible application of these low-cost synthetic inorganic sorbents with conventional and natural sorptive materials in liquid waste treatment technologies to remove long-lived radionuclides such as Cs, Sr, Pu and Am. Results obtained using a bath method, ICP-MS, γ, α spectrometry and β counting revealed that titanium silicates, synthesized using $TiOSO_4$ without reference to the chosen method - a precipitation or sol-gel, showed the highest sorption ability towards studied radionuclides. Magnetite and clay minerals showed better sorption ability towards americium. The highest Pu K_d values and better Pu sorption kinetics were found for synthetic iron oxides. An increase in the Pu K_d value by a factor of 6.8 found for magnetite/hematite composite in comparison with the pure magnetite suggests that this sorbent is efficient for plutonium removal and it is promising for its separation from contaminated solutions. TiSi tested in this study showed close sorption ability towards studied radionuclides in comparison with crystalline TiSi, whereas they were synthesized under mild conditions using cheaper materials. In addition, TiSi prepared by the sol-gel method has certain advantages in comparison with the fine powder TiSi because of a huge potential for tailoring of chemical composition, porosity and surface properties, as well as for the production in the granular form, which is especially important for practical purposes.

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NFC.P09 (Id: 34)
IMPROVEMENT OF RADIONUCLIDE FIXATION IN CEMENT MATRIXES DURING IMMOBILIZATION OF LIQUID RADIOACTIVE WASTE

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Cementation provided by mixing of concentrated radioactive waste (RAW) and cement is the most common procedure for immobilization of medium- and low-level radioactive salt-containing waste of radiochemical industry. However, low degree of inclusion of solidified waste in concrete (especially from concentrated salt-containing solutions owing to deterioration of the concrete properties) and noticeable leaching of some fission elements require development of additional barriers at storage of such RAW. Therefore, addition of RAW into the concrete not in the form of solutions but in the form of sorbents saturated with radionuclides can decrease leaching and, in turn, can improve degree of cesium inclusion without decrease in the strength and some other properties of the concrete monolith. Our studies were aimed at treatment of the solutions with complex chemical composition from the units of RAW concentration and fractionation and of the spent decontamination solutions. To include radionuclides containing in liquid RAW in the concrete the possibility of using of super-stoichiometric sorption was studied. This method allows a saturation increase with respect to many radionuclides by a factor of 5-20. Chemical treatment of the sorbent preliminarily saturated with cesium provides stronger fixation of given radionuclide in the solid matrix. Inclusion of sorbents saturated with radionuclides in the concrete increases ^{137}Cs content in the solidified matrix by one-two orders of magnitude without any changes in the strength properties of the resulting composites. At the same time, the leaching rate of ^{137}Cs decreases by a factor of 4-5. Surface treatment of the resulting composites liquid and supercritical CO_2 (so-called carbonization procedure) was also studied to improve fixation of radionuclides in the concrete blocks. Combination of above procedures allows reducing cesium leaching from the concrete blocks by more than two orders of magnitude.

NFC.P10 (Id: 85)
THE SOLUBILITY OF Ni(II) AND Eu(III) IN THE PRESENCE OF CEMENT SUPERPLASTICISERS

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One of the illustrative disposal concepts considered by the NDA-RWMD (Nuclear Decommissioning Authority - Radioactive Waste Management Directorate) for the disposal of intermediate-level wastes and some low-level wastes is that of grouted waste packages surrounded by a cementitious backfill. The potential use of superplasticisers to improve flow properties of waste encapsulation grouts offers benefits in some applications, e.g. for infilling or self-levelling. However, their impact on the post closure performance assessment of a geological disposal facility (GDF) needs to be considered. First generation superplasticisers such as naphthalene formaldehyde and sodium lignosulphonate showed the ability to increase the aqueous concentrations of radionuclides. The observed effects were not consistent and the use of such additives was, in general, discouraged, however, the composition of modern superplasticisers ('comb' polymers) is better controlled than the composition of compounds used in the past. This suggests that their effects may be better understood and they may behave in a more predictable manner. To determine whether the presence of comb superplasticisers will have an effect of the solubility of radionuclides within the near field of a radioactive waste repository, a study of the solubility of Ni(II) and Eu(III) in high pH (12-13.5) aqueous solutions of the superplasticiser ADVA Cast 551 was conducted. The solubility experiments were approached from oversaturation and were conducted in the following aqueous solutions: 95% saturated $Ca(OH)_2$, 0.1 mol dm^{-3} NaOH and cement equilibrated solutions containing Ordinary Portland Cement (OPC), Pulverised Fly Ash (PFA) or Blast Furnace Slag (BFS). ADVA Cast 551 was present at between 0 and 10% (w/v). Samples were taken at regular intervals over the period of 1 month and measured by Liquid Scintillation Counting or Gamma Spectrometry for Ni and Eu quantification respectively. Results for Ni(II) have shown that the concentration of Ni(II) measured in solution increases with the presence of the cement superplasticisers. The increase observed was greatest in the 95% saturated $Ca(OH)_2$ solution. Results observed in the cement equilibrated solutions were less significant with the solubility increase being within an order of magnitude even in the presence of 10% ADVA Cast 551. Results for Eu(III) will also be presented.

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NFC.P11 (Id: 134)
IMMOBILISATION OF CESIUM AND DIVALENT METALS INTO SINGLE-PHASE STUFFED TRIDYMITE-BASED CERAMICS

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The mixed phosphates with high cesium concentration of the compositions $CsMePO_4$ (Me - divalent metal with a tetrahedral coordination) adopting a stuffed β-SiO_2 tridymite structure are studied as perspective materials for ^{137}Cs γ-radiation sources to be used in medical applications. For the purpose of ^{137}Cs source production from commercial wastes (nitrate cesium solutions containing Me impurities) it is necessary to know the possibilities of single-phase composition formation in the systems $CsMe'_{1-x}Me_xPO_4$ with different Me cations. Phase formation regularities and thermal behavior of mixed complex phosphates $CsMg_{1-x}Me_xPO_4$ (Me = Mn, Co, Ni, Zn, Cu) have been studied. The samples were synthesized by precipitation method. The aqueous solution of phosphoric acid taken in accordance with the stoichiometry sample was added to stoichiometric mixture of cesium and metal (Me) nitrate or chloride solutions. The reaction mixture was dried at 353 K and thermally treated at 873 and 973 K. All the thermal treatment stages were alternated with careful grinding. Crystalline powders were obtained. X-ray powder diffraction measurements indicated that continuous (Me = Mn, Co, Ni, Zn; $0 \leq x \leq 1.0$) or limited (Me = Cu; $0 \leq x \leq 0.4$) solid solutions of the tridymite type were obtained. The samples homogeneity and conformity of their compositions to the theoretical values was monitored by scanning electron microscopy and electron microprobe analysis. The phase formation regularities and the dependences of the unit cell parameters of $CsMg_{1-x}Me_xPO_4$ solid solutions on their compositions x were studied. Three polymorphic forms are possible: monoclinic (sp. gr. P21/a) and two orthorhombic (sp. gr. Pn21a and Pnma). For many phosphates phase transitions P21/a → Pn21a → Pnma are observed with temperature increase. Due to the differential scanning calorimetry results, the phosphates $CsMePO_4$ with Me = Co and Zn had two polymorphic transitions in the temperature interval from 423 to 583 K. The phosphate $CsMgPO_4$ underwent phase transition at 236 K. The second harmonic generation results showed phosphate crystallization in centrosymmetric (P21/a or Pnma) or non-centrosymmetric (Pn21a) space groups. The transition from Pnma to Pn21a space group was accompanied by absorption band disappearance of valency symmetric vibrations in IR-spectra of phosphates. A leaching rate of approximately 10^{-5} $g.cm^{-2}.d^{-1}$ for Cs was determined from Soxhlet leaching of $CsMgPO_4$. Thus, the possibility of simultaneous incorporation of cesium and different metals with oxidation degree +2 (present in commercial radiocesium wastes) is shown in the tridymite structure with formation of wide or continuous solid solutions. Such tridymite-like phosphates may be a perspective candidate both for radiocesium immobilisation and cesium isotopic source production instead of soluble CsCl used nowadays.

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NFC.P12 (Id: 138)
PRODUCTION OF ARTIFICIAL METAL RUTHENIUM FROM IRRADIATED TECHNETIUM

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Long-lived nuclide ^{99}Tc is accumulated as a fission product in 1-3 kg/t quantities in fuel of nuclear power plants. Transmutation of technetium by the action of neutrons is the most cardinal method for its neutralization and disposal; this leads to the production of artificial stable ^{100}Ru, ^{101}Ru. Several technetium targets have been irradiated at SSC RIAR. Metal technetium in the form of disks was irradiated in the neutron trap of the SM reactor. Specimens resulting from the irradiation were found to represent Tc-Ru alloys. Prior to our work no description of the production of artificial metal ruthenium from irradiated technetium was available elsewhere. This paper presents two different procedures for the production of artificial metal ruthenium from irradiated technetium targets. In one of these procedures the targets were dissolved in KOH solutions at the presence of KIO_4 followed by precipitation of Ru(IV) hydroxide with ethanol. To purify the produced ruthenium from technetium traces, it was distilled as RuO_4 into the ethanol aqueous solution where it was reduced and precipitated as Ru(IV) hydroxide. To produce metal ruthenium, Ru(IV) hydroxide precipitate was calcinated up to RuO_2 and then reduced to metal in hydrogen flow. According to the other procedure for separation of stable ruthenium from the irradiated technetium target use was made of a catalytic oxidation with ozone. Firstly, a Tc-Ru alloy specimen was placed into HNO_3 or $HClO_4$ solution containing Ag(I) (or Co(II)) ions. Then the ozone-oxygen mixture was bubbled through the solution. Formed by action of ozone Ag(I) (or Co(II)) ions oxidized the alloy (Tc and Ru) components transferring them to the solution. Then RuO_4 and some part of $HTcO_4$ were distilled to NaOH solution where RuO_4 was reduced to sodium ruthenate. At the next process stage the Ru(IV) hydroxide was precipitated from the solution by the action of ethanol acting as a selective reducer of Ru(IV). The precipitate was placed into water and the ozone-oxygen mixture was bubbled through the hydroxide suspension formed in water. The Ru(IV) hydroxide was reduced to RuO_4 with ozone, which was distilled to ethanol aqueous solution by the bubbled gas flow. The produced Ru(IV) hydroxide was transformed to RuO_2, which was reduced to metal in the helium-oxygen mixture.

NFC.P13 (Id: 185)
STUDY OF NEW FILTER PROPERTIES ^{137}Cs VAPOUR CAPTURE AT HIGH TEMPERATURE

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Vitrification of radioactive wastes and high-temperature synthesis of glass and ceramics for ionizing radiation sources, containing ^{137}Cs, is accompanied by discharge of its vapours. The off-gas contaminated by ^{137}Cs must be cleaned up using different trapping systems. In general two methods of ^{137}Cs vapour caTching are possible, differing both in process character (nature) and realization place in technological scheme:

- "wet" method - "low-temperature" ^{137}Cs vapour condensation in system of gas cleaning (condensers, scrubbers, HEPA-filters). This method leads to contamination of communications and to formation secondary liquid RAW, which require additional repossessing.
- "dry" method - "high-temperature" ^{137}Cs vapour chemisorptions. This method allows fixing ^{137}Cs in stable crystalline and amorphous phases. Earlier it was shown that porous inorganic materials with high content of silica- alumina amorphous phase could be used for effective capturing of ^{137}Cs vapours. Effectiveness of filter depends on the total porosity, porous structure and aerodynamic resistance to off-gas flow. To study these parameters a laboratory scale test facility was built at the KRI.

The following three methods are used to evaluate dust and aerosol capture efficiency at room temperature:

- weighing method: sampling of the dust using special certificated analytical filters followed by weighing of the absorbed precipitate. The filters are sealed in the filter holders;
- radiometric method: if an isotope spike is used, the dust is sampled using the analytical filters as described above followed by measurements of absorbed activity using conventional methods;
- aerosol particle concentration measurements: using laser counters for aerosol particle concentrations. This method determines fraction decontamination factors for the particle sizes ranging from 0.2 to 5 μm.

Report will summarize the obtained analytical data and describe the dependence of filter effectiveness at different rates off-gas flow.

NFC.P14 (Id: 230)
RADIOACTIVE WASTE DESTRUCTION USING MOLTEN SALT OXIDATION

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A molten salt oxidation (MSO) process is being studied for the safe and effective destruction of organic components of radioactive waste. The work involves a laboratory-scale molten salt oxidation system where solid or liquid waste is injected into a bed of molten carbonate salt in the presence of an oxidizing gas. The relatively simple MSO process completely destroys organic compounds, and the carbonate salt neutralizes any generated acid gases and retains the radioactivity. In the past, high melting point salts have been used with air oxidation. In these studies, the use of low melting point salts and stronger oxidizing agents are being investigated for the destruction of radioactive waste oil and ion exchange resins. Work on the recovery of uranium from lignites will also be presented.

NFC.P15 (Id: 255)
FUNCTIONALIZED LATEX PARTICLES FOR PREPARATION OF COLLOID-STABLE NANOSIZED SELECTIVE SORBENTS AND COMPOSITE MATERIALS FOR DECONTAMINATION OF RADIOACTIVE WASTES

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Preparation of a new type of colloid-stable nanosized sorbents based on functionalized latex particles is discussed, which can be directly applied for decontamination of liquid and solid radioactive wastes or used for construction of fibrous or mesoporous composite sorption materials. Carboxylic latex (siloxane-acrylate and polystyrene-acrylate) functionalization is performed via immobilization of nanocrystals of sorbents selective to radionuclides (ferrocyanides of transition metals and sulfates of calcium/barium) into the polymer matrix. The presence of anionic centers on the latex particle surfaces stipulates for the possibility of binding to them ions of divalent metal-precursors of selective sorbents. With the added metal concentration increase one can observe the decrease of electrokinetic potential (by module) at the constant particle size. The residual negative charge on the surface provides the system with high colloid stability. To form stable nanoparticles of a selective sorbent it is crucial to limit the amount of introduced divalent metal ions by the beginning of the plateau on the dependence of electrokinetic potential on the metal concentration in solution. AFM imaging of the functionalized latex particles shows that, as a result of introducing cobalt ferrocyanide nanoparticles, the initially spherical polystyrene-acrylate (PA) particles transform into a cubic shape, which is characteristic for ferrocyanide macrocrystals. Due to immobilization into stable polymer colloids, the selective sorbents nanoparticles preserve high stability at filtration in porous media. After 50 filtration cycles of emulsion of the siloxane-acrylate (SA) latex with immobilized nanoparticles of cobalt ferrocyanide through a layer of natural zeolite, the cobalt content in the emulsion remained nearly constant. When such colloid-stable sorbents are directly applied for decontamination, after radionuclides sorption the stability of such systems can be controllably reduced by addition of cationic flocculants, thus providing high efficiency of radionuclide removal from solution. The combination of high selectivity of nanosized sorbents immobilized into latex particles with ease of their removal from solution by flocculation/coagulation process enables us to recommend the developed schemes for different materials decontamination, first of all, for solid bulky wastes - soils, grounds, metal constructions. To extend suggested approach to the production of different types of sorption materials pre-formed colloidal-stable selective sorbents were deposited on carbon fibers by electrochemical method or used as a template during formation of mesoporous SiO_2, ZrO_2 and TiO_2 sorbents. The selective sorption materials obtained showed good kinetics of radionuclides sorption and distribution coefficients for cesium up to $1x10^7$.

NFC.P16 (Id: 284)
COMPARATIVE STUDY OF THE CORROSION AND SURFACE ANALYTICAL EFFECTS OF THE DECONTAMINATION TECHNOLOGIES

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Decontamination technologies are generally developed to reduce the collective dose of the maintenance and operation personnel at NPPs. The highest efficiency (i.e., the highest DF values) available without detrimental modification of the treated surface of structural material is the most important goal in the course of the application of a decontamination technology. The AP-CITROX procedure has mainly been used for the decontamination of the primary coolant circuit's components (e.g. main circulating pump (MCP) and steam generators (SGs)) at the Paks NPP. While decontamination of the dismountable devices (e.g. main circulating pump) has been carried out in big tanks with heating potential, separable equipments (e.g. heat exchangers) have been treated with special decontamination facilities. The AP-CITROX procedure was used 24 times for the SGs decontamination at units 1-3 of the

Paks NPP during the period of 1993-2001. Our previous studies have revealed that a "hybrid" structure of the amorphous and crystalline phases is formed in the outermost surface region of the austenitic stainless steel tubes of SGs as an undesired consequence of the industrial application of the AP-CITROX decontamination technology. In this presentation, we report some comparative findings on the corrosion and surface analytical effects of the AP-CITROX procedure and the decontamination technology elaborated at our institution. After optimizing its operational parameters the latter technology is suitable for the decontamination of both dismountable (e.g. MCP swivel) and separable (e.g. heat exchanger) equipments. In this semi-plant scale experiments, the passivity, morphology and chemical compositions of the treated surfaces of tube specimens were investigated by voltammetry, and SEM-EDX methods. The newly developed decontamination technology can be effectively applied for the decontamination of the austenitic stainless steel samples; the total oxide-layer can be removed by this technique. The SEM-EDX results revealed that the oxide removal is surprisingly uniform even after 2 or 3 consecutive cycles. The electrochemical studies provided evidences that no unfavorable tendencies in the general corrosion state of the tube samples can be detected in the course of the chemical treatments.

NFC.P17 (Id: 285)
OBSERVATION OF EFFICIENCY OF THE DECONTAMINATION TECHNOLOGIES IN NUCLEAR POWER PLANTS

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The version of the AP-CITROX technology (pre-oxidation: alkaline potassium permanganate, oxide-solution: blend of oxalic and citric acid) applied in the steam generators of the Paks NPP (Nuclear Power Plant) was not adequately developed considering its chemical, analytic and corrosion aspects. To replace the AP-CITROX procedure, a new, efficient „soft" chemical decontamination technology has been developed at the Institute of Radiochemistry and Radioecology at University of Pannonia. In the course of the comprehensive characterization of the efficiency of the novel base-technology the chemical composition and morphology of the oxide layer formed on the inner side of the austenitic stainless steel heat exchanger tube specimens before and after the full decontamination process were studied by scanning electron microscopy (SEM), equipped with an energy dispersive X-ray microanalyzer (EDX). The complete decontamination cycle has been performed under laboratory conditions in a pilot plant circulation system elaborated earlier. In different steps of the chemical procedure the concentration of the main alloying components (Fe, Cr, Ni) dissolved from the surface oxide layer into the decontamination solutions was determined by ICP optical emission spectrometric (ICP-OES) method. Based upon the ICP-OES results the average thickness of the oxide layer removed from the surface into the solutions was calculated. When measuring the activity of the removed radionuclides (^{60}Co, ^{58}Co, ^{110}Ag, ^{54}Mn) in the decontamination solutions we drew conclusions concerning the efficiency of certain steps of the technology and depth distribution of the radionuclides on the treated steel surfaces.

NFC.P18 (Id: 49)
STUDY OF CRYSTALLINE CERAMICS FOR IMMOBILIZATION OF ^{99}Tc

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Technetium-99 is β-active long-lived artificial radionuclide (half-life more than 200,000 years). Because of its long-life, high content in spent nuclear fuel, high ability to oxidize under aerobic conditions as TCO_4^- Tc is one of the most dangerous radionuclides. Development of chemically durable host-phase for Tc disposal is important goal of research. There are no natural minerals of Tc, which might be considered as analogues of stable Tc host-phases. Some chemical elements such as manganese, titanium, rhenium can be used as Tc-simulants. They do not reflect however chemical behavior of Tc in detail. It was suggested to consider development of Tc host-phases with structures of spinel; pyrochlore; fluorite; garnet in three oxide systems: Fe-(Mn,Tc)-O, Zr-(Mn,Tc)-O, and (Sn,Tc)-O using different procedures of precursor preparation and ceramic synthesis conditions. Most samples were synthesized at 1150 °C in inert atmosphere from precursors doped with 5-12 wt. % Tc. Some samples were obtained in air. All the samples obtained were studied using optical and scanning electron microscopy (SEM); powder X-ray diffraction (XRD); microprobe analysis (EMPA) and static leach test in distilled water at 90 °C for 28 days. Content of Tc varied from 0.5-0.8 to 3-6 wt.% in oxide host phases and from 54 to 93 wt.% in metallic inclusions. Leach rates were less than 10-3 g/m^2. Development of optimal methods of precursor preparation and synthesis conditions of Tc-doped ceramic are discussed.

NFC.P19 (Id: 6)
THE SORPTION OF CESIUM ON BEISHAN SOIL UNDER DIFFERENT PHYSICOCHEMICAL CONDITIONS STUDIED BY BATCH AND EDS TECHNIQUES

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Adsorption of Cs^+ to Beishan soil (BS) as function of pH, foreign ions, temperatures, contact time and humic substances was studied in detail under ambient conditions using baTch techniques. The results suggested that the adsorption of Cs^+ was strongly dependent on ionic strength, whereas nearly independent of pH values, and the values of K_d were 1388.78 mL/g in 0.01 mol/L $NaClO_4$ and 740.14 mL/g in 0.1 mol/L $NaClO_4$ solution, respectively. The foreign ions competed with Cs^+ in the sequence: $K^+ > Na^+ > Li^+$ and $Mg^{2+}>Ca^{2+}·Na^+$; however, the influence of anions was not obvious which might be attributed to the very low complexing ability with anions (i.e., Cl^-, ClO_4^- and NO_3^-). The adsorption reaction of Cs^+ to BS was very fast; the values of E_a were 8.944 kJ/mol. The positive ΔH_0 and negative ΔG_0 meant that the holistic process of Cs^+ adsorption to BS was an endothermic and spontaneous process. The adsorption isotherms of Cs^+ were well simulated by the Langmuir model at higher concentration of $NaClO_4$, whereas Freundlich model was better than Langmuir model at low ion strengths (i.e., 0.001 mol/L $NaClO_4$); and the values of q_{max} obtained from the Langmuir model ranged from $\sim 3.33\times10^{-7}$ to $\sim 5.00\times10^{-7}$ mol/g. Humic substances (FA and HA) enhanced significantly the adsorption of Cs^+ to BS.The EDS analysis indicated that the adsorbed Cs^+ were mainly distributed to the fray edges of BS, the locations of these adsorbed Cs^+ coincided with the sodium depletion area, implying the replacement of Na^+ by Cs^+ adsorption.

NFC.P20 (Id: 52)
SELENIUM(IV) RETENTION ONTO ILLITE

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In the context of nuclear waste management, long-term safety assessments have shown that selenium-79, released from the solid waste matrix, could be one of the major isotopes contributing to the global radioactivity potentially reaching the biosphere. Selenium has a quite complex speciation, with four main oxidation states, depending on both the pH and the redox potential of the surrounding environment. Sorption onto minerals can strongly affect the availability and the mobility of selenium. It is thus of great importance to be able to characterize at both macroscopic and microscopic levels the different processes (retention, reduction, surface precipitation, …) that can potentially take place. Regarding the multi-barrier concept considered for deep underground storage of high level and long-lived radionuclides, clays are candidates as host rock as well as backfill materials. Clays like illite, kaolinite, montmorillonite and bentonite constitute the main phases of clay rocks, together with other compounds like iron oxides, titanium oxide, pyrite, calcite and organic matter. We decided to focus our work on illite since it is an abundant and well crystallized clay mineral in soils. Thus, retention properties of illite towards selenium(IV) have been investigated in this study, using a combination of both macroscopic and microscopic measurements. Illite du Puy (France) has been used as the sorbing phase. It has been purified in order to remove auxiliary and minor phases to get a homo-ionic “Na-illite” clay. Then, baTch experiments aiming at studying the sorption behaviour of selenium(IV) onto illite have been performed in $NaClO_4$. The influence of the suspension pH, as well as the ionic strength effect has been investigated. All the experiments have been performed under anoxic conditions in a glove box under N_2 atmosphere ($O_2 < 1$ ppm). Selenium speciation in solution has been checked using Hydride Generation-Atomic Absorption Spectroscopy measurements. The oxidation state of the selenium species once sorbed onto the illite surface has been evidenced using X-Ray Photoelectron Spectroscopy. Electrophoresis measurements have also been performed during this work. Comparison between the ζ potential of the illite surfaces before and after selenium(IV) sorption has been done, to check whether the sorption takes place by chemical bonding formation or rather by electrostatic attraction. Finally, ATR-FTIR measurements have been performed using an ATR ZnSe crystal. By comparison with former IR measurements concerning selenito-ligands containing complexes as well as phases with sorbed selenium species, the fashion binding of selenium(IV) onto illite has been evidenced.

NFC.P21 (Id: 76)
SORPTION OF RADIONUCLIDES TO THE CEMENTITIOUS MATERIAL NRVB UNDER NEAR-FIELD CONDITIONS

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The current concept for the disposal of intermediate- and low-level waste in the United Kingdom involves the emplacement of the grouted waste confined in stainless steel canisters in a cementitious repository deep underground [1]. The NRVB (Nirex reference vault backfill) is a cementitious material specially formulated for this purpose, composed by a mixture of ordinary Portland cement (OPC), hydrate lime ($Ca(OH)_2$) and limestone flour [2]. The movement of the radionuclides away from the repository is expected to be retarded by their limited solubility under the chemical conditions present in the repository (high pH, due to the cement, and low Eh, due to corrosion of the waste canisters) as well as, by sorption processes. In the present work, the sorption of a series of radionuclides (I^-, Cs^+, Ni^{2+}, Eu^{3+}, Th^{4+} and U^{6+}) to NRVB was assessed under near-field conditions (highly-alkaline pH and anoxic conditions).

The experiments were carried using NRVB-equilibrated water (pH ~ 12.6) in NaCl 0.1 mol dm^{-3} as liquid phase, under a N_2 atmosphere (O_2 and CO_2 free), according to the procedure proposed by Sutton et al. [3]. Linear, Langmuir and Freundlich isotherms were studied for all the radionuclides, and in all cases, the sorption of radionuclides fitted well with the linear model. The R_d (C_{solid}/C_{liquid}) observed experimentally varied in a wide interval that ranged from the low values of 0.012 and 0.033 dm^3 g^{-1} for Ni^{2+} and I^-, respectively, to values as high as 2870 dm^3 g^{-1}, obtained for Cs^+. Results of the effect of cellulose degradation product on sorption to NRVB will also be presented. When the R_d (C_{solid}/C_{liquid}) values of the radionuclides of interest for NRVB, were compared with the experimental R_d values for the individual components of NRVB, i.e. OPC, hydrated lime and limestone, it was observed that in the case of nickel and in absence of the organic ligands, R_d(NRVB) · R_d(OPC) + R_d($Ca(OH)_2$) + R_d(limestone), confirming the validity of the additive model for this material. Acknowledgements Authors express their thanks to the Radioactive Waste Management Directorate of the Nuclear Decommissioning Authority for sponsoring this project

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NFC.P22 (Id: 111)
SORPTION PROPERTIES AND BEHAVIOUR OF ^{137}Cs AND ^{90}Sr ON BENTONITE CLAYS AND MAGNOX SLUDGES

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Within the nuclear industry, waste management and disposal issues, both within the plants themselves and in the surrounding areas, are complex and problematic. This is partially a result of many decades of discharges within the storage site. A major problem is in determining the nature and distribution of contamination within the environment, be this on site in surrounding host rock, or determining whether migration has occurred to such an extent that contamination has breached the boundaries of the site and reached the public domain, or whether contamination has been released to atmosphere, for example. It has been suggested that there may be significant quantities of contaminated land arising from breaches in radionuclide storage. Therefore, an understanding of the transportation of radionuclide waste in the environment around storage ponds and silos on waste sites, as well as an understanding of the behaviour within the ponds, is essential in future waste remediation strategies. One area of significant interest is the migration of ^{137}Cs and ^{90}Sr radionuclides in contaminated clay soils, as migration has been shown to be significantly faster than expected in some areas. Although exact figures are not known, information provided by a UK operator suggests that ^{137}Cs and ^{90}Sr are progressing through the surrounding area quicker than the expected annual rate of 6 cm/year, for this type of material. Sorption studies carried out using ^{137}Cs and ^{85}Sr have returned promising results. ^{85}Sr was used in place of ^{90}Sr as it is easier to work with, yet still exhibits the same behaviour in terms of sorption. Although some previous studies have looked at the sorption properties of these two radionuclides within the two environments studied most have focussed on only one or two variables. In these studies conducted at Loughborough University, numerous variables including pH, ionic strength and the effect of competing ions being present in the system have been investigated, while all the time keeping the remaining contributing variables (all of those not being specifically investigated, plus contact time, solid-to-liquid ratio, eTc) constant. This has provided data which will be extremely valuable as any changes in behaviour can be directly associated with the altered variable and results can be directly compared with those previously obtained. The rheology of clays and clay minerals plays an important role in the transport of these key radionuclides. A key area of the repository LTP (Life Time Plan), for research and development is to be able to link the transport in the materials studied to rheology and organic content. Specific clays of interest are bentonite and montmorillonite, as well as the mineral brucite. A final goal of being able to inhibit transport, either by chemical speciation or barrier systems is an aim of this project.

NFC.P23 (Id: 121)
STUDY OF CESIUM AND STRONTIUM SORPTION ON ROKLE BENTONITE IN DIFFERENT ELECTROLYTES

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In most of deep geological repository (DGR) concepts sorption of radionuclides on bentonite represents an important process of retarding radionuclide migration into geosphere. In the Czech Republic, DGR concept takes local bentonite into account as material for both buffer and backfill. The candidate bentonite comes from the Rokle deposit (NW Bohemia) and represents complex mixture of (Ca,Mg)-Fe-rich montmorillonite, micas, kaolinite and other mineral admixtures (mainly Ca, Mg, Fe carbonates, feldspars and iron oxides). This bentonite differs in composition and properties from worldwide studied Na-bentonite (e.g. MX-80, Volclay) or Na-Ca bentonite (e.g. Febex). This fact leads to the need of investigation of Rokle bentonite in greater detail to verify its suitability as a buffer and backfill in DGR. Despite a number of studies concerning cesium sorption on bentonites, there are still some issues requiring clarification, especially the effect of cesium-selective minerals (micas and mica-type clay minerals). Their amount in bentonite and their effect on cesium sorption at different conditions represents great uncertainty in cesium sorption evaluation. In this study, the general trends of cesium and strontium sorption on Rokle bentonite (different samples) were compared with sorption on the well-defined reference material (Ca-montmorillonite SAz-1). Then, the effect of changing solution ionic composition on both Cs and Sr sorption was investigated in detail for average sample of Rokle bentonite and associated uncertainties in distribution coefficient (K_d) determination were evaluated. The comparison with reference sample demonstrates that cesium-selective minerals are very important for cesium sorption even at its trace concentrations. For strontium there are no selective minerals to sorb it and the sorption is influenced mainly by the cation exchange capacity (CEC) of the bentonite. Comparing different electrolytes for cesium, the potassium has the highest competitive effect (competition on selective sites), in contrast to sodium and calcium. For strontium, calcium has the highest competitive effect (competition on regular ion exchange sites), potassium has a lower one and sodium the least one. No significant differences were found for three selected samples of Rokle bentonite in sorption behavior for cesium and strontium, except the effect of total CEC of bentonite. The main uncertainty sources for cesium arise from the presence of cesium-selective minerals in Rokle bentonite samples in varying amounts. For strontium, the amount of smectite (which has the highest CEC in natural bentonite) represents the main source of uncertainties. It also follows from the performed experiments that the cesium sorption on studied bentonite cannot be described using simple K_d value.

NFC.P24 (Id: 224)
SORPTION BEHAVIOUR AND SPECIATION OF NEPTUNIUM(V) ON OPALINUS CLAY

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The sorption of Np on clay such as Opalinus clay (OPA) is one of many important processes that affect the migration and retardation of Np in the geosphere. Clay formations are under consideration in several European countries as a potential host rock for high-level radioactive waste repositories [1]. We studied the sorption of Np(V) onto OPA (from Mont Terri, Switzerland) both by baTch experiments and X-ray Absorption Spectroscopy (XAS). The aim of this study was to obtain a mechanistic understanding of the interaction of Np(V) with the OPA surface. The obtained information is necessary for the safety assessment and detailed planning of nuclear repositories. The baTch experiments were done under aerobic and anaerobic conditions. The sorption of Np(V) on OPA was studied as a function of many chemical parameters such as pH (6-10), background electrolyte (synth. OPA pore water, saturated calcite solution, and 0.1 M $NaClO_4$), partial pressure of CO_2, and presence/absence of humic acid (HA). The sorption curves for $8x10^{-6}$ mol/L Np(V) in saturated calcite solution obtained under aerobic and anaerobic conditions, respectively, show that the adsorption edge occurs between pH 7-8. Maximum sorption was observed at pH 8.5 with 65% sorption under aerobic conditions and 80% under anaerobic conditions, respectively. Under anaerobic conditions the reduction of Np(V) to Np(IV) by Fe(II) minerals of OPA causes a stronger sorption. At pH > 9 Np sorption decreased due to the complexation of Np with carbonate in aqueous solution [2]. Molecular-level information was obtained by X-ray Absorption Fine Structure (EXAFS). EXAFS spectra were measured in fluorescence mode at the Rossendorf Beamline ROBL at ESRF. Several samples at pH 7.6 and 8.5 with a total Np(V) concentration of ~ $8x10^{-6}$ mol/L were prepared under aerobic and anaerobic conditions. The amount of Np sorbed in the EXAFS samples was in the range of 50-121 ppm. Np L3-edge X-ray Absorption Near-Edge Spectra (XANES) showed that in all aerobic samples the pentavalent oxidation state of Np was the dominating one. The Np L3-edge EXAFS spectra of the anaerobic samples prepared in different background electrolytes showed the same EXAFS pattern, indicating that Np(V)-carbonato complexes are formed at OPA surface. More details as well as additional results will be presented and discussed. The authors acknowledge the ESRF for provision of synchrotron beam time and thank the ROBL team for assistance during EXAFS measurements.

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NFC.P25 (Id: 301)
SORPTION AND DIFFUSION CHANGES OF ^{134}Cs, ^{99}Tc AND ^{129}I RADIONUCLIDES ON BENTONITES AT VARIOUS CONDITIONS

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Radionuclides of ^{99}Tc and ^{129}I belong among the fission products in spent nuclear fuel and make the largest contributions to long-term nuclear waste. The high proportion of risk from these radionuclides is due to their large inventories in many types of waste, long half-lives, and the perception that they are highly mobile in the environment and therefore play potentially large role in long-term dose assessment. Computer-modeling methods were used to calculate equilibrium thermodynamic principles, the distributions of predominant aqueous species, and potential solubility controls for the environmentally important oxidation states of each of the contaminants of concern. The Eh-pH diagrams of individual chemical species of the tested radionuclides were calculated by the geochemical software tool Geochemist's Workbench. The data obtained from the model calculations corresponded with experimental results. The chemical stability of bentonite was studied on a series of long-term pressure and temperature loaded bentonite samples that were selected with the aim of constructing a behavior profile of the bentonite buffer material in the experimental container. The chemical stability of the loaded bentonite samples was evaluated on the basis of their ion exchange capacity, the migration behavior of selected radionuclides and determination of the leachable components of bentonite into redistilled water. The results obtained in this study were compared with the values for the unloaded bentonite material. It was found that long-term pressure and temperature gradients do not have a significant influence on the changes of the ion exchange capacity of bentonite material. These results make a major contribution towards determining the migration of radionuclides on bentonite, especially cationic forms of radionuclides. Electromigration methods and thin layer chromatography with radiometric detection were used for the identification of technetium and iodine chemical forms in the studied solid-liquid systems. Migration studies of the radionuclides ^{134}Cs in the form of the Cs^+ cation and ^{99}Tc in the form of the pertechnetate anion were described on the basis of two dominant processes that both include sorption and diffusion. The dominant chemical form of technetium under these redox conditions is the insoluble $TcO_2.nH_2O$, which is formed by the reduction of pertechnetate anion to the oxidation state Tc(IV). The determined values of distribution and diffusion coefficients of the loaded bentonite samples corresponded with the values of the unloaded bentonite material. Ion exchange at the surface sites of bentonite corresponded with the control mechanism of Cs^+ cation sorption on bentonite in that both were simultaneously influenced by competitive cations present in the aqueous phase(Na^+, K^+, Ca^{2+}, Mg^{2+}). To retard the migration of selected radionuclides ($^{99}TcO_4^-$, $^{125}I^-$) released from spent nuclear fuel after the failure of a container, the reducing effects on the concentration of these radionuclides by container corrosion products and of some additives on the bases of Fe compounds in various oxidation states were examined in aqueous media in contact with bentonite.

NFC.P26 (Id: 33)
EVOLUTION OF THE REDOX POTENTIAL IN THE CORROSION SYSTEM

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The knowledge of chemical conditions, especially of redox potential, inside waste packages with radioactive wastes is important for the determination of radionuclides speciation and their leaching rate after failure of waste packages. Conditions inside waste packages will be determined primarily by the reactions of thermodynamically unstable iron with groundwater penetrating into waste packages and by the nature of the iron corrosion products formed. The main corrosion products of iron corrosion are Fe(II) ions, hydrogen and products of reactions of these species with species present in water flowing into waste packages. The composition of water will correspond to the composition of ground water affected by other engineered barriers. The rate of the corrosion reaction, the rate of oxidation and hydrolysis of Fe^{2+} and the rate of the formation of precipitation products will govern the development of Eh inside waste packages. One possibility of estimating chemical conditions in such complex systems is to use advanced geochemical models. A great disadvantage of these models is that a lot of important input data must be estimated. This can lead to great uncertainty about the real conditions inside waste packages. The main aim of this work was therefore to measure redox potential (Eh) in simplified systems simulating the conditions inside waste packages and the effect of iron corrosion on Eh development inside waste packages after ingressions of groundwater. The results can then be used to validate geochemical models. The corrosion systems consisted of the carbon steel plates and the synthetic bentonite porewater. The apparatus was put into anaerobic box ($O_2 < 0.1$ ppm), where redox potential was measured by platinum and gold electrodes. The corrosion rate of carbon steel was determined during experiment by measuring hydrogen evolution and then from weight loss of samples. The results show that the values of the redox potential during the experiments firstly sharply decreases and then slowly increases. Opposite behaviour was observed at measuring pH, where values of pH first slightly increased and then slowly decreased. The concentration of Fe^{3+} ions after the experiments was negligible in comparison with Fe^{2+} concentration. The corrosion rate at 50 °C was almost constant during corrosion experiment, while the corrosion rate at 60 and 70 °C was very fast at the beginning of corrosion but then significantly slowed down. It seems that the effect of temperature on corrosion rate is affected by the nature of corrosion products formed on the surface of metal. The experiments provide useful information about evolution of the redox potential and other parameters which can be expected inside waste packages with radioactive wastes, but time of the experiments carried out so far was relatively short (30 days). In the future, long-term experiments must be performed under various conditions to verify the results obtained.

NFC.P27 (Id: 40)
STUDY OF GAS GENERATION IN REAL L/ILW CONTAINERS

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To obtain reliable estimates of the quantities and rates of the gas production in L/ILW a series of measurements was carried in the last seven years in Hungary. Drums filled with selected original L/ILW coming form the light water PWR type Paks Nuclear Power Plant (Hungary) were placed into special hermetic containers and the gas generation in them was measured carefully. Headspace gas analysis of real radioactive waste vaults closed between 1979 and 1995 in a near surface L/ILW disposal facility of Hungary was also carried out. It was clearly indicated that the gas generation rate is relatively high in the L/ILW drums independently of the chemical type of the main components of the stored waste. Our results showed that the main generated gases in L/ILW are carbon dioxide, methane and hydrogen. The typical rates were 0.05-0.2 STP litre gas/day for CO_2 and CH_4 generation, and less than 0.02 STP litre gas/day for H_2. No explosive gas mixture was indicated in the L/ILW drums during the investigated storage period. Compositions of headspace gases in closed L/ILW vaults were in good agreement with gas generation processes observed in L/ILW drums. The stable carbon isotope measurements show that the main source of the CO_2 gas is the degradation of organic matter in the waste. The low ^{13}C content indicates microbial degradation processes as the main sources of CH_4 in the headspace gas. Typical tritium activity concentrations were between 0.1 and 10 Bq/liter gas in the drums and between 10 and 1000 Bq/liter gas in the vaults. Typical ^{14}C activity values of the headspace gases were between 0.1 and 2.0 Bq/liter gas in the drums and 10 and 1000 Bq/liter gas in the studied vaults.

NFC.P28 (Id: 86)

QUALIFICATION OF LOW AND INTERMEDIATE LEVEL RADIOACTIVE WASTES (L/ILW) WITHIN THE FRAMEWORK OF THE “DEMO” PROJECT BY A TRIATHLER TYPE PORTABLE LIQUID SCINTILLATION SPECTROMETER IN PÜSPÖKSZILÁGY, HUNGARY

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In the Püspökszilágy Radioactive Waste Treatment and Disposal Facility, Hungary, (in operation since 1976) many works started to solve the storage of the non-power-plant radioactive waste of Hungary to fulfil the increasing safety demands. Within the framework of this procedure the exhumation of 66 reinforced concrete near-surface vaults with the size of 70-140 m^3 each (called vault "A") started together with further renewal works. The exhumation of the A11, A12, A13 and A14 vaults was included in the "Demo" project, in the course of which the aim was to find the proper measurement and waste-backfilling technique to adapt them as further vaults. To identify the ^{3}H, ^{14}C and ^{90}Sr isotopes a Triathler type portable liquid scintillation spectrometer was used together with a rapid and well-reproducible swipe sampling method. The expected total activity value was obtained by evaluating 4000 samples. Certain waste packages were removed to gain space allowing the disposal facility to accept further waste in the future. The measurement technique is therefore well-applicable in the case of the "A type" vaults.

NFC.P29 (Id: 87)

„DEMO” PROGRAM IN PÜSPÖKSZILÁGY, HUNGARY: QUALIFICATION OF LOW AND INTERMEDIATE LEVEL RADIOACTIVE WASTES BY A FIELD γ SPECTROSCOPY SYSTEM

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The Radioactive Waste Treatment and Disposal Facility, Püspökszilágy, Hungary, is in operation since 1976. Low and intermediate level radioactive wastes originating from different industrial, medical, educational and scientific activities are disposed of here. At the beginning of its operation the facility had to fulfil only safety requirements but not regulations pertaining to the form and the quality of the wastes. The recent safety analyses aim to improve these temporary solutions to ensure the further safe and proper operation of the facility. A „Demo” project was launched to remove the wastes containing long-lived and highly active isotopes and to gain excess capacity with more organised backfilling, within the framework of which the exhumation of 4 near-surface reinforced concrete vaults (called „A type”) was fulfilled. In the vaults the wastes were in plastic packages. Most of them were undamaged during hoisting, thus the identification of the various isotopes was possible with the help of a field γ spectrometry system. A Genie 2000 Gamma Analysis Software was used together with a Big Mac type CANBERRA γ spectrometry system with a high-purity germanium detector. On the basis of the original records most of the wastes containing different isotopes were identified and the results of the measurements were similar to the ones estimated from the records. After the backfilling it can be seen that the purposes regarding the excess capacity of the vaults can also be achieved. It can be stated that the measurement technique for the further similar vault exhumation procedures is proper and applicable.

NFC.P30 (Id: 294)
IMPACT OF HYDROGEN GENERATED BY IRON CORROSION ON COMPACTED BENTONITE IN DEEP GEOLOGICAL REPOSITORY

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A large number of processes will influence the performance of deep geological repository of radioactive wastes until the activity of radionuclides decay to a negligible level. One of the very important processes is generation of gases, and particularly of hydrogen, which will be formed primarily by anaerobic corrosion of metals and by radiolysis of water. High pressure formed in the vicinity of waste packages due to accumulation of hydrogen can cause failure of sealing materials and contribute to fast release of radionuclides to the geosphere after waste package failure. This contribution presents the results of laboratory experiments, which simulated the phase of repository evolution after ingression of water in failed canister with spent fuel assemblies, which is connected with significant generation and accumulation of hydrogen in free voids of waste packages and at interfaces of waste packages and compacted bentonite used as a sealing material. Corrosion of carbon steel canisters inside walls is simulated by corrosion of iron powder with high surface. It was found that after an increase of pressure of hydrogen to the values exceeding some threshold values, which depends on density bentonite, hydrogen is released to geosphere in pulses due to formation of preferential paths. This is connected with a significant increase of permeability of bentonite from values of approximately 10^{-24} m^2 to 10^{-18} m^2 within the breakthrough time. The pressure needed to reach breakthrough is decreased with the number of breakthrough pulses. Data obtained in experiments enable us to understand more closely the processes occurring in a repository and to avoid conditions that could lead to the failure of sealing materials and fast release of radionuclides to the geosphere and the environment.

NFC.P31 (Id: 39)
DISSOLVED GAS MEASUREMENTS OF THE COOLING PONDS OF PAKS NUCLEAR POWER PLANT, HUNGARY

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The aim of this work was the investigation of the effect of nuclear fuel rods on the composition of the dissolved gas in the cooling water of the cooling ponds of Paks Nuclear Power Plant (Paks NPP). In normal cases after three-year usage in the reactor the spent fuel elements are stored in cooling ponds for years before delivering out from the reactor area. In April 2003 due to the failure of the cleaning procedure of fuel rods from reactor No. 2 some elements remained in service pool No. 1. For better handling of this situation and planning the repairing procedure several parameters must be considered. The gases dissolved in the coolant, especially those produced by fission or decay, are good indicators of the variation in state parameters of the system in this situation as well as in the case of working reactors. Parallel with the measurements of dissolved gases in the coolant for surveying the kilter of the nuclear fuel remained in service pool No.1 we also measured these parameters in the cooling ponds as a reference. The reason was the similar technological situation because in both cases the coolant contains out-of-work fuel elements and the headspace gas above is air with atmospheric pressure. In this work we investigated the effect of the nuclear fuel rods on the composition of the dissolved gas in the coolant of the cooling ponds and the service pool No.1 of the reactor No. 2 of Paks NPP. We measured the quality and the quantity of the dissolved gases and the isotope compositions of the noble gases produced in fission or decay. For these aims sampling and measuring methods were developed. Our results help to ascertain the possible ways and rates of gas generation processes caused by the presence of nuclear fuel rods in the cooling water.

NFC.P32 (Id: 74)
USING NATURAL ORGANIC MATTER AS A REMEDIATION MATERIAL IN ENVIRONMENTAL APPLICATIONS

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Natural organic matter (NOM) is being characterised and investigated for use as a remediation material in various applications. Two readily available environmental materials are being tested: Material A: 68% by weight of the material is organic including 19% humic acid, 28% fulvic acid and 21% humin. 32% of the material is inorganic, containing 23% iron. Material B: 57% by weight of the material is inorganic, 3% is humic acid, 27% is fulvic acid and 13% is humin. The humin fraction of these two materials may contain humic acid that has been immobilised by clays or other minerals. The materials have been tested in three applications; i) As a permeable reactive barrier A permeable reactive barrier is a passive, in situ method that is used to remediate contaminated groundwater. As groundwater flows through it, the barrier extracts contaminants, significantly reducing their concentration. Both materials are capable of extracting metals and organic pollutants from groundwater ii) Extraction of iodine from urine ^{131}I may be given to patients with thyroid problems for diagnosis or treatment. About 30% of the activity (2-3 GBq) is excreted in urine during the first day after administration. In some parts of Europe, the radioactive urine is collected in tanks at the hospital. The objective of this investigation is to determine whether the radioiodine can be extracted from urine and concentrated in a smaller volume of solid. iii) Extraction of iron and other contaminants from industrial effluents During the production of kaolinite or China Clay, brown discoloration caused by Fe^{3+}-containing impurities is removed by the addition of acid and a reducing agent, forming soluble Fe^{2+}. The latter is then removed from the effluent by precipitation with NaOH and flocculants. This study investigates the use of solid NOM to bind iron and co-contaminants directly, avoiding the need for alkali addition and precipitation of large quantities of slurry.

Acknowledgement. Anumaija Leskinen thanks Loughborough University and Enterpris Ltd for financial support

3. NUCLEAR ANALYTICAL METHODS (NAM)

Lectures

NAM.L01 (Id: 201)

SIMULTANEOUS SPECIATION ANALYSIS OF ARSENIC, ANTIMONY AND SELENIUM IN NATURAL WATERS BY NEUTRON ACTIVATION COUPLED TO HPLC, SOLVENT EXTRACTION AND SOLID-PHASE EXTRACTION

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Simultaneous speciation neutron activation analysis (SSNAA) technique is being developed in our laboratory over the last 15 years. This technique can now be used for the simultaneous determination of not only various species of a single element but also species of other elements present in the same sample. Almost all speciation techniques consist of two steps. The first step involves the separation of species from the sample followed by the second step of element-specific detection. Neutron activation analysis (NAA) methods in combination with high-performance liquid chromatography (HPLC) were developed first for the determination of low levels of five arsenic species, namely As(III), As(V), monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), and arsenobetaine (AsB) in water samples. Organically bound arsenic (OBAs) and total arsenic levels were measured. The detection limits of the HPLC-NAA method were found to be 0.005 ng mL^{-1} for OBAs, 0.02 ng mL^{-1} for AsB, DMA, MMA, As(III), and As(V) and 0.12 ng mL^{-1} for total arsenic using the Dalhousie University SLOWPOKE-2 reactor facility. These methods were then extended to include the determination of arsenic, antimony and selenium species, namely As(III), As(V), AsB, OBAs, MMA, DMA, Sb(III), Sb(V), and Se(IV), in natural water samples by solvent extraction and solid-phase extraction. The accuracy of the methods was evaluated by analyzing a Riverine Water CRM. Details of all three methods will be presented along with the concentrations of various species measured.

NAM.L02 (Id: 211)

NAA FOR LIFE SCIENCES AT FLNP JINR: PRESENT AND FUTURE

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Within the broad spectrum of activities in the life sciences at JINR such as nuclear medicine and pharmacy, radiation biology, radioecology, radioisotope production – radioanalytical investigations play a special role due to the long-term experience in multi-element instrumental neutron activation analysis (INAA) at the reactor IBR-2 of FLNP, JINR. Activation with epithermal neutrons along with conventional and cyclic INAA is presently being used in numerous projects on air pollution studies with biomonitors (moss, lichens, tree bark). The results on moss surveys (heavy metal atmospheric deposition study) for some selected areas of Central Russia, South Urals, and countries of Europe (Bulgaria, Croatia, Poland, Romania, Bosnia, Serbia, Macedonia, Slovakia, Western Ukraine, Thace Region of Turkey) have been reported since 1995 to the European Atlas of Heavy Metal Atmospheric Deposition edited under the auspices of the United Nations Economic Commission for Europe. Similar investigations were initiated also in Mongolia, Vietnam, China and South Korea with analytical opportunities offered in Dubna. Applied to the analysis of air filters, INAA is successfully used in assessing quality of London underground air, air of Bratislava, and the Sahara desert impact on the Greater Cairo Area. Epithermal activation analysis in combination with atomic absorption spectrometry and energy-disperse X-ray fluorescence allowed source evaluation of metals in soil from some industrial areas of Russia (South Urals, Kola Peninsula) and Macedonia (Veles province). The analytical possibilities of NAA are favorably used in biotechnology: (i) in the development of new pharmaceuticals based on the blue-green alga Spirulina platensis; (ii) for transformation of toxic forms of elements into non-toxic ones by Arthrobacter oxidans; and (iii) for investigation of bacterial leaching of metals, including uranium and thorium, from low-grade ores, rocks and industrial wastes. Occupational health studies at some fertilizer plants in Russia, Uzbekistan, Poland, and Romania, the quality of foodstuffs grown in some contaminated areas of Russia, Romania and South Africa were investigated in the framework of the IAEA Co-ordinated Research Programmes, the EU 5th Frame Programme and a project with the Nuclear Energy Corporation of South Africa (NECSA). The results of applying NAA to the problem of decommissioning of Nuclear Power Plants and utilization of industrial wastes are demonstrated. The prospects of the above investigations along with the new trends and projects (role of trace elements in the development of atherosclerosis, health impacts of nanoparticles, etc.) to be carried out at the upgraded reactor IBR-2M are reviewed. The educational aspect of using the radioanalytical facility REGATA at the IBR-2M reactor for training young specialists from JINR Member- and non-Member-States is touched upon.

NAM.L03 (Id: 190)

USE OF NEUTRON ACTIVATION ANALYSIS FOR THE CHARACTERIZATION OF SINGLE-WALL CARBON NANOTUBE MATERIALS

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Carbon nanotubes (CNT) are one of the first major nanoscale manufactured products to enter the market. Therefore, reliable and reproducible quantitative measurement and characterization of carbon nanotube samples are important for progress in understanding of these materials and the development of new applications incorporating these materials. An additional value is the development of an information base for CNT toxicology. In response to this need, the National Institute of Standards and Technology (NIST) is developing a single-wall carbon nanotube (SWCNT) Standard Reference Material (SRM). In the course of this work, instrumental neutron activation analysis (INAA) was used to evaluate a variety of SWCNT materials for selection as a candidate reference material as well as to establish elemental composition data on SWCNT materials. INAA proved to be well-suited for the direct determination of catalyst and contaminant trace elements requiring only minimal sample preparation. Prompt γ neutron activation analysis (PGAA) complemented the INAA data in particular with determinations of the light elements. Carbon and hydrogen results provided information on the materials purity and storage properties. INAA and PGAA data were used in the value assignment of mass fractions of catalyst and trace elements in the candidate SRM.

NAM.L04 (Id: 318)

DETERMINATION OF SILICON IN BEER AND BEER PROCESSING MATERIALS BY FAST NEUTRON ACTIVATION ANALYSIS. IS BEER A POSSIBLE PROTECTIVE FACTOR IN PREVENTING ALZHEIMER´S DISEASE?

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Silicon is an essential element for organisms. Although the biochemical mechanisms remain obscure, dietary Si is considered important for growth and development of bone and connective tissues. Recently, it has been suggested that Si and silicic acid may decrease the bioavailability of aluminium by blocking its uptake through the gastrointestinal tract and by impeding reabsorption in kidney [1]. Aluminium, a neurotoxin, has lately been implicated as one of the possible casual factors contributing to Alzheimer´s disease, since the element is conducive to oxidative stress in the brain, and oxidative damage is the primary risk factor for Alzheimer´s disease and other neurodegenerative disorders. While many foods contain Si in polymeric form or as hydrated silica – opal (phytoliths), bioavailable Si in beer is present in a monomeric form as a soluble orthosilicic acid H_4SiO_4 [2,3]. It is the Si form which may decrease the bioavailability of Al and consequently may afford protection against Al toxicity in brain. For this reason, we studied the Si content in various Czech beers and beer base materials using fast instrumental neutron activation analysis. Lyophilized samples of several Czech beers and beer processing materials from one brewery (barley, sweet and hopped wort, extracted grain, yeast, hop extract and granulate, nonfiltered and filtered beer) were combusted in a nickel crucible at 600°C in air. Water used for hot mashing of barley was preconcentrated by evaporation. Irradiation behind a Cd shield was used for determination of Si via the short-lived radionuclide ^{29}Al resulting from the ^{29}Si(n,p)^{29}Al reaction with fast neutrons in the ash from the above samples, and/or in the water aliquots. γ-ray spectra of the irradiated samples and standards were measured by a coaxial HPGe detector coupled to a Canberra Genie 2000 system. For quality control, reference material NIST SRM 2704 Bufallo River Sediment was assayed. The study has shown how Si comes into beer from the individual input materials. In addition to Si contained in water, barley and hop are the main sources of Si in beer. It is mainly the hot mashing technique, which extracts orthosilic acid from barley's and hop's phytolitic silica. The Si concentration in the Czech beers analyzed were in the range of 14 to 44 mg/L, while water used for hot mashing of barley in the studied brewery contained 7 mg/L of Si. Dietary Si intake in the USA is about 20-50 mg per person per day [2], while the British diet has been reported to contain 9-23 mg of Si per person per day, with 60 % derived from cereals with poor bioavailability [4]. Thus it is obvious that moderate beer consumption significantly increases intake of bioavailable Si. Therefore, if the rather controversial hypothesis about aluminium´s neurotoxicity holds, beer drinking should be considered a protective factor in preventing Alzheimer´s disease and other Al induced neurodegenerative disorders.

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NAM.L05 (Id: 170)
TOTAL AND BIOACCESSIBLE FRACTIONS OF TRACE ELEMENTS IN CULTIVATED OYSTER TISSUES BY INAA, PIXE AND ICP-MS

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There is an increasing interest in estimating the total and bioaccessible fraction of elements of both nutritional and toxicological importance in cultivated oysters consumed by Japanese population groups. Oysters are cultivated in Japan by hanging them on an 11-m long rope in the ocean. Levels of 15 elements in oysters cultivated at different depths were investigated by INAA in the present study. Three groups of oyster were collected at 1, 6, and 11 m depths of a single rope. From each group, five oysters were chosen and removed from the shell after washing with tap water. Two groups of organ, namely (i) hepatopancreas and muscle, and (ii) gill and mantle, were separated from soft tissues. These organs were freeze-dried and pulverized. The organs of group (i) are thought to accumulate elements from the ingestion of planktons from sea water while those of group (ii) are simply in direct contact with sea water. One portion of powdered sample was irradiated at the Dalhousie University SLOWPOKE-2 reactor facility in Halifax, Canada for assaying Ag, Br, Cl, Cu, Mg, Na, Se, and V by instrumental neutron activation analysis (INAA) through their short- and medium-lived nuclides. Another portion was irradiated at the Kyoto University Reactor in Osaka, Japan for Co, Cr, Fe, Rb, Sb, Sc, and Zn by INAA using mostly long-lived nuclides. A third portion of the sample was digested in a microwave oven after adding indium solution (for use as an internal standard) and concentrated nitric acid for analysis of more than 20 elements by particle induced X-ray emission (PIXE). Almost all elements except Cd, Cl, and Ni were found to accumulate in soft tissues with increasing depth. The bioaccessible fraction of the elements was estimated by an in vitro enzymolysis method using α-amylase, protease and amylglucosidase followed by ICP-MS. Details of all experiments and results will be presented.

NAM.L06 (Id: 181)
NEUTRON AND PHOTON ACTIVATION AND ION BEAM TECHNIQUES IN GEOCHEMICAL CHARACTERIZATION OF MOLDAVITES AND OTHER IMPACT GLASSES

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The study presents results of geochemical characterization of a large collection of tektites and other impact glasses. Impact glasses are produced through large meteoritic impacts by melting of surface materials. Tektites are impact glasses ejected from the impact site to distant strewn fields. The collection included namely moldavites from the major parts of the Central European tektite strewn field, and irghizites - impact glasses from the Zhamanshin crater in Kazakhstan. Several samples of Australasian tektites and Libyan Desert glass were available as well. The characterization has been based on determination of about fifty elements using various modes of instrumental neutron activation analysis, supplemented by instrumental photon activation analysis, prompt γ activation analysis, PIGE and PIXE. Geochemical data are presented and discussed in view of parent materials and processes involved in formation of various tektites and impact glasses.

NAM.L07 (Id: 193)
DEVELOPMENT OF A RADIOCHEMICAL NEUTRON ACTIVATION ANALYSIS PROCEDURE FOR DETERMINATION OF ARSENIC IN BIOLOGICAL MATERIALS

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Neutron activation analysis (NAA) has played a key role in the certification of As in biological reference materials at NIST. Instrumental neutron activation analysis (INAA) with counting of ^{76}As is typically used to determine ≥1 mg/kg As in biological materials, though lower amounts may be determined, depending on the matrix. At lower levels, As determination is often hindered by the presence of significant amounts of ^{24}Na, ^{82}Br, or ^{32}P. Radiochemical neutron activation analysis (RNAA) with retention of As on hydrated manganese dioxide (HMD) has been used in the certification of As mass fractions in biological reference materials at levels <1 mg/kg [1,2]. Although this method provides very high and reproducible yields, and detection limits at low µg/kg levels, counting geometry uncertainties may arise from uneven distribution of As in the resins. Additionally, the method is not specific to arsenic since other elements (Ag, Cr, Mo, Nb, Sn, Sb, Se, Ta, W) are retained on the column, resulting in possible decreased detection limits for As. Finally, the method does not appear to be easily applicable to all matrices. Two methods are being investigated to yield minimized uncertainties associated with counting geometry, increased specificity for arsenic, and broader applicability. The first involves the use of an ^{77}As tracer to monitor yields and correct for differences in counting geometry. The second method involves the use of a liquid extraction procedure in which ^{76}As is counted in the liquid phase, thus minimizing uncertainties associated with counting geometry. The latter method promises to be specific to arsenic and applicable to a wider range of matrices.

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NAM.L08 (Id: 225)

NAA FOR STUDYING DETOXIFICATION OF Cr AND Hg BY ARTHROBACTER GLOBIFORMIS

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Bacterial reduction and detoxification of potentially toxic metals is one of most promising strategies for the bioremediation of contaminated environmental media. In our previous studies we have established that the Gram-positive bacterial strain Arthrobacter globiformis isolated from basalts taken from the ecologically most polluted region of Georgia (Marneuli) can reduce and detoxify Cr(VI) with high efficiency [1]. In the present investigation instrumental neutron activation analysis (INAA) was applied to study (1) accumulation of Cr(VI) in A. globiformis in the presence of Hg(II); (2) accumulation of Hg(II) in bacterial cells; and (3) effects of Hg (II) and mixture of Cr(VI) - Hg(II) on the elemental composition of bacteria. Our experiments were focused on the dose-dependent effects of Cr(VI) and Hg(II). Cr(VI) as [K_2CrO_4] and Hg(II) as [$Hg(NO_3)_2.H_2O$] were added to the bacterial cell cultures at an early stationary phase of their growth. Two sets of experiments were performed. In the first set the concentration of Hg(II) varied within the range of 50-5000 μg/L. In the second set a 500 μg/L concentration of Hg(II) was added to the bacterial cells at each given concentration of Cr(VI) within the range of 50-1000 mg/L. According to the results obtained, the dose-dependent character of Cr(VI) accumulation by the tested bacterial strain was not significantly affected by the presence of Hg(II). Accumulation of Hg(II), similar to the Cr(VI) accumulation, follows well the Langmuir-Freundlich model. Besides, NAA measurements showed increased content of Fe in bacteria under Hg and Cr action, suggesting that Fe-containing biomolecules play a decisive role in detoxifying heavy metals by A. globiformis. The concentration of 5000 μg/L of Hg(II) was found to be critical for A. globiformis. At this concentration of Hg(II) the concentrations of both essential (Na, Mg, Al, Cl, K, Mn, Zn) and some non-esential elements (Rb, Sb, Sc, As) changed drastically along with a decrease of the biomass of bacteria by a factor of 2. One may assume that under this high exposure to Hg(II) the structure of the bacterial cell wall was destroyed.

Acknowledgement: We are grateful to STCU for their support (Grant #4330)

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NAM.L09 (Id: 368)

NEUTRON ACTIVATION ANALYSIS FOR ASSESSMENT OF BEEF ORIGIN

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Consumer confidence in beef has been hampered by epidemics such as bovine spongiform encephalopathy (BSE) and foot and mouth disease (FMD). Legislation has increased restrictions on beef trade worldwide. For example, the beef trade in the European Union requires traceability throughout the supply chain, allowing the consumers to be aware of the origin of meat available in the market. Nevertheless, the traceability is usually based on labeling and information technology, rather than on intrinsic characteristics of the meat. Analytical methods determining essential and non-essential chemical elements are potential tools to establish the geographical origin of meat, since certain regions may provide specific fingerprints, especially for trace elements. Here, instrumental neutron activation analysis (INAA) was used to assess the origin of beef produced in 3 different countries. For this evaluation, 85 samples of top sirloin were acquired from the local market of Piracicaba, including meat from North, Southeast and South regions of Brazil, in the states of Pará, Minas Gerais and Rio Grande do Sul, as well as from Argentina and Australia. The meat samples were minced in a knife mill, freeze-dried and ground in a ball mill. Analytical portions of approximately 300 mg were weighed into high purity polyethylene vials and irradiated in a thermal neutron flux of 10^{13} cm^{-2} s^{-1}, followed by four measurements of induced radioactivity by HPGe detectors. For quality control, reference material NIST RM 8414 Bovine Muscle Powder and IAEA A11 Trace Elements in Milk Powder were analyzed. The k_0-method was used to provide results for seven chemical elements, i.e. Br, Cs, Fe, K, Na, Rb and Zn. Four elements (Fe, K, Na and Zn) presented low variability among the meat samples analyzed, despite the different origins. Br, Cs and Rb showed a greater variability and a good potential to be used as indicators of origin. However, further studies with a more comprehensive sampling are needed to fully demonstrate this assumption. Moreover, additional chemical elements should be investigated to improve the applicability and the confidence of the proposed methodology for assessment of beef origin.

NAM.L10 (Id: 210)

ELEMENTAL CONCENTRATIONS IN VEGETABLE SPECIES FROM INDUSTRIAL ZONES IN ROMANIA DETERMINED BY INAA

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The environmental pollution by industrial activities presents a significant health risk to human beings. One of the possible pathways of pollutants from the environment to human organisms is the consumption of foodstuffs polluted by industrial activities. This paper presents the levels of elemental concentrations determined by Instrumental Neutron Activation Analysis (INAA) in various vegetable species cultivated in industrial zones of Romania and control sites with no specific type of pollution. The following industries were considered: phosphorous fertilizer production at Turnu Magurele, nonferrous metallurgy at Copsa Mica, iron and steel metallurgy at Targoviste. INAA was applied at IFIN-HH in Magurele (near Bucharest) for the long-lived radioisotopes (TRIGA nuclear reactor in Pitesti) and JINR Dubna for the short-lived radioisotopes (IBR-2 reactor in Dubna). The elements determined were: Al, Ag, As, Au, Ba, Br, Ca, Cd, Ce, Cl, Co, Cr, Cs, Cu, Eu, Fe, Hf, K, La, Mn, Mo, Na, Ni, Rb, Sb, Sc, Se, Sm, Sr, Ta, Tb, Zr, and Zn. Elemental concentration results are compared with Romanian standards and values from the literature. Elemental bioavailability of the investigated plant species and their contamination with trace elements due do industrial activities are discussed. This research was supported by the PN 09370105 and PNCDI II 72-172/01.10.2008 Projects of the Education, Research and Youth Ministry in Romania and Joint Research Project No. 3871-4-08/10 between JINR Dubna, Russia and IFIN-HH, Romania.

NAM.L11 (Id: 131)

SELECTED TRACE ELEMENTS IN POTTERY OF GUARANI ETHNIC GROUP

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Artifacts of pottery belonging to the Guarani ethnic group were investigated by XRF techniques. The Tupi-Guarani is one of the three main ethnics groups representative of the neolithic culture in the Amazonian scope. Such an ethnic group dispersed towards the South; in the Paraguayan area between Paraguay and Parana Rivers several Guarani ethnic movements by both rivers and their tributaries are perceived. The lithologics and ceramics typology have contributed to support that perception. The archaeological findings help to clarify prehistoric cultural aspects and dispersal areas. In that context, the knowledge of the chemical composition of the found ceramic devices, in particular of the REE and other refractory ones provide to information on this dispersion and its expansion. Selected trace elements analysed in samples from eleven devices were Rb, Ba, Nb, La, Ce, Sr, Nd, Sm, Zr and Y using an Am source. Their spidergrams have allowed to identify four different sets of specimens according their areas of provenance.

NAM.L12 (Id: 123)

COMMENTS ON THE CORRECT INTERPRETATION OF K0-FACTORS AND ON THE PROPER USE OF THE SMELS MATERIALS

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Recently plans were devised [IAEA CRP 2005-2009: “Reference Database for Neutron Activation Analysis”] for creating and publishing a library of k_0-factors that are generated differently from the experimentally determined ones published and used in practice thus far [ADNDT, 85 (2003) 47]. For two reasons, we look at this anxiously. Firstly, when realizing that these “new” k_0-factors will be based on values taken from (be it internationally evaluated and up-to-date) files dealing with data for decay (notably γ ray emission probabilities) and activation (notably 2200 m s^{-1} activation cross sections), it is clear that trackability and accuracy are likely to be jeopardized. Indeed, an evaluated cross section is (partially or fully) based on values that are experimentally measured via the “activation method”, thus requiring the introduction of γ ray emission probabilities that are selected by the experimentalist. This implies that the link is broken when they are again combined with recently evaluated γ-ray probabilities - whereby it is irrelevant whether the latter are in se more accurate than the ones introduced formerly. This means that the way of creating these “new” k_0-factors is basically incorrect. Secondly, the release of the thus obtained k_0 library, next to the one for actual use in the practice of k_0-NAA (managed formerly by the INW/Gent - KFKI/Budapest, and in the future by the recently established k_0 Nuclear Data Committee), would inevitably bring about confusion and ambiguity. Also in recent years, we observed from published papers and from communications at conferences that the SMELS materials (Synthetic Multi-Element Standards) are not properly used for what they were developed for [NIMA, 564 (2006) 675]. Indeed, it is seen that the SMELS are systematically applied for the validation of k_0-NAA carried out in a laboratory, whereas their real potential lays in the quality control of the implementation of the k_0-standardization, notably with respect to the calibration of the irradiation facility and of the Ge-detector, including also the correction for dead-time in case of short-time NAA. Indeed, the three SMELS-types are doped with elements that give rise to (n,γ) activation with epithermal-to-thermal cross section ratios and emitted γ-ray energies ranging from low to high (thus giving information on the correctness of respectively the neutron flux characterization and the detection efficiency determination), and (for SMELS-type I) with half-lives ranging from low to high (thus giving information on the correctness of dead-time correction). Regrettably, these built-in potentials of the SMELS are overlooked in nearly all cases described in the literature.

NAM.L13 (Id: 215)
THE k_0-BASED NAA AT BARC: DEVELOPMENTS, QA/QC AND APPLICATIONS

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The k_0-based Neutron Activation Analysis (k_0-NAA) was adapted in Bhabha Atomic Research Centre (BARC) in the year 1994 and since then it is being used in our R&D activities. The present article gives an account of developments and applications of k_0-based NAA as well as internal monostandard NAA (IM-NAA) using research reactors at BARC. Irradiation sites were characterized by cadmium ratio methods by determining sub-cadmium-to-epithermal neutron flux ratio (f) and epithermal neutron flux shape factor (α). The k_0-factors for a large number of short, medium and long-lived nuclides with respect to gold (^{197}Au) were determined. Several (certified/standard) reference materials obtained from IAEA and NIST were analyzed for evaluating accuracy of the method. Recently, synthetic multielement standards (SMELS I, II and III) were analyzed as part of QA/QC of IM-NAA. The detection efficiency, which is an important input parameter in k_0-method, was measured using multi γ-ray standard sources in k_0-NAA studies and in the case of IM-NAA , insitu relative detection efficiency was obtained using γ-rays from the sample itself. The later approach could help us in analyzing samples of non-standard shapes and sizes. Most of our work, except for short-lived nuclides, was carried out by irradiating samples in the swimming pool type Apsara reactor. The measurement of γ-rays of activation products was carried out using a 40 % relative efficiency HPGe-detector coupled to MCA and peak areas were determined using the peak-fit software PHAST. Developed k_0-methods were applied for composition analysis of samples in the field of geology, biology, environmental science, archaeology, material science and nuclear technology. Important contributions were made by determining composition of alloys like zircaloys and stainless steels and impurities in 1S-aluminium without using a standard. We have been contributing towards the development of large sample NAA (LSNAA), under an IAEA CRP, using low flux and highly thermalized irradiation positions at research reactors. Large samples of pottery, coal, uranium ore, rocks and stainless steels were analyzed using thermal column irradiation facility at Apsara reactor. We are also investigating the possibility of using LSNAA method in the graphite reflector position of the critical facility (CF) at BARC and the standardization of the methodology is in progress. Some details and salient results will be presented in this paper.

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NAM.L14 (Id: 157)
NATIVE AND TRANSPLANTED LICHEN AND BARK AS AIR POLLUTION BIOMONITORS AT THREE DIFFERENT METEOROLOGICAL CONDITIONS

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At three sites of Portugal - Viana do Castelo, Sacavém, Sines - bark from Olea europea and the lichen Parmelia caperata, collected in clean areas, were exposed. The situation of these sites represents three different meteorological conditions, from rainy to dry and cooler to warmer. Three different procedures for the lichen exposure were adopted: rectangular flat pieces, the lichen in its original form and biomass (previously ground lichen). Samples were exposed for 9 months and removed after 3, 6 and 9 months (continuous mode) and exposed for 3 months, in a winter campaign, a spring campaign and a summer campaign (discontinuous mode). Conductivity was measured before and after exposure whenever possible. Ten samples of each biomonitor were kept for analysis prior to exposure. All samples were analyzed by Instrumental Neutron Activation analysis (INAA), at the Portuguese Nuclear Reactor, and the chemical elements were determined. Total deposition was collected simultaneously to the exposures and analyzed by inductively coupled plasma mass spectrometry (ICP-MS). Meteorological data were taken from the Portuguese Meteorology Institute and processed together with the chemical element concentrations. Results are discussed on basis of 1) influence of humidity/precipitation and temperature on bark and lichen transplant elemental accumulation, and 2) of different procedure of lichen exposure (biomass, usual one and flat - well-determined exposure area - pieces).

NAM.L15 (Id: 178)
STANDARDIZATION OF k_0-NAA FOR SHORT-LIVED NUCLIDES USING PNEUMATIC CARRIER FACILITY (PCF) AT BARC

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Neutron Activation Analysis (NAA) using short-lived radionuclides is advantageous in terms of less turn-around time of analysis. As a part of k_0-based NAA (single comparator NAA) standardization program for short-lived nuclides, pneumatic carrier facilities (PCF) at CIRUS and Dhruva reactors of Bhabha Atomic Research Centre (BARC) were characterized by determining sub-cadmium-to-epithermal neutron flux ratio (f) and epithermal neutron flux shape factor (α), which are two reactor based input parameters. Bare triple monitor method using ^{197}Au-^{94}Zr-^{96}Zr as well as ^{197}Au-^{50}Cr-^{98}Mo were used for α determination since there is a restriction in cadmium mass to be used as a wrapper/cover for sample irradiation. The f-value was calculated using bare bi-isotopic method. The k_0-factors of the short-lived nuclides (half-lives of nuclides 11 s to 60 min) of elements namely F, Se, Sc, Rb, Al, V, Ti, Cu, Ca, Mg, Br, I, Cl and In with respect to gold (^{197}Au) were determined in the present work. Gold standards (5-10 μg) were prepared on filter paper matrix. The elemental standards were prepared using primary standards in solid or liquid form and were irradiated with gold standard for 1 min in Dhruva reactor and 1-3 min in CIRUS reactor. The measurement of γ-rays was carried out using a 40 % relative efficiency HPGe-detector coupled to a digital spectrometer with loss-free counting facility and the sample-to-detector distance was maintained at 10-15 cm distance. The absolute detection efficiency of the detector was obtained using ^{133}Ba and ^{152}Eu multi γ-ray standard sources. The determined experimental k_0-factors of the isotopes were found to be in good agreement with the recommended k_0-factors in most of the cases. The k_0-factors of isotopes with widely varying Q_0 factors with respect to ^{197}Au (15.7) were compared with recent literature values. The method was validated by determining concentrations of elements like Al, V, Ti, Cu, Ca, Mg, Br and Cl in IAEA reference materials (RMs) namely SL-3, SL-1, Soil-7 and V-10 and Cu, V, I and Cl in synthetic multielement standards (SMELS-I) obtained from SCK-CEN, Belgium.

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NAM.L16 (Id: 253)
THE IMPACT OF POLYETHYLENE VIALS ON REACTOR CHANNEL CHARACTERIZATION IN k_0-NAA

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Reactor channel characterization is commonly performed by irradiation of bare and cadmium-covered "fluence rate-monitors", avoiding as much as possible the use of irradiation vials/capsules and spacers for positioning the monitors inside the channel. However, in routine k_0-neutron activation analysis it is generally necessary to pack the samples in polyethylene vials prior to irradiation. This work aims at studying the impact of polyethylene vials on the f (thermal-to-epithermal flux ratio) and α (epithermal flux distribution) parameters through the bare, cadmium-covered and cadmium-ratio methods. The accuracy of each method will be discussed.

NAM.L17 (Id: 84)
IS EXTRACTION OF Fe FROM IRON BASED MINERALS AN APPROPRIATE METHOD FOR DETERMINING TRACE ELEMENTS?

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Various trace elements in different types of iron based geological reference materials [JSS 804-1 (hematite), JSS 820-2 (limonite), SU-1 and SU-1a (iron-nickel-copper-cobalt ores)] were investigated using k_0-NAA in both forms: instrumental (k_0-INAA) and radioanalytical (k_0-RNAA). To avoid the interference of iron as a matrix element, firstly mineral was dissolved in aqua regia then liquid-liquid extraction procedure by diisopropyl ether (DIPE) in hydrochloric acid media was applied. Trace elements were determined using k_0-INAA after irradiation of water phase. Secondly, the mineral was irradiated and than radiochemical extraction of Fe were done using DIPE and trace elements were determined by k_0-RNAA in water phase. The k_0-NAA method, applied to determine the content of the investigated elements, after removal of Fe enabled to follow the distribution of 39 elements through their corresponding intermediate/medium and long half-lived radionuclides. The elimination of the matrix element lowered the detection limit for some trace elements compared to their corresponding values determined by k_0-INAA in powder form and with certified values for some elements. However, the results for some elements (e.g. As, Ba, Ca, Cs, Hf, K, Na, Zn and Zr) are higher comparing to k_0-INAA in powder due to the adsorption/desorption on glassware. The k_0-RNAA procedure shows better agreement than k_0-INAA with data obtained by k_0-INAA in powder mostly due to negligible blank contribution. The results of this work will be presented and discussed.

NAM.L18 (Id: 83)
ONLINE ^{14}C ANALYSIS OF ULTRA-SMALL SAMPLES WITH ACCELERATOR MASS SPECTROMETRY (AMS)

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^{14}C measurements of samples containing 5-50 μg carbon allow dating of archeological artifacts or environmental materials with only traces of organic carbon. Furthermore, fossil and non-fossil sources of carbonaceous aerosols can be apportioned using such measurements [1]. The widely-used preparation of solid targets for ^{14}C measurements, however, often remains the weak point of ^{14}C analysis of ultra-small samples. Even under optimized conditions, reaction yields are small, which results in large isotopic fractionations or even in total loss of unique samples [2]. These problems are solved with the implication of a gas ion source [3] at the next generation of small AMS machines: the mini radiocarbon dating system MICADAS is a prototype AMS that achieves the terminal voltage of 195 kV for ^{14}C measurements with a vacuum-insulated high voltage platform in contrast to previous systems based on conventional particle accelerators [4]. The self-constructed ion source of MICADAS was modified in order to insert CO_2 gases directly to the focal point of the sputtering cesium beam [3]. A first gas handling system was constructed to supply the gas ion source constantly with CO_2 from the sealed ampules. In this work, the concept of gas ion sources with its gas handling system will be presented. Furthermore, such ion sources allow on-line coupled systems of separation or combustion of organic compounds with AMS, which also will be elucidated. One example is the coupling of an elemental analyzer (EA), which generates a multifunctional system because it provides automated on-line combustion and ^{14}C measurement of any organic material. Therefore, it can broadly be employed in ^{14}C analysis. Carbon dioxide produced in the elemental analyzer is isolated from other combustion gases with a small external zeolite trap [5]. The purified CO_2 is transferred from the external trap to the syringe with a low helium flow. This coupling principle can now be adapted to other combustion systems. Different approaches will be discussed.

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NAM.L19 (Id: 264)
RADON MEASUREMENTS: A WAY TO DISSEMINATE SCIENTIFIC CULTURE AMONG YOUNG STUDENTS IN ITALY

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Usually, the lack of information cause to be afraid about what we don't know, imputing to it a greater hazard. On the contrary we face up without fear activities that have a high level of riskiness, but for which we have direct experience. In other words the subjective perception of the risk very often doesn't correspond to the objective and real risk of an activity. In particular the radioactivity theme is misled because it is almost unknown and the public links this concept to nuclear arms and to its usage in uncorrected way to produce energy in the nuclear power plants, even if in the last 50 years NPP for civil uses has significantly fewer casualties than any other source of energy. However, because public opinion is driven by emotions rather than rational knowledge based views nuclear power's association with nuclear weapons has contributed to its lack of acceptance in many places throughout the world and in particular in Italy the "nuclear issue" has been for a long time a taboo. A way to make the public more trusting to nuclear issue, that the people can have a more rationally reactions and could build up a personal understanding about these issues, is to discuss about this theme and about radioactivity and ionizing radiation, starting from young students. On these bases several Physics Departments with Lauree Scientifiche Project and sections of the National Institute of Nuclear Physics (INFN) with ENVIRAD-SPLASH Project would give to the students of secondary school and to their teachers the opportunity to face these themes with basic information and with an experimental activity. The approach is to have students engaged in activities that will allow them to understand how natural radioactivity is a part of our everyday environment. This would include how radiation enters our lives in different ways, to demonstrate that natural radioactive sources found in soil, water, and air contribute to our exposure to natural ionizing radiation and how this exposure effects human health. Depending on local situations and the specific experiences of the students, several approaches were used. However, collaboration among the schools was an essential element in the program's success. The core idea is that: a) to provide the students a furnished laboratory at their school so that they can measure the natural component of the radioactivity that surround us. In this exercise the measurement of the ^{222}Rn concentration is particularly well suited b) to show the different types of radiations including ionizing radiations and how they each relate to the other; c) to demonstrate how easily ionizing radiations can be measured; d) and to prove the fun a student can derive from discovery and detection of ionizing radiation in the environment. In this paper the experience of Milano reality is been reported in more detail. Another objective is to develop a new technique for teaching physics which will enhance scientific interest of students in applications of nuclear physics in both environmental and physical sciences.

NAM.L20 (Id: 31)
DETERMINATION OF ^{90}Sr IN SOIL, GRASS AND CEREALS

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Environmental samples in Upper Austria have been measured for ^{90}Sr in the year 2005. These measurements were conducted on behalf of the Federal Ministry of Agriculture, Forestry, Environment and Water Management and were part of a project called "Temelin Project". The aim of this project which started in 1992 is to detect any radiological impact of the nuclear power plant Temelin in the Czech Republic on the environment of Austria. After the nuclear weapon tests the average deposition of ^{90}Sr in Austria amounted to 3.3 kBq/m^2. In 1986 the average deposition was 0.9 kBq/m^2. To assess the actual condition in soil, grass and cereals ^{90}Sr was measured in these samples in 2005 and 2006. For all samples oxalate precipitation was conducted and strontium specific columns (Eichrom Industries, Inc.) were used. The calcium concentration in these samples was determined to estimate the amount of resin needed for the preparation. The prepacked 2 mL columns with particle size 100-150 µ were used for soil (lower limit of detection (LLD) below 2 Bq/kg). For grass and cereal samples columns were packed with the 100 - 150 µ resin to gain a LLD below 2 Bq/kg and below 0.1 Bq/kg respectively. After digestion of soil samples hydroxide precipitation was used as an additional separation step. The ^{90}Sr was measured by liquid scintillation counting and via ^{90}Y by Cherenkov radiation counting. For quality assurance reasons, first the initial strontium concentration in the sample was determined then a strontium carrier solution was added and after the separation steps the chemical recovery was determined by ICP-MS. Thus, no radioactive spike and just a small amount of the measuring solution were needed. The results are presented and discussed.

NAM.L21 (Id: 150)
DETERMINATION OF STRONTIUM-90 IN RAW MILK SAMPLES

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A rapid method to determine ^{90}Sr content in raw milk samples is presented. The isolation of radio-analyte from the matrix is accomplished by a pre-concentration stage on a cation exchange resin chromatographic column followed by a 6 hours incineration step at 550 C. Then, the separation of radiostrontium is performed on a column (20 cm x i.d. 12 mm) packed with Sr-Spec (Eichrom) resin. ^{90}Sr content is determined by Liquid Scintillation Analysis, measuring the analyte together with the ingrowing ^{90}Y daughter isotope, before of the establishment of the secular equilibrium. Results in terms of minimum detectable activity and yield mean value are reported.

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RADIONUCLIDE PROFILES OF ^{14}C, ^{90}Sr, ^{228}Th AND ^{232}Th FOR THE PURPOSE OF DATING HUMAN BONE TISSUES OR IVORY

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Radiocarbon dating is a standard method for archaeological findings. But the period which eludes distinct radiocarbon dating ranges from about 1957 to the present. This is due to the shape of the bombing curve of ^{14}C/C in creatures as a result of the above-ground nuclear weapon tests. But just this period is of certain interest for the purpose of determination of the post mortal interval. A basic approach to solve this problem is the compilation of isotopic profiles of suites radionuclides. Besides radiocarbon activity concentration ^{14}C/C further radionuclides as ^{90}Sr/Ca and ^{228}Th and ^{232}Th are of certain interest. During the global nuclear weapon fallout ^{90}Sr was ingested by human beings as well as animals resulting in curves of ^{90}Sr/Ca in human bone tissues being similar to that of ^{14}C /C [1]. The curves of both ^{90}Sr/Ca as well as ^{14}C/C show maximum values at about 1965 and higher values after 1957 than before. Before 1952 the intake of ^{90}Sr is negligible. In contrast to ^{90}Sr radiocarbon is produced by the cosmic radiation resulting in a value of ^{14}C/C of about 0.23 Bq ^{14}C/g C. If ^{90}Sr/Ca is not detected and ^{14}C/C is about 0.23 Bq ^{14}C/g C it can be concluded that life has ended before 1957. If ^{90}Sr/Ca and ^{14}C/C are detected both above the natural caused values the determination of the activity ratio of

^{228}Th/^{232}Th is able to provide a further specification of the post mortal interval. It has been proven from animal [2] and human [3] bone tissue data that the activity ratio of ^{228}Th/^{232}Th is far above unity at death but diminishes after a few years according to the decay of ^{228}Ra to draw near unity after about six decades after death. Applying low-level β-counting with gas-filled detectors for ^{90}Sr and α-spectrometry with semiconductor detectors for ^{228}Th and ^{232}Th and LSC Quantulus 1220 for ^{14}C very low values of lld are realized in combination with certain radiochemical separation and purification procedures which are presented here. They enable to realize values of lld below 0.003 ^{90}Sr/Ca and about 0.00009 Bq ^{232}Th/g ash and a total uncertainty of about 3.8% determining ^{14}C. Sample masses range from about 10 gram (ivory) to about 100 gram (bone). Results are presented dating several samples of ivory.

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NAM.L23 (Id: 160)

PROMPT γ ACTIVATION ANALYSIS AS PERFORMED IN BUDAPEST

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Institute of Isotopes has been a major center in further development of prompt γ activation analysis (PGAA). The paper describes the analytical procedure followed in Budapest. The analysis is based on the careful calibration of the detector system. The counting efficiency and non-linearity of the Compton-suppressed high-purity germanium detector is determined regularly, and enables accurate determination of activities and peak positions. Partial γ-ray production cross-sections have been determined for the most important prompt-γ lines of every naturally occurring chemical element using stoichiometric compounds and homogeneous mixtures. A method for qualitative and quantitative analysis has also been developed in our laboratory for the determination of the chemical composition of samples from the peak areas obtained from the prompt-γ spectra using the spectroscopic data library. The data library and the method have been used for the analysis of a large variety of samples both in Budapest and in other PGAA laboratories.

NAM.L24 (Id: 173)

UPGRADE OF THE PROMPT- γ ACTIVATION ANALYSIS (PGAA) AND NEUTRON INDUCED PROMPT - γ SPECTROSCOPY (NIPS) FACILITIES OF THE BUDAPEST RESEARCH REACTOR

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The prompt-γ activation analysis (PGAA) and the neutron induced prompt-γ spectroscopy (NIPS) facilities are located at the end of the guided beamline No.1 of the Budapest research reactor. In the last few years they were significantly upgraded. The partial replacement and realignment of the neutron guide elements resulted in a factor of four gain of the neutron flux. To make possible the unattended and independent operation of the two stations, a neutron flux monitor and computer-controlled beam shutters have been put into operation. This latter can be programmed to open and close the neutron beam according to a predefined time sequence, thus it will be an ideal tool to perform in-beam activation experiments. In order to make the PGAA facility even more productive, an automatic sample changer has been designed and manufactured. This can accommodate 16 samples, either solids, powders in sealed Teflon bags, or liquids in vials. A computer program has been developed to control such baTch runs. At the NIPS station, however, more attention is paid to the non-destructive analysis of bulky samples. Therefore a new, bigger and partially dismountable sample chamber is under construction. Moreover, the detection conditions are substantially improved by the installation of a new BGO Compton suppressor and massive lead shielding. Space is reserved for a moving table and for a neutron tomograph, to re-establish the combined prompt-γ activation imaging (PGAI) and neutron tomography (NT) equipment. The upgraded facilities offer new possibilities for the routine element analysis and also for the cutting-edge research challenges, and continue to serve international collaborations within the EU FP7 and many other projects.

NAM.L25 (Id: 261)

PROMPT γ-RAY ACTIVATION ANALYSIS FOR H, B, Bi, Pb AND Cd

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Neutron activation analysis (NAA) is a well established method for non-destructive determination of elemental composition and the analysis of trace elements. Prompt γ neutron activation analysis (PGAA or PGNAA) is a complementary radioanalytical technique to NAA for non-invasive investigations of samples with neutrons. One of the main advantages of this latter technique is the easy determination of some light elements like

H, Li, Be, B, C, N and S, that can not be easily determined or cannot be determined at all by NAA. Moreover, for the determination of elements like P, Cd, Gd and Pb PGAA is more effective thanks to the high neutron cross section of some isotopes that are not activated after the irradiation. The next advantage of PGAA is the rapidity: the irradiation takes max. few hours, the data analysis can be performed on the day of measurement and the sample is not activated for a long time. Furthermore, the sample needs no preparation for the PGAA measurement. At the first sight, the disadvantage of the PGAA to NAA are the detection limits. Compared to NAA they are usually higher, aside few exceptions, mainly in the ppm region. But speaking about PGAA, it is more correct to mention "dynamic detection limits" because they are strongly dependent on the matrix of the samples and on the background of the neutron beam (measurements are done during the sample's irradiation). The neutron beam guide NL4b at the research reactor Forschungsneutronenquelle Heinz Maier-Leibnitz (FRM II) in Garching bei München is dedicated to the new prompt γ neutron activation analysis facility. The fingerprint of this PGAA instrument is the cold neutron flux of a very high intensity due to the elliptically tapered design of the neutron beam guide. The neutrons are focused at the sample position with a maximum thermal equivalent flux of $6.3x10^{10}$ n/cm^2 s ($1.7x10^{10}$ n/cm^2 s using the measured mean energy of neutron spectrum, 1.83 MeV). The detection system is made by two HPGe detectors, both with a standard Compton suppression system. In this contribution few exemplary measurements of "typical PGAA elements" will be presented:

- Hydrogen is a very interesting element concerning superconducting materials and some clathrates. Its presence inside these compound can modify the material charachteristics;
- PGAA is very effective for the determination of boron. The dynamic range for this element was determined, as well as the detection limit and used for measurements in silicium wafers and nano-particles;
- One-dimensional distribution of bismuth in lead crystals was also determined with very good statistics.
- Cadmium is another "good element" for this technique. It was analysed in some biological samples for medical research.

NAM.L26 (Id: 199)
DEVELOPMENT OF γ ACTIVATION IMAGING METHOD FOR STUDYING OF ELEMENTS DISTRIBUTION IN SOME ORES

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Recently it has been demonstrated that activation of samples by bremsstrahlung radiation of microtron with further application of digital autoradiography is a prospective method for detection of noble elements micro inclusions in geological samples. A digital version of the autoradiography has been developed [1]. Current investigations have been directed to extension of digital γ-activation autoradiography capabilities to make a reasonable and effective alliance of this nuclear method with non-nuclear methods of local analysis. Among the developed approaches are: analysis of large size samples (tens of cm^2) and development of method for increasing selectivity using analysis of autoradiographic images while decay. Irradiation of large size thin section for fast autoradiographic detection of "zones of interest" containing elements in question seems to be a promising approaching for purposes of screening analysis of samples series. The further detailed analysis of the selected small zones may be done, for example, by SEM. For uniform irradiation of thin sections in the field of bremsstrahlung radiation having quite narrow distribution a special device and software have been developed [2]. It is obvious that activation autoradiography allows detecting distribution of the sum induced radionuclides having suitable decay parameters (β autoradiography). That is why the main task of investigations in autoradiography is optimization of irradiation/cooling times to obtain enough contrast detection of the required elements. Our recent efforts were directed to development of method to increase selectivity of the digital auto radiography by means of analysis of decay dynamics for each pixel of the whole image. It is clear that such analysis may be done just by means of software. The developed program loads a set of coaxial images obtained while sample cooling, necessary information on each image from MS Access data base and then estimates decay parameters (maximum two exponent mixture) for each pixel. It has been proved that even 8 bits depth grey scale images are suitable for stable evaluation of decay data. For visualization of the obtained results the program generates a set of secondary meta images representing "distribution" of the apparent half lives values for the preset ranges and their uncertainties. Using information on the isotope composition (obtained by γ-ray spectrometry), decay schemes one can re-model distribution of the apparent half lives to distribution of the correspondent elements (this part of the software is under testing). The developed method have been successfully applied for analysis of Pt-Pd micro inclusions in polymetallic ores of Norilsk area (North Eastern Siberia, Russia) having complex composition. Comparison with results of analysis by X-ray microanalysis is also given. The work is supporting by Russian Fund for Basic Research (grant N 07-03-00966-a).

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NAM.L27 (Id: 166)

γ-ACTIVATION DIGITAL AUTORADIOGRAPHY OF LARGE SAMPLES. EQUALIZATION OF ACTIVATING DOSE OVER THE MICROTRON-IRRADIATED SAMPLE SURFACE

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γ-activation autoradiography is an effective method for revealing features of noble elements distribution in geological samples [1]. Because the activating field of microtron bremsstrahlung displays sharp decrease of intensity, the area of the analyzed samples usually should not exceed 1 cm^2. However, such relatively small samples may be successfully analyzed by other instrumental methods such as SEM or X-ray microanalysis. To increase the competitiveness of γ-activation autoradiography we suggest to use it for screening analysis of the large area samples (10-30 cm^2), just for detection of "zones of interest" (for example, containing noble elements micro inclusions) implying their further detailed analysis by appropriate instrumental method. Equalization of the activation field over the sample surface may be achieved by suitable sample movement. The suggested kinematic scheme of the irradiation device assumes rotation of the sample and its simultaneous reciprocating movement. Execution is simple enough to provide reliable functioning of the device in strong radiation field. Prior to the construction of a real device the dose accumulation during activation has been modeled on a PC. Program "Dose Modeling" estimates the uniformity of the accumulated dose at different parameters of the sample movement. It is demonstrated that the assumed kinematic arrangement provides considerable smoothing of a dose over a surface though a certain residual non-uniformity of a dose still remains. For full "smoothing" of the activation dose it has been suggested to use mathematical pixel-by-pixel correction of the autoradiogram image by means of the developed software ("Dose Correction"). For such correction a copper disk-monitor should be irradiated together with the sample. The program evaluates the parameters of the distribution of the acquired activation dose of the disk-monitor (single or superimposed Gauss distributions) and makes a correction of the analyzed sample autoradiogram. The disk-monitor autoradiogram is also treated by the program to estimate the quality of dose correction. By means of program "Dose Modeling" the optimal operational parameters of the device have been determined: the ratio of frequencies of the sample rotation to reciprocating movement should be not less than 5.5; duration of an irradiation should be not less than 300 seconds. The device for irradiation of samples has been prepared and tested. It consists of a platform making reciprocating movement by the alternating current motor by means of the crank mechanism; the sample holder and the second motor rotating the sample are fixed on the platform. Results of experimental check of the developed irradiation system have confirmed completely its operational capacity.

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NAM.L28 (Id: 197)

DETERMINATION OF FLUORINE IN SELECTED REFERENCE MATERIALS BY INSTRUMENTAL PHOTON ACTIVATION ANALYSIS

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Possibilities of nondestructive determination of fluorine in selected geological (coals, rocks) and biological reference materials by instrumental photon activation analysis (IPAA) using the MT 25 microtron have been studied. The determination has been based on counting of the non-specific 511 keV annihilation γ rays of ^{18}F, product of the photonuclear reaction F^{19} (γ, n) ^{18}F and a pure positron emitter. To eliminate production of ^{11}C, irradiation was performed below the threshold energy of its formation, i.e., below 19 MeV. The determination was interfered by simultaneous formation of several other positron emitters, particularly ^{45}Ti and ^{34m}Cl. Interference contributions from ^{22}Na, ^{44}Sc, ^{89}Zr, ^{74}As, and ^{84}Rb are small or negligible. By further optimization of beam energy and irradiation-decay-counting times and using correction standards for the interfering nuclides, IPAA allowed determination of fluorine in selected reference materials at the ten mg/kg level.

NAM.L29 (Id: 231)

CHARACTERIZATION OF AN EXTRACTION CHROMATOGRAPHIC RESIN FOR THE SEPARATION AND DETERMINATION OF 36Cl

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The monitoring of long-lived radionuclides is of great importance in the context of the surveillance of nuclear facilities, during their operation as well as during their decommissioning. This is especially true for radionuclides of rather volatile elements as chlorine and iodine, main interest being in ^{36}Cl and ^{129}I. LSC is a widely used measurement technique for the determination of ^{36}Cl that requires a thorough and selective sample preparation in order to give accurate results. Sample preparation methods frequently employed such as volatilization and/or repeated precipitation steps can be rather elaborate and time-consuming, it is thus attempted to develop an easy to use extraction chromatographic resin that allows extraction, and subsequent separation, of Cl and I from pretreated environmental and decommissioning samples and that allows also their determination via LSC. The results of the characterization of the resin including k' values of potential interferents and of the method development are presented as well as some first results of the analysis of real samples.

NAM.L30 (Id: 240)

PROBABILITY FOR TYPE I ERRORS IN γ-RAY SPECTROMETRIC MEASUREMENTS OF DRINKING WATER SAMPLES

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In γ-ray spectrometry the activities are calculated from areas of the peaks appearing in the spectrum. If a peak used in the activity calculations appears in the spectrometer background its count rate must be corrected for the background contribution. The significance of this correction becomes essential in spectral analyzes where activities close to the minimum detectable activity need to be determined. The presence of radionuclei in the sample, that contribute to the spectrometer background, can only be detected from the excess of the count rate in the peaks over their background level. Therefore, the radionuclides can be categorized in two groups: - Radionuclides that do not appear in the spectrometer background. The probability of the wrong detection of these radionuclides is given by the sensitivity at which the peak-search algorithm recognizes the background fluctuations as peaks. - Radionuclides that appear in the spectrometer background. Here, a radionuclide is identified if the count rate in its peaks exceeds the background level. In this case the probability of a false detection is given by a criterion, on the basis of which the number and the significance of the peaks where the excess occurred decide on the presence of the radionuclide in the sample. It follows that for the second group, the minimum detectable activity that can be attained depends on the accuracy with which the background is determined and on its stability over time. It should be noted that if the contribution from the sample to a peak area is small as compared to the contribution of the background, there exists an equal probability that the result of the background subtraction is larger than zero or smaller than zero. To check the rate of falsely detected radionuclides (type I errors) an analysis was made in which the background spectra were treated as sample spectra. From this it is known that in the analysis of these spectra all the detections of radionuclides are false. The spectral analyzes were preformed with Canberra's GENE ESP spectroscopy system. The nuclide library that was used is composed of radionuclides usually found in the environment and radionuclides that occur in the effluents from nuclear power reactors and hospitals. In the case of multi-γ-ray emitters, the nuclide library includes the most abundant γ-rays as well as the γ-rays that cause interferences with other radionuclides, in order to correct the activities for these interferences. The criterion that determines whether a radionuclide has been detected is based on a comparison of the sum of the emission probabilities of the γ-rays, where the count rate in the spectrum exceeds its background value and the sum of the emission probabilities for all the γ-rays of this radionuclide that are included in the library. If the ratio of both sums exceeds a predetermined value, the γ-ray emitter is considered as being present in the sample. In the presentation the rates of type I errors will be presented for single γ-ray emitters and multi γ-ray emitters. The empirical minimum detectable activities for water samples for these radionuclides will be given.

NAM.L31 (Id: 43)

AN INTER-CALIBRATION CAMPAIGN USING VARIOUS SELECTED PU SPIKE ISOTOPIC REFERENCE MATERIALS

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The measurement of amounts of plutonium is recognised as one of the most important tasks in fissile material control. Public opinion is especially sensitive to this element, which presents analysts with a difficult task of measuring plutonium at all levels from large (multi-gram) amounts down to traces in the environment. The Institute for Reference Materials and Measurements (IRMM) is one of the few institutes worldwide that produces and certifies plutonium reference materials. They are part of a systematic IRMM programme to supply spike isotopic reference materials of various isotopes, in particular uranium and plutonium, at different concentrations. Plutonium spike solutions present unusual problems that impinge on the certification of the isotope amounts. Due to radioactive decay certified values have to be constantly corrected, a process that leads to an increase in uncertainty depending on the spike isotope composition. The solutions also have a possibility of being unstable over time, probably due to radiolysis of the solutions. At IRMM plutonium spikes are available for the isotopes ^{239}Pu, ^{240}Pu, ^{242}Pu and ^{244}Pu. This enables us to make an inter-comparison of the values of these spikes. An Inter-calibration campaign using state-of-the art measurement procedures has therefore been carried out at IRMM linking the plutonium spike reference materials available at the institute. The following spikes were used in the program: IRMM-049c (94.1% ^{242}Pu), IRMM-049b (99.9% ^{242}Pu), IRMM-046b (94.3% ^{242}Pu and 98.0% ^{233}U), IRMM-083 (98.97% ^{240}Pu) and IRMM-042a (97.9% ^{244}Pu), IRMM-081a (97.8% ^{239}Pu) from CETAMA were used. As comparison material a recently prepared ^{239}Pu large-sized dried (LSD) spike, IRMM-1027M and external isotopic reference materials NBL CRM-131 and CETAMA MP2 were used for isotope dilution mass spectrometry (IDMS). The uncertainties for the IDMS were estimated according to the Guide to the Expression of Uncertainty in Measurement (GUM). The results obtained by IDMS using the various spike isotopic reference materials will be presented and discussed. This intercalibration study confirms the traceability of the values of the plutonium isotopic contents to the SI. Furthermore conclusions can be drawn for the need of recertification of particular reference materials.

NAM.L32 (Id: 246)

RADIOANALYTICAL DETERMINATION OF ACTINIDES IN REFRACTORY MATRICES BY LINKING ALKALI FUSION WITH SOLVENT EXTRACTION AND CHROMATOGRAPHY EXTRACTION.

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In radiochemical analyses with environmental or industrial samples sometimes happens that certain matrices have a refractory behaviour and usual leaching processes cannot deal with its dissolution properly. There are in the bibliography different ways to solve this problem as the use of microwaves digestions or HF digestions (or both together) and also fusion techniques among others. This work presents a method used previously for ICP-MS measurements that have been modified and readapted for the digestion of refractory samples followed by a separation process with solvent extraction for the determination of actinides (U and Th isotopes) in refractory samples by α-spectrometry. The separation process was made also with extraction chromatography resins (UTEVA resins) in order to test both separation methods and to choose the more suitable one. In the first stage, we worked with samples that were only leached with aqua regia followed of separation process with TBP to finally electroplate the isolated uranium and thorium fractions in stainless-steel dishes. The samples were measured by α-spectrometry with PIPS detectors. Aliquots of these samples were as well measured via γ spectrometry in order to check the results produced by α spectrometry. Disagreement was found between the results obtained by both techniques due to the not complete dissolution of the samples analysed by α-spectrometry. In the second stage, the found disagreement was solved by the application of an alkali fusion technique for a total dissolution of the sample before the application of the separation methods and α spectrometric measurements. The fusion technique with $KHSO_4$ is easily applicable, fast (it takes less than two hours instead of ten-twenty necessary in leachings) and it does not need sophisticated tools to be executed. Regarding the use of UTEVA resins or TBP process for the separation of actinides, it is necessary to point out that both techniques show similar chemical yields for uranium and thorium isotopes although the UTEVAS resins are less time-consuming than the TBP process. In addition, the possibility to reuse the resins makes them more useful from an economical point of view, in opposition to the general idea that many laboratories have about this topic.

NAM.L33 (Id: 109)

DETECTION OF TRITIUM AND α-DECAYING ISOTOPES IN L/ILW BY HELIUM MEASUREMENTS

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As decay products, helium isotopes can clearly indicate the presence of tritium and α decaying isotopes in a closed system. This study presents the helium and neon measurements and their interpretation of long-term headspace gas investigations in L/ILW waste drums from Paks Nuclear Power Plant and closed vaults of the Radioactive Waste Treatment and Disposal Facility, Püspökszilágy, Hungary. Development of special sampling methods and preparation lines as well as isotope-analytical measurements of the headspace gas samples were done in the Laboratory of Environmental Studies in the ATOMKI. In the gas samples helium isotopes as well as neon isotopes have been determined mass spectrometrically. While neon content can be of atmospheric origin only, helium can be produced either by α decay (^4He) or decay of tritium (^3He). He/Ne ratios have been used to determine the different origin of the helium isotopes. Helium isotope ratios always indicated ^3He enrichment in the headspace gases produced by the decay of the tritium in the waste. Using the recent ^3He concentration in headspace gas the total amount of ^3H restored in L/ILW vaults was estimated. The investigated seven different vaults were closed between 1979 and 1995 when they had been full with L/ILW. The calculated tritium activities based on the He measurements showed good agreement with the documented isotope inventory of the vaults. Typical tritium activity concentrations were between 0.1 and 10 Bq/L gas in the drums and between 10 and 1000 Bq/L gas in the vaults. Additionally, one drum showed a higher He/Ne ratio compared to air, which clearly indicates ^4He excess, thus the presence an α source in the waste.

NAM.L34 (Id: 162)

DEVELOPMENT EXPERIENCE AND CONCEPT OF ANALYTICAL SUPPORT COMPLEX FOR INDUSTRIAL SCALE FAST REACTOR FUEL PRODUCTION

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Requirements imposed to nuclear fuel in terms of stoichiometric composition and chemical purity are rather high and depend on the technical and operating conditions of fuel rods in a nuclear reactor. RIAR has long operating experience with the production facility for granulated mixed nuclear fuel (MOX fuel) using electrolytic precipitation of uranium and plutonium oxides from molten chlorides of alkaline metals which are subsequently used for fabrication of vibropacked fuel pins. In connection with this RIAR has developed, metrologically certified and uses a set of analytical procedures, which ensure process control, certification of the obtained fuel and analysis of intermediate process products, waste and environmental objects for the content of uranium, plutonium and transplutonium elements. Analytical control of the pyrochemical mixed fuel production process is performed by periodic analysis of the molten alkali metal chloride samples for the content of uranium, plutonium, americium and lead. The procedures are used for direct spectrophotometric determination of uranium and lead, α spectrometric determination of plutonium and analysis of the above elements with the preliminary extraction-chromatographic separation. Each baTch of the manufactured MOX fuel is subject to certification including determination of the mass portion of uranium, plutonium, americium, chlorine, fluoride, carbon, moisture, cation impurities and nuclide composition of uranium and plutonium, oxygen coefficient (O/M ratio) and bulk granulate density. The set of procedures listed above has been successfully used at RIAR for more than 25 years and addresses the technical requirements imposed on final products. The paper describes the procedures developed at RIAR and proposes the ways of their updating and adaptation for modern equipment.

Posters

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THE USE OF SEDIMENT-WAX PUCKS IN γ-RAY SPECTROMETRY FOR LUMINESCENCE DATING: OPTIMIZED FABRICATION AND PROPERTIES

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Low-background Ge γ-ray spectrometry is a very suitable tool for use in luminescence dating of sediments, because it not only yields the annual (K, Th, U) radiation dose, but also gives information on the secular equilibrium in the ^{238}U decay series. At the Radchem-15 conference (CZ-Marianske Lazne, 2006) we demonstrated that wax is the material of choice for encapsulating the sediment (as a solidified mix of sediment with molten wax), because ^{222}Rn is not leaking out of the source. During the use of these sediment-wax pucks in the course of the years we noticed, however, that the preparation protocol was not optimal. Indeed, the cylindrical puck had a concave top surface; the puck therefore needed shaping into a reproducible geometry, causing a large loss of both time and sample. Quite often, the pucks were also not sufficiently homogeneous due to air inclusions. In this paper, we report on a modified fabrication procedure. The main improvements relate to the shaping of the puck. An improved mould is used, in which the liquid sediment-wax mixture is poured after reaching a more viscous state, i.e. after some cooling down. This eliminates shrinking of the mass solidifying in the mould, thus avoiding air inclusions and deviations from the pure cylinder geometry. The reproducibility and homogeneity of the pucks obtained in this way are excellent. The puck height is determined by availability of sample (as the wax/sediment ratio is kept fixed and the diameter of the mould is constant) and this variation in source geometry must be accounted for. We report on a performance test of the SOLCOI software to compute the detection efficiency for different γ-rays in case of such variations in puck height. To this purpose, calculated efficiencies are compared with values that are experimentally obtained using puck-calibrants of varying height. Finally, we evaluate the extent to which variations in source composition and dimension can be tolerated with specific relevance to (the limits on) the precision and accuracy that are generally attained in luminescence dating of sediments. We conclude that an improved, flexible and user-friendly laboratory practice has been developed for determining the concentration of naturally occurring radionuclides in sedimentary materials using low-background Ge γ-ray spectrometry.

NAM.P02 (Id: 19)

DETERMINATION OF ^{90}Sr AND ^{210}Pb IN DEER BONE SAMPLES BY LIQUID SCINTILLATION COUNTING AFTER IONIC EXCHANGE PROCEDURES

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Environmental monitoring of hazardous radionuclides is an important issue. ^{90}Sr can be found in the environment due to the global fallout from atmospheric nuclear explosions and the Chernobyl accident in 1986. It is one of the most hazardous fission products due to its chemical similarity with calcium, because it can be accumulated in bone tissue delivering irradiation doses to the bone marrow. ^{210}Pb is a naturally occurring radionuclide which also accumulates in bones. If these two nuclides are measured, a natural and an anthropogenic activity concentration can be compared for the respective samples. Deer bone samples were selected as a feasible environmental contamination indicator. This work describes different procedures for the isolation of ^{90}Sr and ^{210}Pb from deer bones by anion exchange methods and their sequential measurement. The aim was to obtain pure ^{90}Sr and ^{210}Pb spectra in order to avoid spectrum deconvolution procedures. To prevent collection of Pb on the Sr•Spec® resin we first separated Pb on a Dowex anion exchange column. Sr, which is not held back on the Dowex column, was then purified using Sr•Spec® resin: first Ca and the Ra isotopes were eluted with 3M HNO_3 and then Sr was eluted with distilled water. With this two-step procedure of lead separation on Dowex followed by Sr purification on Sr•Spec®, pure ^{210}Pb and ^{90}Sr spectra can be achieved by liquid scintillation counting of the respective eluting solutions. The chemical yield of both steps was determined by ICP-MS. Our results of re-measured samples show satisfying agreement with data obtained by a modified Sr•Spec® method and also by the "classical" ^{90}Sr determination using fuming nitric acid.

NAM.P03 (Id: 27)
COMPARATIVE KINETIC STUDIES ON THE CORROSION PROCESS USING TWO METHODS BASED ON THE β-RAYS RETENTION AND ATOMIC ABSORPTION SPECTROSCOPY

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The paper deals with the corrosion kinetics of some steels using two different methods of investigation. The first one is a radiochemical method based on the retention of β-rays emitted by a ^{204}Tl source, by the iron ions transferred into the solution by anodic dissolution of metal samples. The second one is based on the atomic absorption spectroscopy, follows the increase in time of the concentration of the same iron ions in the given aggressive medium. Nitric acid of different concentrations was used as corrosive medium. The obtained experimental data result in two different rate constants that can quantitatively characterize the corrosion process for each given reactant system by the proposed methods. The values obtained by these two methods of investigation are generally in keeping, however more precise will be those obtained through atomic absorption spectroscopy due to its higher precision in the determination of the concentration of the iron ions transferred into the corrosive solutions. However, the proposed radiochemical method seems easier and faster.

NAM.P04 (Id: 42)
GEOCHEMICAL STUDIES IN WATER BODIES OF WESTERN PARAGUAY

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Minor and trace elements composition of bottom sediments from water bodies in Western Paraguay have been investigated by XRF techniques to determine their correlation as well as provenance. The analysis of complex spectra was performed by the AXIL software and the quantitative analysis by the QAES software. Analysed trace elements were refractories Rb, Ba, Nb, La, Ce, Sr, Nd, Zr, Y and other HFSE as Cr, Ni, Cu, together with Zn. Minor elements were Ti, Mn, Fe which are often related to the above refractories. According to their normalized spidergrams two sets of sediments can be differenciated. Those from Pilcomayo and Verde Rivers as well as from km 165 Wetland show LREE enrichment, negative Nb and Ti anomalies and no spike at Zr. On the other hand, sediments from Confuso, Jehe, Negro, and Montelindo Rivers have spidergrams very alike, with strong negative anomaly at Nb, Nd and Ti whereas a positive at Zr. Further, in the former there is a strong correlation of Fe versus refractories; such a correlation does not exist in the latter.

NAM.P05 (Id: 48)
DETERMINATION OF RADIOSTRONTIUM IN FOOD AND WATER SAMPLES USING FUMING NITRIC ACID

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The objective of this work was the determination of radiostrontium content in selected food and tap water samples collected within environmental monitoring programme of Federation of Bosnia and Herzegovina. It included food samples of vegetables, fruit, meat, cereal, milk and milk products. The radiostrontium content in environmental samples was determined by the fuming nitric acid method. This classic way of analyzing radiostrontium makes use of the low solubility of $Sr(NO_3)_2$ in fuming nitric acid solutions. The procedure included successive precipitation of $^{89/90}$Sr as nitrate several times to achieve a good separation from most elements, especially Ca. Chromate precipitations were performed to eliminate Ba, Ra and Pb, followed by hydroxide precipitations to eliminate traces of Y. Counting sources were prepared by weighing $SrCO_3$ on a planchette and measured on a low background proportional gas flow β counter. The Sr yield was determined using Sr carrier and it varied between 60-90%. Quality control was ensured through analysis of reference materials, blank determination and background determination. Activity concentration of radiostrontium varied between 0.012 Bq/kg and 0.428 Bq/kg for vegetables samples, 0.007 Bq/kg and 0.257 Bq/kg for fruit samples, 0.0995 Bq/kg and 0.289 Bq/kg for cereal samples, 0.024 Bq/L and 0.113 Bq/L for milk and milk products, below MDA (0.004 Bq/kg) for meat samples and for tap water samples between 0.878 Bq/kg and 3.78 Bq/m^3.

NAM.P06 (Id: 56)
EFFECT OF AL COATING ON LOSS OF LIGHT ELEMENTS IN POLYETHYLENE NAPHTHALATE FOILS DURING MEV PROTON BEAM IRRADIATION

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For metallic and semiconductor materials, compositional changes as well as structural changes are considered to be negligibly small during ion beam analysis (IBA) with protons and helium ions of MeV energy. For insulators, by contrast, electronic excitations and ionization produced by MeV ion beams sometimes cause degradation of targets. Especially for polymer targets, light elements such as hydrogen, carbon and oxygen tend to be easily released from the targets, resulting in a serious compositional change during IBA. The technique to reduce the compositional change caused by MeV ion beam irradiation is required for precise compositional determination in IBA. In the present study, backscattering and forward scattering spectrometry with 2.2 MeV protons have been applied to detect light elements of a polyethylene naphthalate (PEN) foil. During analyzing a PEN foil of 4 mm thick, all the light elements gradually decreased in areal density as irradiation fluence increased and the contents of 9 % H, 7 % C and 33 % O were eventually released up to a fluence of 2.0x10^{16} protons/cm^2 (corresponding to the integrated charge of 50 micro coulomb). We tried aluminum deposition on the one side of the PEN foil, in order to reduce the loss of light elements. As a result, the losses of H, C, and O were 2 %, 0.5%, and 22 % of initial contents, respectively, considerably smaller than those without Al deposition. The mechanism of light element losses during MeV proton irradiation and the effect of Al deposition will be discussed in the conference. We have been investigating if a PEN foil is one of the promising candidates for an exit window in air-PIXE system, with a help of Al coating.

NAM.P07 (Id: 72)
DETERMINATION OF ^{210}Po AND URANIUM IN HIGH SALINITY WATER SAMPLES

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Among of the naturally occurring radionuclides in the different types of water the long lived uranium isotope ^{238}U, two radium isotopes: ^{228}Ra, ^{226}Ra, and ^{210}Po are of practical importance due to their radiotoxicity. The observed levels of these radionuclides in underground water depend on the chemical compositions of the adjacent geological formation. Under favourable conditions, for example in the presence of chloride and bicarbonate anions, solubility of mineral rock components including trace amounts of radionuclides increases. A role of the underground water supplying systems including geothermal water is recently rapidly growing as they are used not only for energetic, balneological or recreational purposes but as well as a source of drinking water that should be in accordance with EU Directive 98/83/EC and WHO recommendation. The aim of these study was to elaborate a fast and efficient method for simultaneous determination of uranium and ^{210}Po in high salinity water samples. ^{210}Po and uranium radionuclides were preconcentrated from 0.5 dm^3 saline media by co-precipitation with hydrated MnO$_2$, followed by dissolution of the precipitate in 200 mL of 1 M HCl. Uranium radionuclides can be directly determined by ICP MS method, using part of this solution because the total salinity decreases below 1 g/dm^3. From the second part of solution, before ^{210}Po separation for liquid scintillation counting, majority of naturally occurring α-emitting radionuclides (uranium, thorium and protactinium) are stripped by extraction with 50 % solution of HDEHP in toluene. Finally ^{210}Po is simply separated from other interfering nuclides by direct transfer to an extractive scintillator containing 5 % of trioctylphosphine oxide (TOPO) in Ultima Gold F cocktail and determined by a α/β separation liquid scintillation technique. The detection limits are <1 mBq/dm^3 for ^{210}Po and 0.01 ppb for ^{238}U. Quality assurance of the worked out method was checked out in two ways: 1) participation in the IAEA interlaboratory studies (see Table 1) for ^{210}Po determination in water samples 2) determination of uranium and ^{210}Po in Standard Reference Material IAEA-381 - Irish Sea water. Uranium recovery for these samples was in the range 0.88±0.05. ^{210}Po activity is not reported for this SRM, therefore its activity determined by elaborated method was compared with that obtained by direct ^{210}Po deposition on silver discs and α spectrometry counting. These two methods gave practically the same value 30.5±2.3 mBq/dm^3 and 30.0±3.0 mBq/dm^3, respectively.

Table 1. **Results (in Bq/kg) of interlaboratory studies ^{210}Po in acidified water**

Sample.	IAEA value	IAEA unc	Measured value	Measured unc.	Measured/IAEA
1	52.8	1.4	52.0	0.6	0.99
2	101.6	2.8	95.3	1	0.94
3	52.8	1.4	52.1	0.7	0.99
4	101.6	2.8	99.3	1	0.98
5	blank	-	0.3	0.03	-

NAM.P08 (Id: 75)
APPLICABILITY OF K0-BASED NEUTRON ACTIVATION ANALYSIS USING A COMPTON SUPPRESSION γ-RAY SPECTROMETER

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Compton suppression system (CSS) in conjunction with γ-ray spectrometer allows to lower the detection limits for certain elements by reducing the background levels in the acquired γ-ray spectra. The calibration of CSS for use in the k$_0$-based neutron activation analysis (k$_0$-NAA) has been performed in normal mode and applied to Compton suppression mode. In this case, efficiency data are only applied to radionuclides that emit non-coincident γ-rays. In this work the applicability of the k$_0$-NAA method using CSS (k$_0$-CSNAA) in Compton suppression mode for radionuclides that emit coincident γ-rays is tested. The SMELS (a synthetic multi-element standard) consisting of nuclides with different γ-ray energy and Q$_0$ values was used to evaluate the Compton suppression factor for several radionuclides of interest, e.g. ^{60}Co, ^{51}Cr, ^{152}Eu, ^{59}Fe, ^{75}Se, ^{46}Sc and ^{85}Sr. The applicability of k$_0$-CSNAA has been tested by analyzing several reference materials, i.e. Coal Fly Ash (NIST-SRM-1633a), Soil (IAEA-Soil-7) and Lichen Material (IAEA-CRM-336). Preliminary results of the test are presented and discussed.

NAM.P09 (Id: 82)
ALPHA RADIOMETRY OF URANIUM BY LIQUID SCINTILLATION COUNTING AFTER PRE-CONCENTRATION BY CLOUD POINT EXTRACTION

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The aim of this study is the radiometric determination of uranium in waters by liquid scintillation counting (LSC) after pre-concentration of the element by cloud point extraction (CPE). For CPE, tributyl phosphate (TBP) is used as the complexing agent and (1,1,3,3-tetramethylbutyl)phenyl-polyethylene glycol (Triton X-114) as the surfactant. The measurement is performed after phase separation by mixing of the surfactant phase with the liquid scintillation cocktail. The effect of experimental conditions such as pH, reactant ratio (e.g. V(TBP)/V(Triton), ionic strength (e.g. [NaCl]) and the presence of other chemical species (e.g. Ca^{2+} and Fe^{3+} ions as well as humic acid and silica colloids) on CPE has been investigated. According to the experimental results the total method efficiency is (13±2)% and the separation efficiency (60±10)% for the optimum pH and reactant ratio (V(TBP)/V(Triton) = 0.1). Regarding the other parameters, generally Ca^{2+} and Fe^{3+} ions as well as the presence of colloidal species in solution (even at low concentrations) results in significant decrease of the separation efficiency. On the other hand increasing [NaCl] leads to enhancement of separation efficiency. The detection limit under optimum experimental conditions has been found to be 0.5 Bq/L indicating that the method could be applied only to waters samples with increased uranium concentration. Moreover, the negative effect of the chemical species found in natural waters limits the applicability of the method with the respect to environmental radioactivity measurements.

NAM.P10 (Id: 88)
MERCURY IN BACH HO CRUDE OIL OF VIETNAM AS DETERMINED BY k_0-BASED INSTRUMENTAL NEUTRON ACTIVATION ANALYSIS

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The k_0-based instrumental neutron activation analysis (k_0-INAA) using the Dalat research reactor of 500 kW nominal power with a thermal neutron flux about $3.5x10^{12} cm^{-2}s^{-1}$ has been studied and developed in order to determine Hg (total) in Bach Ho crude oil of Vietnam. Both radionuclides ^{197}Hg (2.7 d half-life, 77.3 keV γ-line) and ^{203}Hg (46.6 d, 279.2 keV) were used with different irradiation and measurement modes in order to calculate the concentration of the element. An evaluation of the limit of detection, precision and accuracy as well as convenience in the use of the two radionuclides was performed. The corrections of spectral interferences by ^{75}Se (119.8 d, 279.5 keV) on ^{203}Hg and the deconvolution of complex multiplets in region of 70-85 keV for ^{197}Hg and the problem of Hg loss during irradiation were considered. The mean concentration and range of concentrations of Hg in Bach Ho crude oil of Vietnam were obtained through the analysis of 90 samples and the statistical processing.

NAM.P11 (Id: 94)
ALPHA RADIOMETRIC DETERMINATION OF PLUTONIUM AND URANIUM ISOTOPES AFTER SEPARATION OF THE RADIONUCLIDES BY CATION EXCHANGE AND LIQUID EXTRACTION

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The radioactivity concentration of uranium and plutonium isotopes (^{236}Pu and ^{232}U) in water samples has been determined by means of α spectroscopy after chemical separation of the radionuclides by cation exchange and liquid-liquid extraction using the Chelex-100 resin and 30%TBP/dodecane. Method calibration using $^{236}Pu/^{232}U$ standard solutions results in a detector efficiency of 19% and in a cation exchange separation efficiency which varies between 60% and 90%, and 50% and 70%, for uranium and plutonium, respectively, depending on the radionuclide concentration in the sample. Application of the method to test solutions of constant radionuclide concentration and variable composition (0.1, 0.5 and 1 M NaCl; 0.1 and 0.5 M $Ca(NO_3)_2$; 0.1 and 1 mM $FeCl_3$; 10 and 100 ppm SiO_2; 10 and 100 ppm humic acid) shows generally that the cation exchange efficiency is not significantly affected by the presence of major components, except for increased Fe(III) concentrations in solution (> 0.1 mM). The liquid-liquid extraction efficiency is almost similar for both radioelements and varies between 50% and 80%, depending on the radionuclide concentration in the sample. Taking into account that the electrodeposition of the radionuclides under study is quantitative, the total method efficiency is calculated to vary between 30-70% and between 25-55%, for the uranium and plutonium isotopes, respectively. The detection limit of the α spectrometric system has been found to be 0.2 mBq/L, suggesting that the method could be successfully applied for the radiometric analysis of uranium and plutonium isotopes in natural waters

NAM.P12 (Id: 104)
PULSE SHAPE ANALYSIS TO REDUCE THE BACKGROUND OF BEGE DETECTORS

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Three methods for reducing the background and thus the detection limits for γ spectrometry using HPGe-detectors are combined here. 1. A passive reduction of the background is achieved by using radiopure materials for the detector and its shield 2. An effective shielding from cosmic rays is achieved in the laboratory, which is located 225 m underground. 3. An active background reduction is achieved using pulse shape analysis. Since the invention of the Ge-detectors in 1962 researchers have investigated ways of using the pulse shape for discriminating between pulses from various types of interactions in the Ge-crystal. The lack of success in this field has triggered developments of more complicated detector systems such as segmented crystals, pixels and strip detectors. When p-type point contact Ge-detectors with a shaped electric field (called BEGe-detectors, Broad Energy Germanium, by the company Canberra) started to appear on the market, the possibility opened for using solely the pulse shape of the one electrode for making analysis. The drift of the electrons and holes towards the electrodes produces different pulse shapes depending on how and where the energy of the photon was deposited in the crystal. When all the energy is deposited in one point, single-site event (SSE), the pulse shape has one peak, but when the energy is deposited is several points, a multiple-site event (MSE), the shape of the pulse presents several peaks. The work presented here is aimed at the study and characterization of the background and pulse-shape of two BEGe detectors with 19% and 50% relative efficiency by using a pulse shape discrimination technique. Since the signal of the read-out electrode depends on the size of the detector the discrimination level that can be achieved with each detector is different.

NAM.P13 (Id: 153)
NORM AT INDOOR ENVIRONMENTS USING AEROSOLS PASSIVELY COLLECTED AT CLASSROOMS OF LISBON BASIC SCHOOLS

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This work aims to develop methods characterizing the composition of the total particulate matter (TPM) collected inside classrooms of three elementary schools, in Lisbon, Portugal. The selection of the schools was based on their different distance from the city centre: one at downtown, other at the city border and the other at middle distance between both. Quartz and polycarbonate filters are being used. Each filter has a 47 mm diameter and was exposed inside plastic Petri slides. TPM masses are being collected passively during 2009/2010. Although the filters are being exposed inside a small area, significant differences between the masses are observed. Exposed filters and blanks are being measured during a 3 day period in a γ-X hyperpure germanium detector aiming the detection of natural radionuclides. The following natural radionuclides could be found: 1) ^{210}Pb, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{234}Th originated from ^{238}U, 2) ^{212}Pb and ^{228}Ac originated from ^{232}Th, 3) ^{208}Tl originated from ^{237}Np, 4) ^{40}K originated from ^{39}K. Millipore polycarbonate filters were quite adequate for all measurements except for ^{210}Pb, ^{226}Ra and ^{234}Th for which quartz filters were found to be more suitable. Results are compared with literature ones and discussed under the point of view of their origin. Methods are developed in order to get accurate efficiency calculations for the 47 mm diametr filters.

NAM.P14 (Id: 154)
THE USE OF COINCIDENCE SUMMING EFFECT IN γ SPECTROMETRY FOR THE DETERMINATION OF FULL ENERGY PHOTOPEAK EFFICIENCY AND ACTIVITY OF THE ^{60}Co-60 POINT SOURCE

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A study of coincidence summing of γ-rays of ^{60}Co will be presented. In γ spectrometry with germanium detectors, the summing effect have to be taken into account at low source detector distance. The peaks due to coincidence summing of X+X, X+γ,γ+γ rays can complicate the spectrum obtained by this type of the detector and significantly change the counting rate of single peaks. There is a theoretical model developed for coincidence summing of X and γ rays for radionuclides with complex decay scheme. This model enables to write equation for single peaks, the coincidence sum peaks and total count rate. The efficiency of detection and the activity of ^{60}Co source are the unknowns in the count rate equation system. They can be determined by simultaneously finding the roots of the equation system.

NAM.P15 (Id: 165)

SEPARATION, PRECONCENTRATION OF LANTHANUM FROM MONAZITE AND ITS DETERMINATION BY ^{138}La USING NON-DESTRUCTIVE γ RAY

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A method has been developed for separation, preconcentration and determination of lanthanum element with two isotopes, the stable ^{139}La and the radioactive ^{138}La with the half life of 1.05×10^{11} years. The stable isotope ^{139}La has a very high abundance ratio of 99.911±0.001% while ^{138}La has the abundance ratio of only 0.089±0.001%. ^{138}La also has two main γ ray energy lines, one at 788.4 and the other at 1435.8 keV which are used for the determination of ^{138}La by Hyper Pure Germanium detector, HPGe. Based on this method, the percent of ^{139}La has been calculated with high precision in synthetic samples. The results were compared with inductively coupled plasma-mass spectroscopy, ICP-MS, technique. The detection limit was calculated as MDA (minimum detection activity) and it has been found to be equal 1.46 Bq/kg for ^{138}La at 788.4 keV and 1.52 Bq/kg at 1435.8 keV γ ray energy lines with mean value 1.49 Bq/kg. Application of this method was successfully used for the determination of lanthanum in natural sample, monazite. The developed method was based on new chemical steps; precipitation and solvent extraction of crude monazite sample was used for the lanthanide determination. The precipitation method is used for removal of Th (IV) and U (VI) and the solvent extraction is used for removal of Ac from REEs. Molar solution of HDEHP in benzene was used for the extraction of REEs and actinium, Ac, from 0.1M HNO_3 while the 0.2M HNO_3 is used to stripe Ac in residual REEs in the organic phase. Therefore, lanthanum in the residual REEs was accurately determined.

NAM.P16 (Id: 183)

CHEMICAL COMPOSITION OF SILICA PHYTOLITHS. COMPARISON OF DIFFERENT ISOLATION METHODS

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The silica phytoliths, also called biogenic opals, are formed in many plant species. It has been found that information on their chemical composition is limited, except for several major elements. Comparison of different methods for isolation of silica phytoliths from plant material was performed in this work. Dry ashing and acid digestion were used and their influence on chemical composition of the prepared samples was studied. Barley (Hordeum vulgare L.) was used as a model plant and the phytoliths were extracted from different parts of the plant body - stems, leaves, awns. The analysis of the elemental composition was performed by instrumental neutron activation analysis in both short and long term irradiation modes which allowed determination of more than 30 major and trace elements.

NAM.P17 (Id: 192)

CHARACTERIZATION OF TUPIGUARANI TRADITION ARCHAEOLOGICAL CERAMICS FROM ESPIRITO SANTO, BRAZIL, APPLYING NEUTRON ACTIVATION AND MULTIVARIATE STATISTICAL ANALYSES

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The Portuguese settlers arrived in the coast of the Brazilian State of Espírito Santo in sixteenth century and built several “Missões Jesuíticas” (Jesuitical Missions). In spite of living within the Mission and been catechized, the Indians, Tupiguarani Tradition, kept on producing traditional handcraft, as the decorated ceramic, however, they introduced European elements during the production to the decoration. The archaeological interventions carried out along the coast of Espírito Santo, found the Tupiguarani Tradition vestiges, where the main evidence is a pretty ceramic with the occurrence of plastic and painted decoration. The ceramic presents high archaeological value among the vestiges. It is a material very resistant to time and to the natural conditions in the surrounding areas. Its chemical composition, once determined, can help understand the connection between the past and the present. Therefore, archaeology can fill the gap between ancient population and modern society elucidating the evidences found in archaeological sites. The main purpose of this paper was to contribute to the understanding of the Tupiguarani Tradition occupation in this region. During the research expeditions, many sherds were found in 13 archaeological sites in the study area. Inside this context, seventy ceramic fragments were analyzed by neutron activation technique, k_0-standardization method, at CDTN/CNEN using the TRIGA MARK I IPR-R1 nuclear reactor, in order to characterize their elemental composition.

The results were treated with multivariate statistic analyses - Cluster and Principal Components Analysis - and discussed.

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NAM.P18 (Id: 196)
MEASURING OF GROSS α AND β ACTIVITY BY MEANS OF LSC

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Measuring of gross α and β activity of radioactive aerosols is one of the tasks of field analyses of radioactive substances in the army of the Czech Republic. It is performed by suction of air through filters and measuring of gross activity. Furthermore the field analysis includes determination of gross α and β activity of water, milk, soil, food, smear and γ spectrometric measurement. The problem of determination of gross α and β activity of aerosols filters by means of LSC is solved. This method is perspective and appropriate for rapid measurement of gross α and β activity because of α and β separation, thus simultaneous measurement of α and β radionuclides and 4 π geometry without any filter modification. The method is based on Automatic TDCR Liquid Scintillation Counter Hidex 300 SL. Model radionuclides were chosen due to their military significance, radio toxicity and possibility of misusing by terrorists - ^{90}Sr, ^{210}Pb, ^{239}Pu and ^{241}Am. Three type of filters were investigate - paper and glass filters in LSC cocktail insoluble and soluble or partially soluble nitrocellulose filters. The attention was also paid to selection of convenient type and volume of the LSC cocktail.

NAM.P19 (Id: 198)
MODIFICATION OF PRECISE TECHNIQUE FOR DETERMINING PU MASS FRACTION BY AUTOMATIC COULOMETRIC TITRATION METHOD

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Technique for determining plutonium total mass fraction in Pu preparations and uranium-plutonium mixtures, including MOX fuel for fast reactors by the automatic coulometric titration is developed and successfully used in RIAR for a long time. The paper presents the optimization results of sample preparation and titration conditions on the facility, developed in RIAR to minimize the total analysis error. Use of standard (CO) potassium dichromate specimen without CO PuO_2 for facility calibration during analysis result verification is experimentally proved. Metrological certification of the developed technique version indicated that total analysis error of the Pu mass fraction in the specimen can be less than 0.05%. First, the developed precise technique for plutonium determination was applied during the certification of state PuO_2 standard specimen of the first class for its plutonium mass fraction content.

NAM.P20 (Id: 203)
DETERMINATION OF MAGNESIUM IN BIOLOGICAL MATERIALS BY NEUTRON ACTIVATION AND ANTI-COINCIDENCE γ-RAY SPECTROMETRY

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Magnesium is considered as an essential element at moderate levels. Both deficiency and toxic effects of Mg in humans have been reported in the literature. Titrimetry, spectrophotometry, and atomic absorption spectrometry are generally used for the measurement of Mg levels. Instrumental NAA (INAA) is also an attractive tool for the rapid, simple and reliable determination of Mg. However, due to high background activity in biological samples, the measurement of γ-rays emitted by ^{27}Mg is generally difficult. The main objective of the present work has been to study the advantages of anti-coincidence γ-ray spectrometry for the determination of low levels of Mg in biological materials using INAA. The ^{27}Mg nuclide has a half-life of 9.46 min and it emits two major γ-rays, namely 843.8 and 1014.4 keV, which are not coincident. Therefore, the use of anti-coincidence counting should not cause any reduction in peak efficiency of either of the photopeaks. The peak efficiency reduction factors of the two peaks have been measured as 0.98 and 1.00, respectively. It has been observed in many biological materials that the background around the 1014.4-keV peak of ^{27}Mg is mainly due to the 1778.9-keV peak of ^{28}Al, 1368.6-keV peak of ^{24}Na, 1642.7-keV peak of ^{38}Cl, and 1810.7-keV peak of ^{56}Mn. Anti-coincidence counting technique can be beneficially used under such situations. Several biological reference materials (RM) and standard reference materials (SRM) were analyzed for Mg by INAA. Between 200 and 700 mg of these materials were irradiated in the Dalhousie University SLOWPOKE-2 Reactor facility at a neutron flux of 5×10^{11} cm^{-2} s^{-1} for 1 min, allowed to decay for 1 min, and counted for 10 min. The anti-coincidence γ-ray spectrometer used in this work consisted of a HPGe detector and a 10"x10" NaI(Tl) guard detector with a 3"x3" NaI(Tl) plug. The peak-to-Compton plateau ratio of this system is about 590:1. The 843.8-keV peak suffers from interference by the 846.8-keV peak of ^{56}Mn which has a longer half-life of 2.58 h. In the anti-coincidence counting mode, the percentage overlap of the 843.8-keV peak is less because of the suppression of the 846.8-keV peak. We have developed a simple correction method which has been used for the determination of Mg in 15 NIST RM and SRM using the 843.8-keV γ-ray of ^{27}Mg. The measured values have been found to agree well with the certified values.

NAM.P21 (Id: 205)
ELEMENTAL CONTENT OF INDIGENOUS BACTERIA UNDER DIFFERENT CHROMIUM LOADINGS

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Indigenous bacteria can be successfully used to either detoxify or immobilize toxic heavy metals. These bacteria are under continuous investigation, and in-depth molecular understanding has been developed for some of them. However, up to date the dependence between the ability of bacteria to reduce or immobilize metals and their elemental compositions is not clear yet. For the first time epithermal neutron activation analysis method (ENAA) has been applied to determine the elemental content of bacteria before (control) and after exposure to different loadings of chromate. As a model, Cr(VI)-reducer bacteria of Arthrobacter genera, isolated from polluted basalts from both the Republic of Georgia and the USA, was used. Concentrations of the following elements: Na, Al, Cl, K, Fe, Co, Zn, As, Br, Rb, Sr, Sb, Ba, Th, U have been determined. In the cells treated with Cr(VI) significant alterations in the concentrations of some elements playing essential role in the life processes of microorganisms were observed. To understand the mechanisms of microbial resistance to Cr(VI) electron spin resonance (ESR) spectrometry was used to trace the formation and behaviour of chromium species (Cr(V)/Cr(III)) in bacterial cells. The concentrations from 12 to 19 elements were determined in the bacterial cells.

NAM.P22 (Id: 208)
COMPOSITION ANALYSIS OF ZIRCONIUM ALLOYS BY k_0-BASED PGAA USING BUDAPEST COLD NEUTRON BEAM FACILITY

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Zirconium and its alloys are important materials in nuclear power technology. Zirconium alloys, namely zircaloy 2, zircaloy 4 and Zr-Nb alloy, are mostly used for fuel cladding and pressure tubes in water-cooled nuclear power reactors due to their low neutron absorption cross sections, high corrosion resistance and good thermal conductivity. For nuclear quality control, it is important to analyze these alloys to ensure their conformity to specifications. Analytical methods, which are non-destructive in nature, bulk sample analysis capability with negligible matrix effect, are preferred for the analysis of such materials. Neutron induced prompt γ-ray activation analysis (PGAA) is one of the suitable methods. In the present work, PGAA was used for the composition analysis of three types of zircaloys, namely zircaloy 2, zircaloy 4 and Zr-2.5%Nb alloy. All the measurements were performed at the PGAA facility of the Budapest Research Reactor using cold-neutron beam. The thermal equivalent neutron flux at this position is 1.2×10^{8} cm^{-2} s^{-1}. Samples in the mass range of 0.3-0.5 g, kept inside thin Teflon bag, were irradiated in vacuum and the prompt γ-rays were measured for 3-10 h with a HPGe detector (sample-to-detector distance 23 cm) surrounded by a Compton-suppressor and passive shielding. The absolute counting efficiency of the detector was determined using ^{133}Ba and ^{152}Eu and prompt γ rays from $^{14}N(n,\gamma)^{15}N$ and $^{37}Cl(n,\gamma)^{38}Cl$. The γ-ray spectra were complex due to presence of multi-γ producing elements/isotopes and they were analyzed for peak areas by Hypermet PC (MS DOS Version 5.01). The minor elements detected in three different zircaloys, in addition to major Zr, are: Sn, Fe, Cr and Ni in zircaloy 2, Sn, Fe and Cr in zircaloy 4, and Nb, Fe and Cr in Zr-2.5% Nb alloy. The trace elements detected in general are B, Hf, Ti, Mn and Co. Trace amounts of Cl were also detected in zircaloy 2 and 4. Since all the major and minor elements were amenable to PGAA, the absolute concentrations of all the elements were determined using mass balance procedure, i.e., without using concentration of any external or internal standard. The advantages of the method are: (i) it is non-destructive, (ii) it could detect trace amounts of important elements like B, Hf and Cl in the presence of major/minor elements, and (iii) all elements determined except for B have multiple γ-rays, which help in self-validation of methodology. Acknowledgements This work was carried out under INSA-HAS international exchange program (No. IA/Misc. 2009-2010/4237) and the financial assistance by INSA and BARC (India) and HAS (Hungary) is duly acknowledged. Authors from BARC acknowledge Institute of Isotopes, Budapest for providing PGAA facility.

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NAM.P23 (Id: 214)
ANALYSIS OF LARGE AND NON-STANDARD GEOMETRY SAMPLES OF ANCIENT POTTERIES AND BRICKS BY INTERNAL MONOSTANDARD NAA USING INSITU DETECTION EFFICIENCY

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Studies of archaeological artifacts constitute an important area of research that might provide clues to unravel the past human activities, art, trade, eTc. Archaeologists are mainly interested in the provenance studies of the artifacts. Archaeological artifacts like potteries, bricks, coins and paintings are mainly studied for their provenance and one of the valid, and accepted methods is through their chemical composition analysis in conjunction with statistical methods. Among the archaeological artifacts, potteries are most widely studied and occasionally bricks are also analyzed. Since they have a strong correlation with the clay/soil source from which they are prepared, elemental concentrations could be used for arriving at correlations. Elements, which are mainly present at trace levels in the artifacts, give authentic clue to their origin compared to the major elements. Elemental concentration ratios of Al and Sc, due to their non-volatile nature and La and Ce, due to their similar geochemical properties are used for preliminary grouping. The confirmation of grouping is done from statistical analysis like principal component analysis (PCA) and cluster analysis (CA). The present paper describes application of large sample instrumental neutron activation analysis (LS-INAA) method for provenance study of potteries and bricks using scandium as internal monostandard. In the present study, samples of ancient potteries and bricks belonging to 4th-5th century BC to 4th century AD were excavated from 15 different Buddhist sites in Andhra Pradesh, India. Samples in the larger mass range of 10-50 g were taken for the experiment, irrespective of their geometry. Samples were irradiated for 7 hours in Apsara reactor, BARC. Radioactivity assay was carried out using a 40% relative efficiency HPGe detector coupled to 8k MCA. Peak areas were determined using peak-fit software PHAST. The k_0-based internal monostandard INAA (IM-INAA) was used for arriving at the elemental concentration ratios with respect to Sc. Since the method uses insitu detection efficiency, it is geometry independent and non-standard geometry samples could be analyzed. Concentrations of about 18 elements such as Na, K, Sc, Cr, Fe, Co, Ga, As, Br, La, Ce, Nd, Sm, Yb, Hf and Th were determined in both potteries and bricks. The results of elemental concentration ratios with respect to Sc show that the both potteries and bricks fall into four major groups. It was observed that in some samples of potteries and bricks, collected from same location, correlations exist indicating the source of clay/soil may be the same. We would like to highlight that the provenance / grouping study of archaeological samples could reliably be carried out through their elemental concentration ratios than the absolute concentration values and thus the present method is a suitable one for this study. Acknowledgement Authors from GITAM University acknowledge UGC-DAE Council of Scientific Research, Mumbai for the financial assistance.

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NAM.P24 (Id: 217)
DETERMINATION OF SILVER AND GOLD IN COPPER CONCENTRATE

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Many copper ores contain significant quantities of silver and gold. Copper concentrate, the intermediate in the ore processing, is an important source for production of gold and silver. Methods for the determination of noble metals have been reviewed by Beamish and Van Loon [1]. The most common method for determination of noble metals in geological sample is atomic absorption spectrometry with a fire-assay preconcentration step. One of the most sensitive method for analysis of noble elements is neutron activation analysis [2]. Instrumental neutron activation has been applied to determine noble elements in ores, but generally it is required to separate noble elements from interfering and matrix activity. Several radiochemical schemes have been reported to isolate one or more of the noble metals during analysis. These schemes include solvent extraction, distillation, ion-exchange, precipitation and others. Simultaneous determination of silver and gold in copper concentrate sample was carried out by neutron activation analysis method. Initially, copper concentrate sample was analysed by ED-XRF to obtain qualitative information and found that copper and iron were present in major proportions. Instrumental neutron activation analysis (INNA) was followed by radiochemical or chemical neutron activation analysis. During INAA, cooling time of about a week is required before ^{198}Au could be measured because of high activity due to ^{64}Cu and ^{24}Na whereas ^{110m} Ag was measured after further cooling of a week. To reduce the analysis time, radiochemical neutron activation analysis and chemical neutron activation analysis were carried out. The noble elements form anionic complex with moderate molarity of HCl that are readily retained on anion exchanger. The distribution coefficient values for gold and silver are very high at lower HCl normality whereas copper and iron have negligible values. Sodium is not retained. Hence, ion exchange separation and pre-concentration of gold and silver were carried out from 1N HCl solution. The value of gold by INAA agreed well with the values obtained by RNAA and CNAA. However, the value for silver was significantly lower in case of RNAA and CNAA method compared to INAA. This may be due to loss of silver as silver chloride. The separation procedure was modified to avoid loss of silver during separation.

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NAM.P25 (Id: 223)
RADIOANALYTICAL DETERMINATION OF ^{210}Pb AND ^{210}Po IN SEA WATER SAMPLES

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^{210}Pb and ^{210}Po are naturally occurring radionuclides belonging to the natural radioactive chain of ^{238}U. As their parent nucleus they are ubiquitously distributed in the rocks and soils of the Earth's crust and they reach the atmosphere and natural waters as a result of the decay of ^{222}Rn, which easily diffuses through the interstitial cracks of the Earth's crust itself. Human beings have always lived in contact with natural background radiation, nevertheless some particular human activities, such as the extraction and processing of the Earth's crust materials, removed these materials from their original and inaccessible location and increased the concentration of radionuclides contained in them. These activities have produced the so called TENORMs (Technologically Enhanced Naturally-Occurring Radioactive Materials), which constitute an alteration of the natural level of radiation, enhancing the risk for man to radiation exposure. Typical sources of TENORMs are phosphogypsum and waste products, coming from industrial production of fertilizers and phosphoric acid. Moreover the uncontrolled release of such wastes in the environment over time may be a source of environmental contamination, so the determination of ^{210}Pb and ^{210}Po at very low levels is particularly important, because of their major contribution to the internal dose in humans. In fact the high specific activity of ^{210}Pb (2.82×10^6 Bq mg^{-1}) and ^{210}Po (1.66×10^8 Bq mg^{-1}) makes them two of the most toxic natural nuclides. Moreover, ^{210}Pb remains in the skeleton long enough to produce the highest skeletal dose of any natural nuclide under average conditions of background exposure, and ^{210}Po is highly radiotoxic, emits high energy α particles, and is accumulated by many terrestrial plants and animals, through which it enters human dietary. Within the institutional tasks of the Radiotoxicology Laboratory of the Radiation Protection Institute of ENEA CR Saluggia, we performed measurements on several seawater and suspended particulate matter samples for the determination of the two radionuclides of interest. We simultaneously separated lead and polonium, that were firstly concentrated in water samples, through co-precipitation with iron (III) hydroxide and the addition of ammonia solution till pH 9.5 - 10, and then solubilised in HCl. For lead determination a second chemical procedure has been considered and observations about the difference between the two methods are reported. The radiometric measurement of ^{210}Pb has been performed both indirectly, through the measurement of its daughter ^{210}Bi in secular equilibrium, by the use of a proportional counter system, and directly, by the use of VLL LSC. After spontaneous deposition of ^{210}Po onto silver planchets, its activity has been measured with α spectrometry at high resolution. Some samples have been spiked with ^{209}Po for the determination of the radiochemical yield, but due to the difficulty to get polonium radiotracers, because of international events, we considered the possibility to determine it subtracting a mean blank spectrum to a mean spiked spectrum, traced with ^{210}Pb, as ancestor of ^{210}Po in equilibrium.

NAM.P26 (Id: 228)
METAL DETECTION IN SOLUTION BY NEW MASS-SPECTROMETRICAL METHOD

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New method - extraction ions from solution at atmospheric pressure - analogue of well-known ESI-MS method, was proposed for analysis of metals in radioactive solutions. Advantages of this method in comparison with known methods - (ICP-MS, ICP OES, ESI-MS, eTc.) - small volume of sample, high sensitivity, possibility of automatization.

NAM.P27 (Id: 232)
APPLICATIONS OF NUCLEAR ANALYTICAL METHODS IN THE HISTORICAL GLASS IN THAILAND

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Cultural heritage objects are highly heterogeneous. Due to the complex nature of materials and objects, their analysis needs to use extremely sensitive, spatialy resolved, multi-elemental and versatile methods that should be as a non invasive as possible and give complementary information at different scales; from the macroscopic to the nanometer scales. In this work, XRF, SEM-EDS, PIXE, EPMA and XAS were used to characterize the chemical composition and microstructure of the historical glass in Thailand such as ancient glass beads, ancient decorative glasses and old-styled gold mosaic glass. It can be concluded that these are powerful tools for investigation of heterogeneous glassy materials.

NAM.P28 (Id: 233)
GROSS α ACTIVITY DETERMINATION IN WATER AND ^{210}Po

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The gross α activity is defined as the total activity of the α emitters. Gross α and β activity screening methods have been developed for determining whether the specific analysis of radionuclides is required for characterizing the water quality. There may be a loss of radionuclides during storage of water sample caused by the adsorption onto the container wall and by the precipitation and coprecipitation. It is generally recognized, however, that these effects can be minimized by acidification of the samples after collection utilizing HNO_3 or HCl. Polonium 210 is an α emitter, a member of the uranium decay series. This radionuclide originates by radioactive decay of ^{222}Rn. In water with elevated content of ^{222}Rn, the presence of ^{210}Po can contribute significantly to the measured value of the gross α activity. The methods for gross α activity analysis of drinking water are often based on sample evaporation and heating at the temperatures exceeding 100 C. At such temperatures, can ^{210}Po become volatile in dependence of its chemical form and therefore the gross α activity can be underestimated. Our contribution will compare and discuss several routines of sample preparation considering the possible losses of ^{210}Po.

NAM.P29 (Id: 235)
ROUTINES OF ^{210}Po DETERMINATION IN FLUVIAL SEDIMENTS FOR DATING PURPOSE

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During the last decades, ^{210}Pb and its daughter ^{210}Po have become widely used radionuclides for sediment dating. In environmental sediments, the total amount of ^{210}Pb is given by two components: (a) supported ^{210}Pb, produced by radioactive decay of ^{222}Rn inside the material, and (b) an unsupported ^{210}Pb component derived from ^{222}Rn which diffuses into the atmosphere where it decays. Subsequently, ^{210}Pb is removed by atmospheric precipitation or dry deposition, falling on the land or water surfaces. The dating method often applies determination of ^{210}Po supposing the radioactive equilibrium with ^{210}Pb. Utilized analytical procedures generally consist of several basic parts: (a) isolation of measured radionuclide by leaching or total decomposition of sample matrix or polonium distillation; (b) measurement by α spectrometry or by liquid scintillation counting; (c) determination of supported ^{210}Pb. Our contribution will compare several sample preparation techniques and possibilities of measurement with regards to sample matrix, time and instrumentation requests.

NAM.P30 (Id: 237)
NEW KITS OF RADIATION SOURCES FOR THE PURPOSE OF ENERGY DEPENDENT CALIBRATION OF COLLIMATED IN-SITU-γ-SPECTROMETRY

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A very powerful tool during decommissioning of nuclear facilities is the application of collimated in-situ-γ-spectrometry [1]. Even more complicated distributions of activities of γ-emitters in walls, soils, containers or other materials can be determined. The trend is an increasing need in assessing activity concentration as well as contaminations of the surface of walls of nuclear facilities which are decommissioned. To examine the skills of this method objectively a calibration phantom, named K-RISK, was built [2]. K-RISK calibration phantom enables the creation of completely reversible three-dimensional activity distributions in wall structures close to reality. This was realized by building a concrete wall of about 2 m of height, about 2 m of length and about 0.5 m of thickness containing a grid of 256 cylindrical channels with a diameter of about 14 mm through the whole wall in which point-shaped calibration sources of important radionuclides are placed. Two kits of calibration radiation sources containing ^{60}Co have been made. The low-level-kit simulates a surface contamination below 0.5 Bq/cm^2 the high-level one those of a few Bq/cm^2 with relative uncertainties of below 8 % for the high level kit and about 30% for the low-level kit. In comparison with many other calibration phantoms K-RISK offered a variety of advantages. A first inter-comparison campaign applying K-RISK showed the expertise to fulfil high quality standards [2]. Therefore a need of further kits has been noticed containing radionuclides as ^{137}Cs and ^{152}Eu. By order of the Bavarian State Office on Environmental Affairs the URA-Laboratory has made two calibration kits of ^{152}Eu (low and high activities) and one of ^{137}Cs for the purposes of calibration collimated in-situ-γ-spectrometry. The activity values are deduced from NIST-Standard and enable to simulate values of surface contamination of the high level kits with total uncertainties of below 10% at a confidence level of 95%.

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NAM.P31 (Id: 238)
RADIOCHEMICAL TRACE ANALYTICAL PURIFICATION PROCEDURES DURING DECOMMISSIONING OF NUCLEAR FACILITIES: EXPERIENCES AND RESULTS

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The decommissioning of nuclear facilities demands an optimisation of analytical procedures to determine a variety of radionuclides in a variety of different materials on a wide activity scale ranging from very low to very high activities. Severe analytical problems arise from disadvantageous decay properties of a number of radionuclides. The results of recently developed analytical and nuclear detection methods are presented which have been applied successfully for determining radionuclides like ^{55}Fe, ^{60}Co, ^{59}Ni, ^{63}Ni, ^{90}Sr, U, Pu, Am, Cm [1-3]. The applied analytical methods have to fulfil high quality standards including very low lower limits of detection, very high selectivity, sufficient precision and accuracy even in samples containing enhanced amounts of interfering elements. Results are presented analysing different types of materials [1]. Corrosion products of several types of steel and alloys have been analysed to support the radiological characterisation of nuclear power plants during decommissioning. At present new challenges arise analysing Pu and Am in concentrated solutions of salt. Some aspects of radiochemical purification procedures have to be adapted to this kind of matrices to realise lower limits of detection of about 0.001 Bq/L for ^{239}Pu, ^{238}Pu and of about 0.005 Bq/L for ^{241}Am.

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NAM.P32 (Id: 244)
DEVELOPMENT OF k_0-CYCLIC NEUTRON ACTIVATION ANALYSIS AT THE PORTUGUESE RESEARCH REACTOR

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The fast pneumatic transfer system (SIPRA) for short irradiations at the Portuguese research reactor has been upgraded with a new spectrometer incorporating Zero-Dead Time corrections and new software for use alongside a modern computer. The development of cyclic NAA based on the k_0 methodology for SIPRA has been performed. To ensure the accuracy of the system for very short lived nuclides (e.g. ^{77m}Se and ^{20}F), the calibration of transit time with millisecond timing resolution was performed. The results of k_0-cyclic NAA for standard reference materials have been shown with a particular focus on selenium.

NAM.P33 (Id: 247)
ASSESSMENT OF CONTAMINATION LEVELS AND DISPERSION IN A MINING IMPACTED AREA BY INAA AND μ-PIXE ANALYSIS

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Past mining activities, random disposal without proper precautionary and rehabilitation measures, erosion, leaching, and atmospheric dispersal of mine soils, tailings and waste may contaminate the surrounding environment and nearby locality with highly toxic trace elements, and natural radionuclides in the vicinity of Sao Domingos mine, Portugal, a copper-sulphide open pit mine. Toxic trace elements, rare earth elements and natural radionuclides were measured in mining soils, plants, lichen and moss samples by using instrumental neutron activation analysis (INAA) and μ-PIXE. This study has carried out the assessment of magnitude of contamination levels and dispersion of the above contaminants in and around the mining area. The levels of the elements were very high in all types of samples including the soils, lichens, moss and plant: the highest levels were recorded in the samples collected close to ore processing sites and decreased with distance. The concentrations of As, Sb and other elements in the soils, plant, lichen and moss samples were much higher than the regional background levels reflecting the high magnitude of contamination in and around the mining area and the possible health risk for the inhabitants living nearby the mining affected area. Patterns of bioaccumulation of elements throughout the area under study were quite similar for widespread pollutants such as As, Sb, Cu, Zn, Fe, Br, Co, La, and Cr. The lichen, moss and plant samples are important biomonitors of terrestrial and atmospheric pollution in mining affected areas. Some of the plant species with high biomass and extensive root systems, grown abundantly and spontaneously in the area, are suggested as the suitable candidate for the phytoremediation of the mining contaminated soils.

NAM.P34 (Id: 257)

k_0-INAA USING COMPARATOR AND NEUTRON FLUX MONITOR AT CDTN/CNEN, BRAZIL: ADVANTAGES AND DISADVANTAGES

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Many laboratories apply the k_0-INAA and several of them use comparators and spread sheet instead of neutron flux monitors and software for analysis and calculations. At the Laboratory for Neutron Activation Analysis, CDTN/CNEN, two variations of the k_0-INAA are used, one is called in house k_0-"monostandard" method - using comparator - and the other is the well known k_0-standardization method - with neutron flux monitor. The first one, k_0-"monostandard", is an alternative method and it is used in some specific situations. This method is carried out with sodium as comparator, the values for f and α are average values for the carousel facility of the TRIGA MARK I IPR-R1 reactor and each sample is irradiated in one irradiation channel. The calculations are carried out with spread sheet. Concerning the k_0-standardisation method, irradiation of the samples pile in the irradiation vial and intercalated by neutron flux monitor Al-Au (0.1%) IRMM-530RA foil cut into 6 mm diameter and 0.1 mm thick is applied. The irradiation is performed in one irradiation channel in which the values for f and α were determined in this specific channel. For the calculation of the elemental concentrations, a software package called Kayzero for Windows is used. In this paper, advantages and disadvantages of both procedures are discussed basing on the results of the reference material GBW 07401 (soil) analysed by both procedures.

NAM.P35 (Id: 265)

AIRBORNE AND TERRAIN γ SPECTROMETRY MONITORING OF NATURAL AND ARTIFICIAL RADIOACTIVITY

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Airborne and terrain γ-ray spectrometry are very effective methods for radiation control in environment, prospecting of radioactive contamination and localization of radioactive sources. These methods were proved in the system of the military radiation reconnaissance and radiation monitoring of the Czech Armed Forces. The IRIS-XP airborne γ-ray spectrometer (4x4 liters NaI(Tl) crystals) and terrain γ-ray spectrometer PGIS-128 (2 liters NaI(Tl) crystal), both produced by PICO Envirotec, Inc. Canada, were used for in-situ experiments. The helicopter Mi-17 was used for airborne measurement and the mobile devices and footpath variant were utilized for measurement on terrain surface. The semi-calibration of described systems was performed by comparison with semiconductor HPGe in-situ γ-ray spectrometry on selected terrain locality. The spectra of natural radionuclides, such as ^{40}K, ^{232}Th and ^{238}U, were measured for these purposes. The radiation sources ^{137}Cs and ^{60}Co with activity from 100 MBq up to 3 GBq were used for measurement optimization and simulation of radiation events. The terrain γ-ray spectrometry was also tested under the condition of real contamination in Chernobyl "exclusion zone".

NAM.P36 (Id: 268)

ISOTOPIC CHARACTERIZATION BY Q-ICP-MS OF SOME α EMITTERS IN CUBAN SOIL

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The determination of α emitters, namely uranium, thorium and plutonium isotopes, in the soils, is a useful way to evaluate the radioactive fallout and to better understand the isotopic distribution profiles in different soil layers. Due to the particular geographic position and the economical and political conditions, Cuba could be considered as a unique "natural habitat" in the boreal hemisphere. The total lack of human activities having as a main consequence or as a side effect the production of artificial radionuclides, with the only exception of the temporary installation of strategic missiles and the patrolling of the territorial water by some nuclear ships, makes the island territory free from any radiological human contamination. As the Cuba Island has never been contaminated by artificial human activities employing α emitters, these soils could be specifically considered as a reference soil samples for the Central American zone, for the determination of the radiological contamination due to a fallout consequent to performing air experiments, testing weapons or nuclear power plants accidents. It is well known that the analytical results, especially for the determination of some actinides, are strongly influenced by some characteristics of the samples. Inhomogeneity, broad distribution of the particles and very low concentration effects were considered. For these reasons, a monitoring campaign regarding the determination of amount of radionuclides such as thorium, uranium, radium and plutonium in different environmental matrices is very interesting. However the most commonly used analytical methods (α-spectrometry, neutron activation analysis) are too laborious, too expensive and time consuming to enable an accurate characterization of the territory and a constant monitoring of the radioactive levels in the environment. To overcome these disadvantages, a new analytical approach was used to develop a rapid, simple and reasonably cheap method, which can be exported towards other environmental laboratories. This method was tested by comparing the results obtained in different laboratories using ICP-MS and different nuclear measurement techniques, such as γ and α spectrometry.

NAM.P37 (Id: 269)
APPLICATION OF MONTE CARLO SIMULATION TO DESIGN A MODULAR ^{241}Am-Be NEUTRON IRRADIATOR

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Neutron irradiator facilities with ^{241}Am-Be sources are worldwide available in order to perform neutron activation analysis (NAA), to investigate materials in different research areas or to test and calibrate neutron detectors and environmental or personal dosemeters. The use of a neutron irradiator is advantageous because have a very stable neutron flux, though it is many orders of magnitude lower than the one of a nuclear reactor or a particle accelerator. Many of irradiators are realized with neutron sources located at fixed positions and accordingly the characteristics of neutron spectrum previously assessed do not change. However, an interesting chance is to have a modular facility, capable of varying conditions of irradiation and setting the prevalence of fast or thermal neutron spectrum component. In this work we report the Monte Carlo studies devoted to design a modular ^{241}Am-Be neutron irradiator making use of different moderators (water, graphite, polyethylene and so on) in order to obtain a neutron energy distribution useful to test neutron detectors or personal dosemeters. The optimization of thicknesses and shape of the moderators and shielding materials was obtained with a Monte Carlo simulation with MCNP5 code. An experimental test to verify the reliability of the simulation by means of NAA of selected materials was also performed. Once realized, the designed neutron irradiator will provide an useful facility for radiochemical material studying and testing and calibration of personal dosemeters or neutron measurement equipments.

NAM.P38 (Id: 278)
DEVELOPMENT OF THE METHOD FOR DETERMINATION OF TECHNETIUM IN ENVIRONMENTAL AND BIOLOGICAL SAMPLES

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Technetium is a one of the uranium fission products and therefore could be found in a trace concentration in the Earth's crust. It was estimated that a kilogram of uranium contains 1 nanogram (10^{-9} g) of technetium. However, besides of naturally occurred Tc, in the last several decades its relatively high amount was artificially produced in the nuclear reactors by the thermal neutron fission of ^{235}U and ^{239}Pu reactor fuels with the fission yield product of ^{99}Tc of about 6.1%. The ^{99}Tc has a half-life of $2.13\text{x}10^{5}$ a, giving a specific activity of 650 MBq g^{-1} and decays by β emission with E_{max} 300 keV. It has been released into the environment as the results of nuclear weapons testing and low- and intermediate-level waste disposal, and will be an important component of high level wastes when these are finally disposed of. Geochemically, Tc exists in two stable oxidation states depending on the redox conditions; it forms reduced species, predominantly Tc^{4+} and its complexes and compounds, however, it could also occur as Tc (VII), which forms the pertechnetate anion, $[TcO_4^-]$. The environmental redox chemistry of technetium is thus very important with respect to its mobility. As a β-emitter the ^{99}Tc is determined with commonly used low-level β-counters. However, due to the often complicity of the analyzed samples (e.g. high concentrated salt samples) a separation procedure prior to the counting should be applied. In such case an internal standard should be used in order to find out a chemical yield of the overall analyzing method. For the determination of recovery of ^{99}Tc the short-lived isotope ^{99m}Tc (half-life: 6 hours) is usually used which is sometimes unacceptable due to the rare availability, relatively high specific activity and low half-time and high costs of metastable ^{99m}Tc. In the present study we propose a new methodology for the assessment of the overall recovery of ^{99}Tc determination by combining the conventional β-counting method with relatively fast inductively coupled plasma mass spectrometry (ICP-MS) measurements. The principle of the developed method is to use the naturally occurred rhenium (as a homolog element to technetium) as an internal standard in order to find out the losses of the analyte during the sample preparation procedure. The determination of rhenium was performed by ICP-MS method and its recovery values were used for the correction of results for ^{99}Tc measurements. The accuracies of Tc and Re recoveries observed by application of such approach were found to be in the range of 95-105%. After the recovery studies the developed procedure was applied for analysis of Tc activity in available standard solutions as well as in real samples: environmental (high concentrated salt samples) and biological (urine) samples. The results showed a good agreement of measured technetium content with the predictable values and will be discussed during the presentation in details.

NAM.P39 (Id: 298)

DEVELOPMENT OF RADIOACTIVE XENON ANALYSIS SYSTEM BY DIAGNOSTIC ASSESSMENT USING MASS SPECTROMETRY

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In order to diagnose each function of currently adopted xenon acquisition system (Gammadata, SAUNA II), two kinds of preliminary test have been carried out. First of all, the mole fraction of the air concentrated in the transport columns adopted for the SAUNA was measured by utilizing mass spectrometer. It has been verified that the terrestrial xenon was well concentrated to be 0.2% of mole fraction from the trace amount of 0.087 ppm of the atmosphere and that also the moisture as well as carbon dioxide contained in the air was removed efficiently. The SAUNA II is designed to adsorb xenon physically into the molecular sieve (5A) in the transport column at room temperature, although the adsorption efficiency of xenon is enhanced under the cooling system. For this matter, subsequently, the adsorption efficiencies at room temperature were compared with that at -196 °C, measuring the Xe concentrations in the stainless steel frit packed with 5A by mass spectrometry. We experimentally demonstrated the adsorption efficiency was two times higher at the lower temperature. Based on the preliminary test, we propose an innovative structure for xenon acquisition system which has a cooling bath equipped to the transport columns and a portable mass spectrometer for real-time diagnosis. Xe transferred from the transport column is highly enriched in the tiny sample reservoir to be mounted on the high-purity germanium (HPGe) well detector.

NAM.P40 (Id: 311)

RAPID AND SIMULTANEOUS DETERMINATION OF NEPTUNIUM AND PLUTONIUM IN ENVIRONMENTAL SAMPLES BY EXTRACTION CHROMATOGRAPHY USING SEQUENTIAL INJECTION AND ICP-MS

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An automated analytical method for the rapid and simultaneous determination of plutonium and neptunium in environmental samples was developed. Extraction chromatographic column packed with Eichrom TEVA® resin was incorporated in a sequential injection (SI) system to automatically separate plutonium and neptunium from matrix elements and interfering radionuclides. Since valence adjustment is a crucial step to warrant the same chemical behavior of plutonium and neptunium on TEVA column, and therefore actualize their simultaneous separation and detection, we investigated and compared distinct methods for the valence adjustment of plutonium and neptunium to Pu(IV) and Np(IV). The results showed that two steps valence adjustment using disulfite and concentrated nitric acid as redox reagents was the most effective method. The analytical results for both plutonium and neptunium in three reference materials were in agreement with the reference or informative values at the 0.05 significance level. The column separation time within the SI system for a single sample was less than 1.5 hours. The developed method significantly improves analysis efficiency and reduces labor intensity, and enables a rapid and simultaneous determination of plutonium and neptunium which is especially important in emergency situations.

NAM.P41 (Id: 317)

A PRELIMINARY ASSESSMENT OF AIR QUALITY IN MANGABEIRAS' PARK, BELO HORIZONTE, BRAZIL, USING EPIPHYTIC LICHENS AS BIOMONITOR

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Lichen, or lichenized fungi, is one of the most efficient on air pollution biomonitoring among the biomonitors. Biomonitoring has been used as an alternative method to study the air pollution in several countries, however, in Brazil, systematic use of lichens as biomonitors of environmental pollution is quite rare. In order to make an assessment of the air quality of the Mangabeiras' Park, this study was conducted by measuring the concentration of elements accumulated in the lichen thallus. This park, located in Belo Horizonte (Minas Gerais, Brazil), is the greatest green area in the city, and an apparent region of non polluted air. During the development of the study, epiphytic lichens of three genus of urban lichens (Canoparmelia sp, Pyxine sp and Parmotrema sp) were collected using a steel stainless knife, taking samples of similar sizes from 1.5 m from the soil. Soil samples were also collected in order to calculate the enrichment factor. The elemental concentration determination was carried out applying the neutron activation technique, k_0-standardization method, using the TRIGA MARK I IPR-R1 research reactor located at CDTN/CNEN. The lichen samples presented expressive concentrations of Br, Fe, K, Mn, Na and Zn. Using Sc as normalizing reference crustal element; the enrichment factor (EF) values of 19 elements were calculated. Among these 19 elements, 14 of them (Al, As, Co, Fe, Hf, K, La, Mn, Na, Sb, Th, U, W and V) present EF values equal or lower than 1. The presence of characteristic soil elements from the region (As, Fe, Th, U) and the low values of EFs evidence that soil elements are expressive in the air composition of this park. The presence of iron mining companies in the surroundings of the Mangabeira's Park seems to interfere in the air quality of this area. Therefore, this study shows one of the most known impacts of mining activities: the emission of particulate matter.

NAM.P42 (Id: 323)
CHARACTERIZATION OF ABANDONED CONTAMINATED VALONGO MINING AREA AT PORTUGAL, BY INAA

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At Sao Domingos mining (southern Portugal near Spanish border), we have collected soils and vascular plants growing there [1-3]. The values we found for arsenic in soils were between 400 and 3600 mg/kg. All the plants we could find growing there were: Agrostis castellana Boiss. & Reuter; Corrigiola litoralis L.; Erica andevalensis Cabezudo & Rivera; Erica australis L.; Eucalyptus camaldulensis Dehnh.; Genista polyanthos R. Roem. ex Willk.; Juncus acutus L.; Nerium oleander L.; Nicotiana glauca R.C. Graham; Piptatherum miliaceum (L.) Coss.; Rumex scutatus L. subsp. Induratus. The maximum values we have found for arsenic transfer from soil to these vascular plants was 0.3%, which is not sufficient to bioremediation purposes, i.e., seeding one of the plants in the arsenic contaminated area would not results in a large amount of arsenic removed from the soil. Therefore, we went to other mine (Valongo mine, northern Portugal, near Oporto) and we collected again soil and vascular plants which existed there. Soil results pointed out arsenic values up to 0.6%. The vascular plants which we found there are: Agrostis curtisii Kerguélen; Agrostis fouilladei P. Fourn.; Amanita caesarea Scop.; Digitalis purpurea L.; Erica australis L. subsp. aragonensis (Willk.) Cout.; Erica cinerea L.; Juncus conglomeratus L.; Pteridium aquilinum (L.) Kuhn; Pterospartum tridentatum (L.) Willk. subsp. cantabricum (Spach) Talavera & P.E.; Ulex minor Roth. We also collected some moss samples and water from the wells and small river of the area, to compare with previous water sampling at Sao Domingos mine, where we found arsenic values between 0.006 and 1.78 mg/L. The aim of this work it to determine arsenic (and other chemical elements) in the mosses and vascular plants collected at Valongo mine, by neutron activation analysis, in order to determine the transfer coefficients between soil and mosses or vascular plants and eventually find the suitable biorremediator for Sao Domingos mine.

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NAM.P43 (Id: 329)
TRANSFER COEFFICIENTS FROM SOIL TO ROOTS OF WHEAT PLANTS BY INAA

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An extensive investigation of elemental levels in cereals and their cultivation soils is being carried out across the main production areas of mainland Portugal, with a view to an eventual Se-biofortification of major cultivars (under research contract PTDC/QUI/65618/2006; FCT-MCTES, Portugal). Breads and cereal derivatives (breakfast blends, pastas, eTc) make up a sizeable share of the Portuguese diets, thus an increase in the bioavailability of essential elements through crop biofortification may contribute to an upgrade in the health status of the whole population. For this purpose of nutrient supplementation, and accounting for its relative weight in the realm of cereal consumption, samples from two areas with an intensive wheat production (bread and durum; Triticum aestivum L. and Triticum durum L., respectively) were collected through the summer of 2009. Such areas are located north and south of the Tagus river, deep into the inner territory of mainland Portugal. This work aims at an elemental characterisation of the cereal-cultivation soils, and their role in the nutritional uptake of wheat plants. Four certified reference materials were used for the quality control of the analytical technique here (instrumental neutron activation analysis = INAA): NIST-SRM 1567a (Wheat Flour), NIST-SRM 1568a (Rice Flour), GBW07404 (NRCCRM Soil NCS DC 73322) and GBW07406 (NRCCRM Soil NCS DC 73324). For NIST-SRMs, only a few elements were above detection limit in the present conditions - As, Br, K, Na, Rb, Se, Zn - even if in good agreement with their certified values, accounting for uncertainties. The elements that could be compared with certified values for the Chinese soils were: As, Br, Ce, Co, Cr, Cs, Cu, Eu, Fe, Ga, Hf, Ho, K, La, Lu, Na, Nd, Pr, Rb, Sb, Sc, Sm, Ta, Tb, Th, W, Yb, Zn and Zr. Overall, a good agreement was found as well. Wheat-cultivation soils from the area south of Tagus river contain higher concentrations of As, Cs, Ga, K, Rb, Ta, Th, U, W and Zn, thus featuring adequate levels of nutrients such as Cs, K, Rb or Zn. The northern-area soils contain higher concentrations of Co, Cr, Eu, Fe, Hf, Na, Sb, Sc, Tb, Yb and Zr, and feature large amounts of the nutrients Co, Cr, Fe and Na. Similar results were obtained for Br and the rare earths Ce, Ho, La, Nd and Sm. Some differentiation of rare-earth elements occur for Eu, Tb and Yb, i.e., for the heavier ones. No depletion or enrichment was found for Ce, since its ratio to La is 2. For wheat roots, Cd and Hg were found in some samples at levels of 1 and 0.1 mg per kg dry weight (d.w.), respectively. The highest soil-plant transfer coefficients (root d.w. contents/soil d.w. contents) were found for Br, Cr and Zn (above 30 %), followed by As, Co, Ga, K, Tb, U and W (between 10 and 20 %), whereas the lowest values (below 10 %) were found for Ce, Cs, Eu, Fe, Hf, Ho, La, Na, Nd, Rb, Sc, Sm, Ta, Th, Yb and Zr.

NAM.P44 (Id: 334)

DETERMINATION OF SELENIUM IN SOFT AND DURUM WHEAT (PLANT AND GRAIN) USING THE SHORT-LIVED NUCLIDE

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The increasing attention paid to the role of selenium (Se) and selenoproteins in human health stems from an equally growing body of evidence on their actual (general) importance for a healthy immune system, and on their protective (specific) effects against cardiovascular disease, asthma, male sterility, and, especially, certain forms of cancer. The current Recommended Dietary Allowance (RDA) for adult men and women (> 14 y) is 55 µg of the element per day. The Portuguese situation is hard to assess due to scarce information and lack of consistent studies on this subject, though unlikely to differ much from Europe at large, where falls in Se intake - and corresponding drops in blood indicators of Se status - have raised a widespread concern. In these terms, an extensive investigation of Se levels in cereals and their cultivation soils is being carried out across the main production areas of mainland Portugal, with a view to an eventual Se-biofortification of major cultivars (under research contract PTDC/QUI/65618/2006; FCT-MCTES, Portugal). Total Se is assessed by instrumental neutron activation analysis (INAA), through short irradiations on the fast pneumatic system (SIPRA) of the Portuguese Research Reactor (RPI-ITN), with an acquisition system by ORTEC - a DSPEC Pro digital γ-ray spectrometer and a liquid-nitrogen-cooled, high-purity-germanium detector (HPGe), connected to a 4096 multi-channel analyser. The short-lived nuclide ^{77m}Se, that features a half-life time of 17.5 s, is used to determine the Se content in SIPRA. Full optimisation of the procedure for Se determination is still ongoing. Still, several tests on the SIPRA with the reference materials NIST-SRM 1567a (Wheat Flour) and NIST-SRM 1568a (Rice Flour) have been yielding results that are quite satisfactory already. Reference samples have been put through cyclic neutron activation analysis (CNAA), at a flux of 2.6×10^{16} neutrons per square meter and second. The number of cycles for each sample was 15, with both irradiation and counting times of 20 s, and a decay time of 5 s. Three replicate samples of 80-110 mg (in polyethylene capsules) have been used for each reference material; concentrations were determined by the relative method. This paper is focused on wheat samples - soft (bread) and durum wheat; Triticum aestivum L. and Triticum durum L., respectively - from the 2009 campaign, and discusses the Se distribution in grains and other parts of wheat plants (roots, leaves), toward an eventual supplementation of the element in different growth stages. A few other experiments with the Chinese soil standards GBW07404 (NRCCRM Soil NCS DC 73322) and GBW07406 (NRCCRM Soil NCS DC 73324), as well as with rye roots and soils from rye fields, were not that conclusive due to a very high activity of the samples. Such tests will be repeated with the DSPEC Pro in lieu of the DSPEC jr, since the latter does not properly account for counting losses.

NAM.P45 (Id: 341)

DETERMINATION OF ^{238,239,240,241}Pu, ^{241}Am, ^{242,243,244}Cm, ^{90}Sr, ^{55}Fe AND ^{63}Ni IN LOW AND INTERMEDIATE LEVEL OPERATIONAL RADWASTE

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Radioanalytical method for the determination of radionuclides so called “difficult to determine” has been developed to characterize liquid and solid operational low and intermediate level radioactive waste. The main steps of the method involve digestion of sample of various matrixes, primary separation of radionuclides from matrix with the aim to reduce high γ activities and final purification of radionuclides using extraction chromatography. The commercially available Eichrom resins (UTEVA, TEVA, TRU, Sr Resin and Ni Resin) have been applied. The method enables the simultaneous determination of ^{238}Pu, ^{239,240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm, ^{243,244}Cm, ^{90}Sr, ^{55}Fe and ^{63}Ni from a single sample. Radionuclides were measured by means ofααα spectrometry and liquid scintillation. After measurements of ^{238}Pu and ^{239,240}Pu, plutonium isotopes were removed from the stain steel disc and following additional purification ^{241}Pu was measured using liquid scintillation. The method was applied for determination of radionuclides in samples of different matrixes derived from Ignalina NPP e.g. reactor water, spent fuel pool water, evaporated concentrate, spent resins, dust, graphite eTc. Accuracy and precision of Pu, Am and Cm analysis were tested in intercomparison runs organized α by the Risø National Laboratory, Denmark and in proficiency test organized by National Physical Laboratory, UK. The authors would like to acknowledge the financial support of European Commission (European Atomic Energy Community [EURATOM]) for funding project 211333 in the Seventh Framework programme.

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NAM.P46 (Id: 344)

DETERMINATION OF THE IMPURITIES OF REACTOR CORE GRAPHITE WITH NEUTRON ACTIVATION ANALYSIS, X-RAY FLUORESCENCE AND MASS SPECTROMETRY TECHNIQUES FOR GRAPHITE WASTE MODELING

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In nuclear energetics is very important to handle radioactive waste properly. After closing nuclear facility significant amounts of radioactive waste will occur, which disposal will depend on their activity. The main source of radioactivity in the NPP's equipment are the neutron activation products. Their amounts depend on the elemental composition of materials used for building a nuclear facility. In the cores of channel and gas cooled reactors huge amounts of high purity graphite is used as a moderator. Because being in an intensive neutron flux for a long irradiation period minor graphite impurities such as Cs, Sr, Eu, U, Cd, U, Th play the major role in the formation of medium and long-lived radioactive nuclides. As graphite is difficult to dissolve, advanced measurement techniques must be used. In this work measurement methods of neutron activation analysis, X-ray fluorescence and mass spectrometry for the evaluation of minor graphite impurities will be presented and their results critically compared. We show a good agreement between results of an expensive neutron activation analysis and rapid mass spectrometric measurements. We have found that minor graphite impurities of interest are in 10 ppm - 10 ppt concentrations range. These results will be used for the Ignalina NPP graphite decommissioning.

NAM.P47 (Id: 346)

TRITIUM CONTENT DISTRIBUTION IN JEJU ISLAND GROUNDWATER USING Ni-Ni ELECTROLYTIC ENRICHMENT METHOD

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Jeju is a volcanic island located about 90 km south of the Korean peninsula. This area is covered with highly permeable basaltic rocks from the Pliocene through the Quaternary. These rocks are highly permeable and forming the principal aquifers. Therefore, ground water is the sole fresh water resource and provides almost all of the water demand because of the low content of the surface water. In this work, tritium levels in Jeju island groundwater were studied with a liquid scintillation counter(LSC) and electrolytic enrichment method using Ni-Ni electrodes. Tritium enrichment parameters are estimated with different current and total current charge variation. From the established optimum tritium enrichment condition, we analyzed fifty eight mountainous area groundwater samples of Jeju island. The tritium separation factor was from 8 to 36 with a current density variation. The detection limit of tritium measurement is about 0.06 Bq/L using 1000 mL sample and 600 min counting time. The tritium concentrations in fifty eight groundwater in Jeju island were ranged <0.06 TU-0.46 Bq/L and averaged value was 0.25 Bq/L.

NAM.P48 (Id: 351)

QUANTITATIVE EVALUATION OF ESSENTIAL AND TRACE ELEMENTS IN NINE MEDICINAL PLANTS BY IPAA AND XRF TECHNIQUES

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The concentrations of some trace and essential element constituents in nine medicinal plants surveyed in Bucegi Mountains in Romania, namely Achillea millefolium, Chelidonium majus, Cynara scolymus, Hypericum perforatum, Tilia cordata, Matricaria recutita, Mentha, Rosa canina and Urtica, were determined. The above medicinal plant species were used for the experiments as they are an important source of drug with many pharmaceutical effects as well as bioaccumulators of the trace heavy metals and other bioactive chemicals. Samples of about 2 g have been measured by the combined use of photon neutron activation analysis (IPAA) followed by high-resolution γ-ray spectrometry and X-ray fluorescence (XRF). The analytical approach allowed the determination of 34 elements in the 9 medicinal plants commonly used in Romania. Usually the trace element concentrations were higher in tree leaves than in herbs, following the order Tilia cordata > Chelidonium majus > Cynara scolymus > Hypericum perforatum > Achillea millefolium > Mentha,> Urtica > Matricaria recutita, especially for Mn, Fe, Cu, and Rb. Most elements vary in a narrow range by a factor of 2 - 4 while a few others as Na, Mn, and V, vary in a wide range. All the medicinal herbs contain K, Cl, Mg, P, and Ca as minor constituents along with significant trace amounts of Cr, Cu, Fe, Mn, Se, and V. The detection limits of the analytical method for different elements are discussed in some detail. The interrelationships of the trace metal concentrations of medicinal herbs by factor correlations between pairs of elements have been examined.

NAM.P49 (Id: 352)
TRACE ELEMENT DISTRIBUTION IN HUMAN TEETH BY X-RAY FLUORESCENCE AND MULTIVARIATE ANALYSIS

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X-ray fluorescence spectrometry (XRFS) was used as a multielement method of evaluation of individual whole human tooth or tooth tissues for their amounts of trace elements. Measurements were carried out on human enamel, dentine, and dental cementum, and significant differences in tooth matrix composition were noted. In addition, the elemental concentrations determined in teeth from subjects of different ages, nutritional states, professions and gender, living under various environmental conditions and dietary habits, were included in a comparison by multivariate statistical analysis (MVSA) methods. By factor analysis it was established that the trace inorganic components of human teeth varied consistently with their source in the tissue, with more in such tissue from females than in that from males, and more in tooth incisor than in tooth molar. The heavy metal concentrations in the tissue did not varied greatly from dentin to enamel or to dental cementum, whatever the age and occupation of the subject. The statistical analyses performed seem to indicate that deciduous teeth might be a suitable indicator of environmental exposure to several trace elements. The results demonstrated that XRFS supplemented by MVSA is an useful and practical approach for the investigation of trace heavy metal incorporation and distribution on the surface of teeth as well as in inner layers. The features of the approach applied to environmental monitoring are also discussed.

NAM.P50 (Id: 353)
TRACE ELEMENT DISTRIBUTION IN CROPS GROWN UNDER INDUSTRIAL STRESS: A MULTIDISCIPLINARY APPROACH

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The impacts of the trace heavy metals released by different industries on croplands were monitored by different environmental media. All the sampling locations based on a regular monitoring grid network were selected. To all samples collected in area of the smelter complex (20 x 20 km^2), one to three locations has to be contributing to each sampling location. The spots were further categorized by groups in relation with the downwind distance from the smelter complex. In the developed approach, trace element concentrations in soil, water, epiphytic mosses and crops were determined. The analyses were carried out on the vegetation pieces and grain fractions < 2 mm by using the photon activation analysis and X-ray fluorescence methods. Some of the experimental trace metal values exceeded the threshold established by Romanian and EU regulations to protect vegetation and explain the estimated significant crop losses. Factor analysis of the elemental data showed always the component loaded with specific elements coming from industrial emissions. For example, the survey showed that in the monitored area bordering the oil refinery complex there are zones with significantly elevated concentrations of Ni, Pb and V and other heavy metals as As, Cr, Cu, Fe, Sb and Zn linked to anthropogenic activities in the oil and hard industry.

NAM.P51 (Id: 174)
OPTIMISATION OF NEUTRON FLUX PARAMETERS DETERMINATION FOR k_0 STANDARDIZATION DURING IRRADIATION AT REACTOR LVR-15 IN ŘEŽ

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Multipurpose research reactors such as LVR-15 in Řež require frequent monitoring of neutron flux parameters (f, α) when k_0 standardization in NAA is to be used. These parameters may change quite unpredictably, because experiments in channels adjacent to those used for NAA frequently require a change of the reactor operation parameters and/or active core configuration. For monitoring the neutron flux parameters in each irradiation container the bare triple monitor method is very convenient. However, when using Zr-Au monitors we have observed significant variations of f and α values as a function of irradiation and decay time employed. This was especially noticeable if the irradiation time was very short (in case of short-term irradiation induced activities of monitors are too low) or if the decay time before the first count of these monitors was longer than the recommended decay time of one day (the long-term irradiated samples may be too hot to be handled and/or measured one day after irradiation). Therefore, we have tested other sets of neutron flux monitors consisting of Au, Mo and Cr (Au, Mo and Sc in case of short-term irradiation) as proposed recently by Koster-Ammerlaan et al. [1]

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NAM.P52 (Id: 292)
^{129}I IN FINNISH WATERS

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^{129}I is a long-lived β-emitting (E_{max} 154.4 keV) radioisotope of iodine. Its half-life is 15.7 million years. ^{129}I is produced mainly by human nuclear activities and especially it has been released to the environment from the spent nuclear fuel reprocessing plants. In the pre-nuclear era ^{129}I/^{127}I ratios in the environment were approximately 10^{-12}. Nowadays ^{129}I/^{127}I ratios have reached values from 10^{-10} to 10^{-4}. In this study, activity concentrations of ^{129}I and its distribution into various chemical species (iodide I^-, iodate IO_3^- and bound in organics) were analyzed from four different lakes in Finland and from four different sea locations on the Gulf of Finland, the Bothnian Sea and the Bothnian Bay. ^{129}I was also analyzed from four rainwater samples. Samples were taken in the summer of 2009. After filtering the 0.3 L water samples, separation of various iodine species was done by anion exchange chromatography: $^{129}IO_3^-$ passes through an anion exchange resin bed in NO_3^- form while $^{129}I^-$ absorbs into the bed. $^{129}I^-$ is eluted from resin with NaClO. Finally samples were precipitated by $AgNO_3$ to form AgI and ^{129}I was measured by accelerator mass spectrometry (AMS). Stable iodine (^{127}I) was analyzed by inductively coupled plasma mass spectrometry (ICP-MS). First results from a lake in the southern Finland and from water taken from the Finnish Bay in front of Helsinki show that levels of ^{129}I in lake water are around 1×10^{9} atoms per litre while in sea water the levels are 4–5 times higher. ^{129}I occurs both in lake and sea water mainly in iodide form and the fraction of iodate form is only about 5%. The ^{129}I/^{127}I ratio has clearly elevated compared to natural levels, and are approximately the same in sea and in lake, 14×10^{-8} and 8×10^{-8}, respectively. These results are only preliminary and a better picture of the situation will be obtained after finalizing the project. The results obtained so far are, however, at the same level as obtained in Swedish studies at the same latitudes.

NAM.P53 (Id: 371)
^{236}U IN WELL WATER - A TOOL FOR URANIUM PROSPECTION?

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^{236}U (half-life 23 Myr) is produced in uranium ore via thermal neutron capture on ^{235}U. The neutrons originate mainly from (α,n) reactions caused by α-particles from the uranium decay series. The equilibrium ratio of ^{236}U/U in natural ore is proportional to the thermal neutron flux, which is expected to be proportional to the uranium concentration in first approximation. Since this fingerprint of high grade ore should stay unaltered in withering and dissolution, it should still be detectable in well water which was in subsurface contact with the ore; thus, such wells should be useful as natural probes for uranium prospection. We expect this signature to be more unambiguous than the uranium concentration in water, recently investigated e.g. in [1]. However, measurement capabilities for ^{236}U were developed recently only at very few AMS (Accelerator Mass Spectrometry) facilities, among these at VERA (Vienna Environmental Research Accelerator).

We will discuss the feasibility of uranium prospection by using natural ^{236}U in well water, and summarize existing measurements. Uranium ores show up to $^{236}U/U=10^{-10}$, but measurements of uranium from low-concentration rocks or deep well water are sparse or unavailable [2]. The connection between uranium concentration and the ^{236}U/U ratio will be discussed, since trace isotopes (serving as (α,n) targets and "neutron poisons") and water content of the rock can alter the ^{236}U production significantly.

A known highest grade ore deposit was located in Jáchymov, Czech Republic. It is mainly depleted now, but is a perfect test case to investigate whether high grade ore is indicated by high ^{236}U/U concentration in well water in the vicinity. Thus, the Jáchymov region could be a perfect test case to study ^{236}U in well water as a tool for uranium prospection.

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NAM.P54 (Id: 372)

(RADIO)CHEMISTRY IN AMS – A SERVANT OR A PARTNER?

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The world demand for ultratrace analyses of radionuclides with long half-lives has been increasing not only in dating, material, environmental, geo-and cosmochemical studies, but also recently in the pharmaceutical and pharmacological applications. The most sensitive tool for such analyses is the dynamically advancing method of Accelerator Mass Spectrometry (AMS) which enables the determination of long-lived radionuclides in concentrations or amounts lower by up to 6 orders of magnitude than by radiometric methods.

AMS is a very powerful but technologically challenging analytical method that is, at present, mainly a domain of physicists. The sample preparation is often carried out by physicists without wider background in chemistry and its procedures or, in a better case, by chemists without practice in the work with radionuclides and/or sufficient knowledge of AMS needs and the collaboration between chemists and physicists in AMS, similarly to some other ultratrace analytical methods seems to be rather insufficient. However, making full use of the sensitivity of AMS is possible only if precise, reproducible, and well defined methods for sample preparation are used.

We suggest that here is the place where radiochemistry, after more than 100 years of its development, can help with its powerful toolbox of micropreparations, carrier and tracing methods, tracking of contamination sources, numerous separation procedures optimised for radiometric determinations and, in some cases, even with chemical suppression of isobars interferences. Full integration of radiochemistry separation and preparation methods into AMS sample preparation could create a synergy resulting in quality samples/methods well suited for high precision AMS measurements at further decreased background and improved sensitivity.

Thus, our contribution is an appeal on a closer collaboration between (radio)chemists and physicists in the field of Accelerator Mass Spectrometry, thus contributing to a better use of its potentials.

4. CHEMISTRY OF ACTINIDE AND TRANS-ACTINIDE ELEMENTS (TAN)

Lectures

TAN.L01 (Id: 286)

NEW NUCLEUS ^{277}Hs: IN BETWEEN TWO ISLANDS OF STABILITY

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The nuclear fusion reactions ^{244}Pu(^{48}Ca,3n)^{289}114 and ^{244}Pu(^{48}Ca, 4n)^{288}114 were studied at the new, highly efficient gas-filled recoil separator TASCA. The large number of thirteen observed decay chains assigned to ^{288}114 and ^{289}114 enabled reliable cross sections measurements. The highest cross section was 8.2 (+4.0 / -2.8) pb measured for the 4n evaporation channel. The new nucleus ^{277}Hs was observed. It decayed by spontaneous fission after a lifetime of 4.5 ms. Model calculations, which reproduce the properties of nuclides in the region around longer-lived deformed doubly-magic ^{270}Hs well suggest ^{277}Hs to be located near a minimum of shell stabilization between this island of deformed nuclei and the island of stability of spherical superheavy nuclei.

TAN.L02 (Id: 282)

CHEMICAL IDENTIFICATION OF ELEMENT 114 AT TASCA

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The newly built gas-filled separator TASCA, at the GSI Darmstadt has been successfully commissioned. TASCA was designed to study the physics and chemistry of the heaviest element and it has been demonstrated that TASCA is the most efficient device for the production of superheavy elements in complete fusion reactions with a ^{48}Ca ion beam. Two experiments on the physics and chemistry of element 114, produced in the ^{244}Pu(^{48}Ca;3-4n)^{288,289}114 reaction, were performed at TASCA in 2009 within the framework of the international TASCA collaboration. 17 decay chains from element 114 were detected. The decay properties of ^{288,289}114 and the high cross sections reported by Oganessian et al. [1] have been confirmed. Here, we report on the results of the experiment on the chemistry of element 114 that was performed at TASCA in September 2009.

Evaporation residues (EVRs) of element 114 were produced in the irradiation of a ^{244}Pu target (~ 580 μg/cm^2) with ^{48}Ca ions at an excitation energy of 42 MeV. The EVRs were separated in TASCA, which was operated in the Small Image Mode [2]. They then passed through a 3.3 μm Mylar window (30 x40 mm^2) into a Recoil Transfer Chamber (RTC) of 29 cm^3 in volume made from Teflon. The atoms were thermalized in the RTC in a He/Ar gas mixture (7:3) and then flushed into the COMPACT detector [3], which was directly connected to the exit of the RTC. Two chromatography channels made from 32 pairs of PIN diodes covered with a thin gold layer were connected in series: the first one was kept at the room temperature, and the second one had a negative temperature gradient from +20 to -160°C. This allowed efficient detection of chemical species with different volatilities: from non-volatile Pb to very volatile Rn. The distribution of Pb and Hg in COMPACT was measured in separate experiments with short-lived Pb and Hg isotopes that were preseparated in TASCA. For monitoring the detection system, a small amount of ^{219}Rn was added to the carrier gas and was responsible for α particle background in spectra. No other peaks from α particles were found. Two decay chains from element 114 were detected: one from ^{288}114 and one from ^{289}114 produced in 4n and 3n evaporation channels, respectively. Both decays from element 114 isotopes were observed in the first detector channel, held at room temperature. From this observation, we can conclude that element 114 is less volatile than element 112 and more similar to Hg. This is in agreement with theoretical predictions [4] and contradicts previous chemical experiments with element 114 as performed by the PSI group at FLNR Dubna [5].

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TAN.L03 (Id: 252)
INDEPENDENT VERIFICATION OF ELEMENT 114

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Production of numerous superheavy elements (SHE) in ^{48}Ca irradiations of actinide targets was reported by the Dubna Gas Filled Recoil Separator (DGFRS) group in past years. Independent verification of these observations has been of paramount importance, but confirmation experiments failed to produce SHE in ^{48}Ca induced reactions. We report on the first successful independent verification of the production of element 114 in the reaction ^{48}Ca(^{242}Pu,3-4n)^{287,286}114 at a center-of-target energy of 244 MeV. Two genetically correlated decay chains were observed during 8-day experiment at the Lawrence Berkeley National Laboratory's (LBNL) 88-Inch Cyclotron. Based on the observed decay properties these decay chains were attributed to decay of ^{287}114 and ^{286}114 produced in 3n and 4n channel, respectively. Implications of this observation on chemistry and physics of the heaviest elements will be discussed.

TAN.L04 (Id: 133)
ELEMENT 114 CHEMISTRY AND WHAT IS NEXT?

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Based on the periodic table, element 114 should be placed in the group 14, which includes elements like C, Si, Ge, and Pb. E114 is expected to reveal a more metallic behaviour than its lighter homologues. However, relativistic quantum chemical calculations predict different chemical behavior, namely, higher inertness in comparison with Pb [1-3]. In this paper experimental determination of deposition temperature of element E114 on gold is presented.

During the irradiation of ^{242}Pu with ^{48}Ca (see [4]) a decay chain was observed, which was assigned to the primary product of the nuclear fusion reaction – the isotope ^{287}E114 ($T_{1/2} = 0.5$ s). A deposition temperature for this isotope of -88°C was observed. This unusual first chemical observation of element E114 was confirmed switching to the projectile target combination ^{48}Ca and ^{244}Pu. The production of ^{288}E114 and ^{289}E114 are reported in the reactions ^{244}Pu (^{48}Ca, 4n) and ^{244}Pu (^{48}Ca, 3n), respectively [5]. During this experiments two more decay chains attributed to the isotope ^{288}E114 were observed, fully confirming the first observation. A kinetic Monte-Carlo model of gas adsorption chromatography [6] assesses the adsorption enthalpy ($-\Delta H_{ads}(Au)$) from the observed deposition pattern of element 114 on the gold surface in the COLD at the applied experimental conditions as $-\Delta H_{ads}(Au)(E114) = 34\pm54(11)$ kJ/mol (95% c.i.). Recent relativistic density functional calculations predict the formation of a metallic bond between E114 and gold [3]. A semi-empirical macroscopic metal-metal adsorption model [7,8] predicts adsorption enthalpy of a metal-like E114 on gold of $-\Delta H_{ads}(Au)(E114) = 183$ kJ/mol. The adsorption enthalpy of a noble-gas like element 112 on gold surfaces was estimated to $-\Delta H_{ads}(Au)(E112) = 42\pm5$ kJ/mol [9]. The comparison between these theoretical values and our experimental result concludes the formation of a noble-gas-like weak physisorption bond between atomic 114 and a gold surface in contrast to the expectations from the relativistic models and from empirical predictions. On a 95% confidence level E114 is interacting weaker with gold compared to mercury. New possible experimental techniques pointed to investigation of chemical properties of super heavy elements will be discussed.

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TAN.L05 (Id: 172)
VACUUM THERMOCHROMATOGRAPHY - THEORY AND MONTE CARLO SIMULATION

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Mixtures of ultra low amounts of species can be separated in a tube, evacuated well below a microbar, along which the temperature gradually decreases from the closed hot end, by placing the sample into this hot end (zero coordinate). Then the Knudsen flow results in that the components with dissimilar adsorbability on the tube surface make deposits, which peak in different temperature intervals away from zero. It happens despite the absence of any convective flow, which is the basis of all true "chromatographies". A few experimental works published to date demonstrate separations of both very volatile and quite nonvolatile species.

This "vacuum thermochromatography" (VCT) is based on erratic independent displacements of the molecules up and down the tube. Indeed, a molecule experiences free flights between two sequential collisions with the tube wall, while each strike results in an adsorption event and so in an adsorption sojourn time. Thus the molecule does not keep memory about its incoming velocity vector; it is commonly supposed that the angular distribution of the rebounds follows the cosine law.

The paper presents the first rigorous consideration of the microscopic picture of VCT to substantiate MC simulations. Formulae for the flight length and its projection on the tube axis were derived. Algorithms of calculating random values and the probability density functions for the above quantities were obtained. Erroneous formulae and values met in literature were corrected.

In an isothermal tube, the picture obtained by MC simulations was indistinguishable from the solution of the corresponding diffusion problem. It is the half-Gaussian – because the migration is reflected at zero – and the mixture components differ only in the width of that distribution. However, in the case of VCT, the average adsorption sojourn time, which is proportional to the Boltzmann factor (depending exponentially on the desorption energy), rapidly increases with lower temperature. As a result, two molecules displaced from the given coordinate in opposite directions have different probability per unit time to make next steps. This is the background for evolution of the adsorption peak, which "moves" towards lower temperature with longer duration of the experiment.

Empirical regularities in the peak coordinate and temperature of the simulated peaks as a function of some experimental conditions were obtained. Some reasonable function for fitting the peak profiles was proposed. Attempts to use the diffusion ansatz for the interpretation of VCT data were undertaken though the strong temperature change of the effective diffusion coefficient with the coordinate brings problems in diffusional description.

VCT as a method of preparative separation offers prospects of solving some specific task in radiochemical analyses. One of the goals is finding the energy of desorption of various species. The here obtained equations with semi-empirical parameters allow thermodynamic interpretation of the experimental data. The earlier attempts on these lines are critically discussed.

As to the studies of superheavy elements, the advantages are in that the surface seen by the atoms can be kept very clean and no correction of the measured α particles energies is necessary.

TAN.L06 (Id: 143)

GAS PHASE CHEMICAL STUDIES OF SUPERHEAVY ELEMENTS USING THE DUBNA GAS - FILLED RECOIL SEPARATOR – STOPPING RANGE DETERMINATION

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The use of the Dubna Gas-Filled Recoil Separator (DGFRS) [1] as a preseparation device for gas phase chemistry experiments with elements 112 and 114 led us to the task of reevaluating the stopping force (STF) of superheavy elements (SHE). In the experiment the DGFRS and the IVO-system [2] were separated by a thin Mylar window of variable thickness so that the evaporation residues (EVR) can pass it. The reaction product collection (RPC) chamber, filled with gaseous argon and mounted behind the Mylar foil, had to be as small as possible to increase the transport efficiency to the thermochromatography detector COLD. Due to scarce availability, we measured STF data for Hg, Rn, and No in Mylar and gaseous argon at energies of around 30 to 50 MeV. These data were used for an extrapolation to STF of SHE 114.

A 265 MeV ^{48}Ca beam was delivered from the U-400 cyclotron at the Joint Institute of Nuclear Research (JINR) in Dubna, Russia. The beam was passed through a 1.6 μm Ti window, a 1.6 μm Ti target backing into the target materials. Three different target materials were irradiated: neodymium oxide (nat. Nd_2O_3, 0.30 mg/cm^2), dysprosium oxide (nat. ^{203}Dy, 0.33 mg/cm^2) and lead (^{208}Pb, 0.42 mg/cm^2, 1 μm Cu degrader in front). The mainly produced EVR's in the fusion of ^{48}Ca with target nuclei are according to HIVAPSI [3] ^{185}Hg, ^{206}Rn or ^{254}No, respectively. The EVR's were separated in the DGFRS according to their mass to charge ratio and guided on an exchangeable Mylar foil of 2, 3 or 5 μm thickness separating the RPC-chamber from the DGFRS. The RPC-chamber had a depth of 18 mm and was filled with thermalizing argon gas of a pressure between 0.0 and 0.4 bar. In the center of the focal plane of the separator at the back wall of the RPC-chamber a 20 x 20 mm silicon detector (PIN-diode) was mounted to detect the signals and the α decays of implanted products. Different data points were gained by varying the Mylar foil thickness and the argon gas pressure.

For the extrapolation of the stopping range to 114 we used the assumption of proportionality of the STF of a heavy ion to the velocity dependent effective charge (q_{eff}) and the equivalent STF of a proton (epSTF) [4]. These epSTF's which are uncorrected for any other STF terms, were plotted against 1/ln(Z) an empirically established dependence of the STF on Z of the heavy ion. The obtained values were compared with the stopping code SRIM 2008 [5] and finally used to setup the gas chromatography experiment with elements 112 and 114. The results are accepted for publication [6].

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TAN.L07 (Id: 7)

PROGRESSES ON THE STRUCTURAL CHEMISTRY OF THE ACTINIDE PHOSPHATES

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Data on structure and properties of actinide phosphates are needed for the development of phosphate ceramics envisaged as possible hosts for long-term sequestration of actinides generated by the nuclear fuel cycle. In that aim an extensive study on the crystal structures, thermal stability, and thermodynamic properties on a number of crystalline trivalent and tetravalent actinide phosphates was undertaken. $PuPO_4$ with monazite structure was obtained and characterized in terms of thermal stability and thermal expansion. The decomposition of $PuPO_4$ to Pu_2O_3 was observed at 1573 K. Its high-temperature heat capacity is to be measured up to 1500 K by drop calorimetry. $M^{II}Np(PO_4)_2$ (M^{II} = Ca, Sr, Ba) were obtained by solid state reaction. The influence of ionic radius of M^{II} (M^{II} = Ca, Sr, Ba) on the crystal structure is further discussed. Pure phase of $CaNp(PO_4)_2$ with monazite structure (P2/n) was obtained by solid state reaction after several thermal treatments and readjustments in stoichiometry. The lattice parameters are a = 6.65085(3) Å, b = 6.83893(5) Å, c = 6.35378(3) Å, and β = 104.117(9) o. $SrNp(PO_4)_2$ seems to adopts an inedited double-monazite structure with a = 6.895 Å, b = 13.272 Å, c = 6.892 Å, and β = 99.22 o, probably due to the Sr/Np ordering responsible for the non-negligible reflections with k = 2n +1; in this case, the two cations seem too different in size to allow disorder, contrarily to $CaNp(PO_4)_2$. Pure $BaNp(PO_4)$ with monoclinic C2/c was also obtained. Its structure was inferred with the one reported for $RbEu(SO_4)_2$, the lattice parameters being a = 12.69095(13) Å, b = 5.36179(6) Å, c = 9.40949(11) Å, and β = 102.516(68) o. The pattern looks similar to the one previously found for the Th-counterpart.

TAN.L08 (Id: 97)

SINGLE CRYSTAL X-RAY DIFFRACTION AND RAMAN CHARACTERIZATION OF TERNARY CE(IV) AND PU(IV) PEROXO-CARBONATE COMPOUNDS COMPRISED OF RARE DIMERIC MOLECULAR UNITS

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The environmental fate and transport of transuranium elements and the search for alternative f-element separation methods require a better understanding of the speciation of transuranics under oxidizing alkaline conditions, especially in the presence of ubiquitous ligands such as peroxide and carbonate. From our exploration of the Ce and Pu peroxo-carbonate systems several new structures containing the known $[(CO_3)M(O_2)_2Ce(CO_3)_3]^{8-}$ (M = Ce(IV) or Pu(IV)) dimer were found. A simplified synthetic procedure was used to prepare crystalline products of several novel compounds, namely $[NH_4]_8[(CO_3)_3Ce(O_2)_2Ce(CO_3)_3].6H_2O$, $[C(NH_2)_3]_8[(CO_3)_3Ce(O_2)_2Ce(CO_3)_3].4H_2O$, $Na_8[(CO_3)_3Ce(O_2)_2Ce(CO_3)_3].12H_2O$ and $K_8[(CO_3)_3Pu(O_2)_2Ce(CO_3)_3].12H_2O$. Structural characterization of the aforementioned compounds was performed using single crystal x-ray diffraction. It was observed that the crystal packing of the $[(CO_3)_3M(O_2)_2Ce(CO_3)_3]^{8-}$ dimeric unit varied significantly depending on the cation incorporated into the structure (e.g., Na^+, K^+, NH_4^+ , and $C(NH_2)_3^+$). Additional characterization of the f-element compounds was performed on single crystals using a high-resolution microscope-based Raman spectrometer.

TAN.L09 (Id: 128)

A COMPARITIVE STUDY OF U(III) AND U(IV) COMPLEXES IN A ROOM TEMPERATURE IONIC LIQUID SYSTEM

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Room temperature ionic liquids (RTILs) were known as early as the 19th century beginning with alkylammonium nitrates. The field grew in the 1970s by using combinations of air and water sensitive aluminum halides with an organic cation salt. While these ionic liquids were useful and had many desirable features, they were extremely sensitive to air and water, limiting their utility. RTILs today are salts that are liquids from as low as -96 °C to up to 100 °C. RTILs are of high interest in many fields of study because of their negligible vapor pressure, high electrochemical stability, high conductivity, and wide electrochemical windows. In addition, RTILs are useful systems for studying f-elements in the absence of hydrolysis. Included in this work are studies of multiple uranium complexes in a room temperature ionic liquid system, $[Me_3NnBu][N(CF_3SO_2)_2]$. Uranium materials in the 3+ and in the 4+ oxidation states were prepared in inert atmosphere conditions. Infrared and ultraviolet-visible spectroscopy was utilized to give details regarding the speciation of each system. Electrochemical analysis was executed, including cyclic and square wave voltammetry as well as electrodeposition from both oxidation states. The resulting electrodes were then analyzed using scanning electron microscopy, energy dispersive spectroscopy, and x-ray diffraction to evaluate the nature of the uranium deposit on the electrode surface. A comparison of the resulting oxidation and reduction processes observed during electrochemical analysis as well as the electrodeposits will be made.

TAN.L10 (Id: 177)
STRUCTURAL ASPECTS OF URANYL COMPLEXES WITH LIPOPOLYSACCHARIDE

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It is well-known that microorganisms play an important role in bioremediation. Because of the high retention capability of heavy metal ions, they significantly influence mobilization and immobilization of cations in soils. The prediction of the radionuclide transport in the environment and the improvement of technical bioremediation strategies require a detailed understanding of the binding mechanisms on a molecular level. Lipopolysaccharide (LPS), the main part of the outer membrane of Gram-negative bacteria, sticks out of the cell wall and is in direct contact with the (aqueous) environment. With its high content of negatively charged functional groups (mainly carboxyl and phosphoryl groups) it plays a key role in protection of contaminants. We have investigated the uranyl LPS interactions to gain detailed information about the coordination sphere in the molecular environment of the uranyl ion. We focused on the identification of the coordinating functional groups over a wide pH range (from 2.5 to 7) and under different stoichiometric conditions. In particular, the discrimination between carboxyl and phosphoryl groups and their binding behaviours was elucidated by extended X-ray absorption fine structure (EXAFS) spectroscopy at the U LIII-edge and attenuated total reflection Fourier-transform infrared (ATR FT-IR) spectroscopy. With respect to environmental conditions, samples with an excess of LPS were investigated by EXAFS. Furthermore, samples with equimolar ratios of uranyl and functional groups of LPS according to a slight deficit of phosphoryl groups were determined with FT-IR. EXAFS spectra show great similarities to the uranyl mineral phase meta-autunite. A four-fold complexation of the uranium was derived from very short $U\text{-}O_{eq}$ distances of 2.28 Å and U-P distances of 3.58 Å indicating unidentate coordinated phosphoryl groups. Furthermore, U-U interactions can be observed at 5.2 Å and 6.9 Å. FT-IR spectra show spectral evidence for both, carboxyl and phosphoryl coordination. A downshift of the antisymmetric streTching mode of the carboxylate group from 1576 (uncomplexed LPS) to 1530 cm^{-1} and an upshift of the symmetric streTching mode from 1404 (uncomplexed LPS) to 1455 cm^{-1} upon complexation provides evidence for a bidentate complexation to carboxylate groups. The antisymmetric and symmetric streTching modes of the complexing phosphoryl groups are observed at 1105 and 1060 cm^{-1}. In summary, we determined at high LPS excess preferential phosphoryl coordination, whereas with an increasing relative amount of uranyl ions, corresponding to a decreasing number of functional groups of LPS, additional carboxylate coordination becomes important. This complexation behaviour remains within a broad pH range from slight acidic to neutral values. Under the investigated experimental conditions, the coordination of uranyl ions to the LPS molecule is obviously controlled by the U/LPS concentration ratio irrespective from prevailing pH.

TAN.L11 (Id: 296)
THEORETICAL STUDY ON „ITINERANT" PROPERTIES OF YTTRIUM AND AMERICIUM(III) IN THE LANTHANIDE SERIES WITH RESPECT TO STABILITY OF THEIR COMPLEXES

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As regards the stability of metal complexes with hard ligands, yttrium can be located among heavy lanthanides, as expected from the ionic radii of the metals. However, in the case of complexes with soft ligands, yttrium behaves as a light lanthanide. This effect has been explained in terms of covalent shortening of the metal-ligand bond [1], due to the participation of 4f orbitals of Ln(III) in bonding [2]. The ligand-dependent itinerant properties of yttrium enable developing efficient procedures for its separation from lanthanide mixtures, e.g. by solvent extraction. Trivalent actinides undergo analogous shifts across the lanthanide series, but to the opposite direction [3], which was explained as the effect of still greater contribution from the 5f orbitals to the bonding. In this work quantum mechanical calculations were carried out on model complexes of Y(III), Am(III) and some selected Ln(III) with nitrates (hard ligands) and thiocyanates (soft ligands). The aim of the work was to reproduce the experimental shifts of the positions of Y(III) and Am(III) in the two series of lanthanide complexes, and to examine the contributions from the 4f and 5f orbitals into the bonding (metal-ligand orbitals). All the calculations were done by using Gaussian 03 package. Density functional theory was used in the form of Becke three-parameter hybrid functional B3LYP. Stuttgart-Dresden SDD small-core pseudo-relativistic basis set was employed for metal atoms and standard all-electron Pople 6-31G(d) double valence basis set was used for other atoms. Geometry optimizations were followed by frequency calculations to prove that the obtained stationary points had been the true minima on the potential energy surfaces. The results and the calculation approach may appear helpful in further studies on the origin of selectivity of tetra-N-heterocyclic ligands (BTBP) for actinides(III) over lanthanides(III) in solvent extraction SANEX processes. Quantum mechanical calculations for the M(III)-BTBP complexes are extremely troublesome. Various approaches to actinide selectivity in similar systems have been presented in a recent review [4].

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Posters

TAN.P01 (Id: 132)
THERMOCHROMATOGRAPHIC INVESTIGATION OF ^{113}In, ^{125}Sb AND ^{125m}Te IN QUARTZ COLUMNS

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Investigation of chemical properties of super heavy elements (SHE) produced in ^{48}Ca-induced nuclear fusion reactions with actinide targets [1-4] represents a challenging task for chemists world wide. In the present work ΔH_{ads} of carrier-free ^{113m}In, ^{125}Sb and $^{125m}TeO_2$ species on quartz surface was determined. Lighter homologues of SHE elements ^{113m}In ($T_{1/2} = 99$ min), ^{125}Sb ($T_{1/2} = 2.7582$ y) and ^{125m}Te ($T_{1/2} = 57.40$ d) were prepared by neutron irradiation. For that purpose 0.5 g of nat-Sn was irradiated at the SINQ-NAA at PSI for 2 h. The irradiated sample was used as source for thermochromatographic investigations with quartz as stationary surface. Chemical behaviour of In, Sb, and Te species was studied under different experimental conditions (carrier gases, gas mixtures and flow rates). Specially designed gradient oven allowed to achieve a temperature gradient of 1300 °C down to -140 C. Interaction of metallic Sb and In with quartz surfaces was investigated using a highly purified H_2 to exclude trace amounts of water and oxygen. The entire thermochromatographic column was encapsulated in a steel tube. The gas had to pass a Ta getter (1000 °C) before hitting the tracer source deposited on Al_2O_3 and heated up to 1300 °C. Afterwards, the released isotopes passed again over a hot Ta getter. This setup provided for the elemental state of the quite reactive Sb. It was found that deposition temperature Sb in elemental state is 580°C in good agreement with literature data, whereas deposition temperature for In is higher ($T_{dep} = 710$ °C) than the reported one [6]. Monte-Carlo simulation [7] was applied to obtain the adsorption enthalpy of the species on quartz surfaces at zero surface coverage (ΔH_{ads}). Calculated enthalpy of adsorption for In was -235 kJ/mol and for Sb was -205 kJ/mol. Using H_2/H_2O gas mixture it was possible to determine deposition temperature of ^{113}In(OH) ($T_{dep} = 320$ °C) and $^{125}Sb(OH)_3$ ($T_{dep} = 360$ °C). The deposition temperature results in $\Delta H_{ads}(InOH) = -145$ kJ/mol and $\Delta H_{ads}(Sb(OH)_3) = 155$ kJ/mol. The interaction of Te species with quartz surfaces was investigated using O_2/H_2O gas mixture. $^{125m}TeO_2$ was deposited at 590 °C resulting in an enthalpy of adsorption $\Delta H_{ads}(TeO_2) = -205$ kJ/mol. The data obtained will be useful for the design of experimental set-ups for gas chromatographic experiments with real superheavy elements, especially E113-E115.

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TAN.P02 (Id: 209)
APPLICATION OF COPRECIPITATION FOR THE STUDY OF CHEMISTRY OF RF

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Chemical identification of radionuclides in the decay chains of the recently discovered transactinide elements may serve as a method for confirming the discovery [1]. The longer half-lives of radionuclides of transactinides discovered in the An + ^{48}Ca reactions extend the application of radiochemical techniques to their chemical characterization [2]. The method of cocrystallization can give valuable information on properties of transactinide compounds, not available in the macroscopic, therefore the identification of coprecipitation mechanism of lighter homologues of transactinides is of scientific interest. Here we report the coprecipitation behavior of Ti, Zr, Hf and Th as lighter homologues of Rf with lanthanum fluoride. The kinetics of coprecipitation and distribution of group 4 elements between the liquid and solid phase, both preformed and formed during the distribution process, was studied as a function of concentration of the micro- and macrocomponent, foreign multivalent ions, HF, pH in solution. The solid phase of prepared samples was characterized by electron microscopy (SEM and TEM). The results show that the coprecipitation mechanism of Zr and Hf with LaF_3 is different from Ti and Th and can be assigned to heterovalent isomorphism. A possibility of using coprecipitation with lanthanum fluoride for comparative investigation of the behavior of Rf in HF solutions was demonstrated.

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TAN.P03 (Id: 212)
SORPTION OF Ti, Zr, Hf AND Nb, Ta AS HOMOLOGUES OF Rf AND Db FROM MIXED AQUEOUS-ORGANIC HF SOLUTIONS

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In order to optimize conditions for isolation and study of chemical properties of Rf and Db [1], we have investigated sorption of their lighter homologues Ti, Zr, Hf, Nb and Ta, from HF solutions in water-miscible organic solvents using the Dowex 50x8 cation exchange (CIX) and AG 1x8 anion exchange (AIX) resins. We have studied dependence of sorption on concentration of HF and acetone. We found that using the AIX resins the distribution coefficients of Ti, Zr, Hf and Nb increased and those of Ta decreased with increasing content of acetone in HF solutions. The results indicated that in concentrated (> 3 M) HF solutions containing acetone, group 4 elements formed MF_7^{3-} (M = Ti, Zr, Hf) complexes whose stability decreased in reverse order of sorption Zr > Hf > Ti. Obtained results are in agreement with previously reported data [2,3] where using CIX chromatography and HCl-HF mixed solutions group 4 elements were separated and eluted in order of Zr > Hf > Ti > Th. Under the same conditions, group 5 elements predominately formed complexes $NbOF_5^{2-}$ and TaF_7^{2-} with stability considerably different, which decreased in sequence Nb > Ta. We have found optimal conditions for separation of the elements using CIX and AIX chromatography. The advantages of studying chemical properties of Rf and Db in aqueous HF solutions mixed with organic solvents are briefly discussed.

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TAN.P04 (Id: 15)
THERMODYNAMIC STUDY ON THE U(VI) COMPLEXATION WITH "ALIPHATIC" AND "AROMATIC" DI-CARBOXYLIC ACIDS BY MICRO-CALORIMETRY

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The thermodynamics of complex formation of U(VI) with oxydiacetic acid (ODA), thiodiacetic acid (TDA), iminodiacetic acid (IDA), isopthalic acid (IPA), 2,5-frandicarboxylic acid (FDA) and dipicolinic acid (DPA) were studied by the micro-calorimetric titration technique. So far, extensive data on stability constant have been obtained and summarized in databases for a variety of combinations of actinide ions and complexants which could be found in the geosphere to predict the migration behavior of actinides in deep underground for the safety assessment of radioactive waste disposal. However, enthalpy and entropy data of actinide elements are scarce in comparison with stability constants. These data are indispensable not only for understanding of the reaction mechanism but also for the estimation of stability constants at elevated temperatures outside the range of 20-30 °C by the extrapolation from those at a room temperature with using thermodynamic models. Following our previous study reporting thermodynamic data (ΔG, ΔH and ΔS) of complexation of 8 di-carboxylates with U(VI) [1], this paper presents the thermodynamic quantities of U(VI) complexation with three "aliphatic" di-acetic acids and three "aromatic" di-acetic acids. Those ligands have a different center atom between two carboxylic groups, i.e., oxygen for ODA and FDA, sulfur for TDA and nitrogen for IDA and DPA, which may coordinate with a metal cation. The obtained thermodynamic quantities are compared with those of U(VI) complexation with glutaric acid (GA) for the discussion of the effect of structural difference, i.e., aliphatic- and aromatic structure and types of central atom in the ligands on the complex formation thermodynamics. From the thermodynamic quantities, i.e., ΔG, ΔH and ΔS determined by the potentiometric and calorimetric titration techniques, it is indicated that these reactions are mainly driven by the entropy change while the enthalpy change is not promoting the progress of the reaction. The ΔG, ΔH and ΔS of 1:1 IDA complex show remarkable difference from those of GA complex, while those of ODA and TDA complexes are almost equal to those of GA complex. ΔG of 1:1 IDA complex with U(VI) was 30 kJ/mol larger than that of GA complex, which is generally referred to the chelate effect of the ligand. Our calorimetric measurement has revealed that this "chelate effect" should be ascribed to the "entropy effect" since the entropy change ($T\Delta S$) of IDA complex was 50 kJ/mol larger than that of GA complex. The origin of this large entropy effect is discussed in the presentation with the help of structural information of the complex calculated using density functional theory (DFT).

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TAN.P05 (Id: 64)
A STUDY OF HETEROGENEOUS EQUILIBRIA IN THE SATURATED AQUEOUS SOLUTIONS OF URANOARSENATES AND URANOPHOSPHATES OF ALKALINE AND ALKALINE-EARTH ELEMENTS

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Uranoarsenates and uranophosphates belong to a numerous difficult soluble compounds that can be formed by the ingress of various uranium-containing waste in the environment and can also be used as uranium-fixing matrices in technological processes. Research of the state of these compounds in aqueous solutions, of their hydrolytic stability and solubility, and of transformations of uranoarsenates and uranophosphates into other chemical compounds both in the solid phase and in a solution is important. In this work detailed research of the state of $Mk(BvUO_6)_k.nH_2O$ compounds (Bv - As,P; Mk - H^+, Li^+, Na^+, K^+, Rb^+, Cs^+, NH_4^+, Mg^{2+}, Ca^{2+}, Sr^{2+}, Ba^{2+}) in aqueous solutions at 25 °C within a wide range of acidity is presented. It has been found that the behavior of all $Mk(BvUO_6)_k.nH_2O$ compounds in aqueous solutions obeys the general laws due to their structural and functional similarity. Acidity has the most essential effect on the chemical stability of uranium compounds under study. Composition and structure of the solid phase, ionic-molecular forms, and concentrations of U(VI), As(V), P(V) and Mk in equilibrium solutions depend on pH. In acidic medium ($pH<2$) difficult soluble acid with composition of $HBvUO_6{\cdot}4H_2O$ is formed as a result of ionic exchange in the solid phase. In the case of the $Mk(BvUO_6)_k{\cdot}nH_2O$ interaction with aqueous solutions of the alkaline elements hydroxides ($pH>10$) As(V) and P(V) are leached into the solution and insoluble uranates are formed. In the intermediate interval ($2<pH<10$) secondary solid phases are formed in quantities defined by acidity of the equilibrium solutions. $(UO_2)_3(AsO_4)_2{\cdot}12H_2O$, $(UO_2)_3(PO_4)_2{\cdot}8H_2O$ and $UO_3.2H_2O$ as synthetic analogues of the natural minerals should be mentioned among these phases. The solubility of all compounds with formulae $Mk(BvUO_6)_k.nH_2O$ has minimal value in neutral solutions (10^{-6}-10^{-5}M) and sharply increases with addition of acid or base. The uranophosphates are characterized by higher chemical stability than uranoarsenates. The finer distinctions in behavior of $Mk(AsUO_6)_k.nH_2O$ and $Mk(PUO_6)_k.nH_2O$ are caused by the nature of the Mk elements and they reveal themselves in conversions of the secondary phase formation reactions and in the acidity-base intervals of the primary solid phase existence. A physicochemical model characterizing quantitatively the equilibrium state in the heterogeneous systems under study is proposed for predicting the processes occuring when inorganic uranium compounds dissolve. Using the model it is possible to calculate the state diagrams of the solid phases and the equilibrium solutions over the phases. In addition, it is possible to evaluate various heterogeneous equilibria constants.

TAN.P06 (Id: 78)
SYNTHESIS, STRUCTURE AND PROPERTIES OF SYNTHETIC TROGERITE $(UO_2)_3(AsO_4)_2{\cdot}12H_2O$

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Synthetic trogerite (ST) $(UO_2)_3(AsO_4)_2{\cdot}12H_2O$ can be obtained by mixing uranyl nitrate and arsenic acid solutions in equivalent ratio 3:2. In this case the following competitive reactions become possible: $UO_2(NO_3)_2 + H_3AsO_4 + 4H_2O \leftrightarrow HAsUO_6{\cdot}4H_2O(s) + 2HNO_3$ (uranoarsenic acid formation) $3UO_2(NO_3)_2 + 2H_3AsO_4 + 12H_2O \leftrightarrow (UO_2)_3(AsO_4)_2{\cdot}12H_2O(s) + 6HNO_3$ (ST formation) Increase of acidity results in uranoarsenic acid formation while its decrease enhances triuranyl diarsenate hydrates formation. The partially dehydrated ST and a mixture of crystallohydrates are formed at elevated temperatures. So far ST has not been obtained in the form of an individual crystal compound, free of any impurity phases. Its X-ray data and crystallographic characteristics are unknown. In this work the ST synthesis technique is proposed. Dehydration, thermolysis, structure and properties of ST were studied by X-radiography, IR-spectroscopy, X-ray fluorescence analysis, and scanning calorimetry. It has been found that dehydration is the only temperature-caused transformation in the range of 20-800°C. In the interval 50-200°C trogerite loses 8 molecules of water and transforms into the well known $(UO_2)_3(AsO_4)_2{\cdot}4H_2O$. Both crystallohydrates with crystal lattices built of coordination polyhedrals belong to the U(VI) compounds with layered structure. Layers are formed by pentagonal bipyramids UO_7, connected by the general edge to produce continuous chains with AsO_4^{3-} tetrahedrons. Opposite layers of tetrahydrate are strongly bound by covalent chains $(O_3)As$-O-$U(O_5)$-O-$As(O_3)$ of the length of 8.782. It is not long enough for similar bonding of opposite layers at a distance of 11.079 in $UO_2(AsUO_6)_2{\cdot}12H_2O$. Therefore bonding of layers in trogerite is due to non-rigid H-bonds, formed by H_2O molecules coordinated to U(VI) atoms. Three H_2O molecules in $UO_2(AsUO_6)_2{\cdot}4H_2O$ exhaust coordination possibilities of interlayer U(VI) atom in the pentagonal bipyramid. The fourth H_2O molecule is located in zeolite-similar cavities. It is easily released at the onset of $UO_2(AsUO_6)_2.12H_2O$ dehydration. Release of H_2O molecules proceeds in 2 stages at 185 °C and 336 °C. $UO_2(AsUO_6)_2$ with a strongly deformed crystal lattice due to coordination unsaturation of interlayer U(VI) atoms is formed as a result of full dehydration of trogerite. It is almost immediately hydrated by atmospheric moisture, forming the tetrahydrate mentioned above.

TAN.P07 (Id: 147)

INVESTIGATIONS ON STABILITY AND STRUCTURE OF THORIUM COLLOIDS

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This work presents results about the formation of thorium(IV) colloids and precipitates in the presence of silicic acid. Three methods were used for the preparation of the thorium compounds: - Experiment A: Silicic acid concentrations and ionic strengths were adjusted in the presence of preformed stable ThO_2 colloids at neutral pH (0.4 mM Th; 0.3 mM Si; I = 0.05 M $NaClO_4$) - Experiment B: Precipitation of ThO_2 colloids in the presence of silicic acid by neutralization from the acidic side of the pH scale (1 mM Th; 0-4.2 mM Si; I = 0.1 M $NaClO_4$) - Experiment C: Precipitation of Th(IV) colloids in the presence of silicic acid by neutralisation of a Th(IV) carbonate solution from the alkaline side of the pH scale (1 mM Th; 0-3 mM Si; I = 0.1 M $NaClO_4$) For the characterization of the colloids, ζ potential measurements as well as NMR and extended X-ray absorption fine structure (EXAFS) spectroscopy were applied. The ζ potentials of the thorium compounds in experiments A, B and C were determined by laser Doppler velocimetry at pH values of 2 - 10. These measurements allowed the estimation of the isoelectric points (IEPs) of the colloids and conclusions about the colloidal stability of the suspensions formed. By the addition of silicic acid to preformed ThO_2 colloids (experiment A), a significant shift of the IEPs from pH ~ 8.5 to lower values (pH ~ 6.5) was observed. Similar results were found in experiment B. Here, the IEP shifted from pH ~ 7.3 to pH ~ 6.0 in the presence of silicic acid. For both experiments the shift of the IEP was a function of the silicic acid concentration. Obviously, the significant amounts of silicate in/on the Th(IV) particles shift the IEP toward the IEP of pure silicic acid (pH < 3) [1]. On the other hand, the results of the ζ potential measurements on the Th(IV) colloids from experiment C demonstrated that the IEP did not change in the presence of varying amounts of silicic acid but remained constantly at pH ~ 5.2. It was also significantly lower than the IEPs of the colloids from experiments A and B formed under acidic conditions. The IEP behaviour of the experiment C colloids is not yet fully understood. The results of ^{29}Si solid state NMR and EXAFS spectroscopy of the thorium samples from experiment C showed that silicate was built in into the solid structure of ThO_2. A possible mechanism is the partial replacement of the Th-O-Th bonds by bridging silicic acid and the formation of Th-O-Si-O-Th links. It is discussed whether the integration of silicic acid in the solid structure results in the formation of a still unknown thorium compound or a thorite ($ThSiO_4$) like material was formed.

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TAN.P08 (Id: 321)

STRUCTURES OF HYDRATED AND HYDROLYZED THORIUM(IV) DETERMINED BY EXAFS AND LAXS

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The hydrated thorium(IV) ion is nine-coordinate in strongly acidic aqueous solution with a mean Th-O bond distance of 2.45(1) Å as determined by extended X-ray absorption fine structure (EXAFS) and large angle X-ray scattering (LAXS) [1]. The presence of a second hydration sphere was indicated by LAXS with cca 18 water molecules at 4.66(2) Å. Structural studies in aqueous solution of the hydrolysis products of thorium(IV) identified three different types of hydrolysis species: a μ_2O-hydroxo dimer, $[Th_2(OH)_2(H_2O)_{12}]^{6+}$, a μ_2O-hydroxo tetramer, $[Th_4(OH)_8(H_2O)_{16}]^{8+}$, and a μ_3O-oxo hexamer, $[Th_6O_8(H_2O)_n]^{8+}$. Strongly acidic solutions are required to stabilize the hydrated thorium(IV) ion. With increasing concentration and pH thorium(IV) hydrolyzes, first, to a dimer with double hydroxo bridges, then to a tetramer with the thorium(IV) ions forming a diamond (represented by two dimers merged together) [1]. Finally, at pH value close to precipitation of thorium(IV) oxide/hydroxide, a hexamer is formed with the thorium atoms forming a rhomboctahedron. Potentiometric studies suggested a hexamer composition $[Th_6(OH)_x(H_2O)_n]^{24-x}$, x = 14 or 15 [2]. However, the rhomboctahedral structure prevents for sterical reasons such a large number of hydroxide groups to be bound. Instead 7-8 oxo groups are bound to thorium ions as from proton activity point of view 16 hydroxo groups are equivalent to 8 oxo groups, making it impossible to distinguish between e.g. $Th_6(OH)_{16}^{8+}$ and $Th_6O_8^{8+}$ complexes. A compilation of reported solid state structures of actinoid(IV) compounds with oxygen donor ligands show a strong correlation between the An-O bond distance and the coordination number [1]. EXAFS studies of uranium(IV), neptunium(IV) and plutonium(IV) ions in aqueous solution have reported accurate An-O bond distance, but the coordination numbers were less accurate and often overestimated. The hydrated tetravalent actinoid ions in aqueous solution all seem to be nine-coordinated [1].

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TAN.P09 (Id: 155)
ELECTROCHEMICAL BEHAVIOUR OF CURIUM IN CHLORIDE MELTS

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Curium isotopes contained in spent nuclear fuel have a high specific heat rate and long half-life and they should be recovered from high-level wastes (HLW) and transmutated. Molten chlorides have more high radiation resistance than aqueous and organic media. This fact is of significant importance because curium has a high specific activity. There is not much information about the behaviour of curium in molten chlorides. Data available are referred to the conventional standard potential of curium in the alloy formed with zinc [1] and distribution coefficient between salts and liquid metal phase [2]. Data on thermodynamics of formation for oxygen-containing curium compounds in chloride melts were obtained by authors [3] with using of potentiometric titration. This work is devoted to study of non-oxygen curium compound formation in chloride melts. The following standard electrochemical methods were used: CV, SWV, DPV, chrono potentiometry and dynamic galvanometry. Dependences of formal standard electrode potential of curium, Gibbs energy change and other parameters on temperature and effective radius of solvent cation were obtained. Also Pourbe diagrams were drawn.

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TAN.P10 (Id: 156)
STUDY OF REACTION OF CmO^+ AND CmOCl FORMATION IN CHLORIDE MELT BY SPECTROSCOPIC METHOD

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Molten chlorides have a higher radiation resistance than aqueous and organic media. This fact is of significant importance because curium has a high specific activity. And, correspondingly, it can be used for partitioning of curium from waste before its vitrification. Data on the thermodynamics of formation for oxygen-containing curium compounds in chloride melts were obtained by authors [1] by potentiometric titration. This work is devoted to more precise determination of these data by the spectroscopic method. Spectrum of Cm^{3+} and CmO^+ complex in melt $NaCl\text{-}2CsCl\text{-}CmCl_3$ at 550, 600 and 650°C over argon atmosphere and different partial pressure of HCl/H_2O has been obtained. It is demonstrated by the appearance of peak for CmO^+ complex in the range of 358 nm as a result of three different reactions, leading to formation of oxide ions in melt. Dependences of Cm^{3+} and CmO^+ concentration on partial pressure of HCl/H_2O were obtained. From these experimental data dissociation constants of CmO^+ and CmOCl have been evaluated.

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TAN.P11 (Id: 163)
KINETICS OF NEPTUNIUM(V) CONVERSION IN STRONG NITRIC ACID SOLUTIONS CONTAINING POTASSIUM PHOSPHOTUNGSTATE, $K_{10}P_2W_{17}O_{61}$

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Behavior of Np(V) in strong nitric acid solutions with different strength (1.0 ÷ 3.0) $mol.L^{-1}$ and KPW (1 ÷ 5)$x10^{-3}$ $mol.L^{-1}$, containing potassium phosphotungstate, $K_{10}P_2W_{17}O_{61}$ (KPW), is examined by spectrophotometric method. It has been established that Np (V) final conversion products under studied experimental conditions are Np (IV) and Np (VI), and the process is going in accordance with the first-order rate law with regard to neptunium(V) concentration.

TAN.P12 (Id: 219)
ELECTROCHEMICAL AND SPECTROSCOPICAL INVESTIGATION OF URANIUM IN ROOM-TEMPERATURE IONIC LIQUIDS

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Room-temperature ionic liquids (RTILs) consist of organic cations and organic or inorganic anions. Beside their fluidity over a large temperature range, they have versatile physical and chemical properties, e.g., low vapour pressure, thermal and chemical stability [1]. In our investigation, the wide electrochemical window of RTILs is of particular importance. RTILs are considered as “green solvents” and may open up new options in industrial actinide separation [2]. This work is about the redox behaviour of hexavalent uranium in 1-butyl-3-methylimidazolium based RTILs. Two different methods have been used: cyclic voltammetry to achieve information about the redox behaviour of uranium(VI) and electrochemical deposition to get samples for X-ray photoelectron spectroscopy (XPS). In both cases the same uranium RTIL solutions have been used. Uranium oxalate ($UO_2C_2O_4$) has been dissolved in 1-butyl-3-methylimidazolium methylsufate (BmimMsu) and 1-butyl-3-methylimidazolium thiocyanate (BmimSCN) under argon atmosphere. Afterwards the solutions (c(U) ~ $8x10^{-3}$ mol/L) have been dried under reduced pressure and heating to minimize the water content in these stock solutions. The amount of water has been determined via Karl-Fischer titration ($\leq$ 10 ppm). For the cyclic voltammetric measurements glassy carbon has been used as working electrode, titanium as counter electrode and platinum as quasi-reference electrode (versus ferrocene/ferrocenium). All experiments have been carried out under argon atmosphere. BmimMsu and BmimSCN provide an electrochemical window of 4.5 - 5.0 V, therefore the reduction of U(VI) to U(IV) should be possible. Actually for both $UO_2C_2O_4$ RTIL solutions cyclic voltammograms have been achieved, which showed a similar progression with a shift of the redox potentials. The cyclic voltammograms suggest a quasi-reversible reduction of uranium(VI) to uranium(IV). The potentials for the U(VI)/U(IV) reduction in BmimMsu and BmimSCN were -1.9 mV and -2.0 mV (vs. SHE), respectively. To verify the reduction of U(VI) to U(IV), samples have been prepared under argon atmosphere for XPS measurements. The uranium was deposited on a titanium foil applying a potential of 2 V for 3 h. The measured binding energies for the U 4f and U 5f XPS-lines confirm the reduction of hexavalent uranium to the tetravalent oxidation state. By combining two different methods, it was possible to demonstrate that U(VI) can be reduced to U(IV) and deposited as tetravalent uranium on titanium foils using RTILs as solvent.

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TAN.P13 (Id: 220)
NOVEL U(VI) SCHIFF BASE COMPLEXES: SYNTHESIS, STRUCTURAL CHARACTERIZATION, AND EXTRACTION STUDIES

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The coordination chemistry of uranium in different oxidation states has recently generated much attention due to several reasons. The most important reason is the separation of U(VI) present in radioactive waste, but also the effects of U(VI) on our environment are of great interest [1-5]. The extraction and separation of U(VI) and other actinides, especially the separation from lanthanides, is most difficult due to their similar chemical behavior [6]. However, the introduction of soft heteroatoms, as imine nitrogen, in the ligand systems could be used as a tool for more selective and effective binding and extraction. We have synthesized and characterized some novel U(VI) complexes using multidentate Schiff base ligands. Structures of these complexes were characterized by X-ray crystallography and DFT calculations. It is the intention of this work to determine not only the structure of these complexes but also the extraction ability of the ligands towards U(VI) and Eu(III).

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TAN.P14 (Id: 117)
PRODUCTION AND INVESTIGATION OF THIN FILMS OF METAL ACTINIDES (Pu, Am, Cm, Bk, Cf)

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With respect to the limited availability of transplutonium metals special techniques and methods of their production have been developed that combine the process of metal reduction from a chemical compound and preparation of a sample for examination. In this situation the evaporation and condensation of metal onto substrate becomes the only possible technology. Thin film samples of metal curium (^{244}Cm, ^{248}Cm) and ^{249}Bk were produced by thermal reduction of oxides with thorium at approx. 1700°C. Metal layers of 0.6-180 μm and 2.5 μm thickness, respectively, were deposited onto the tantalum substrate. For production of metal californium ^{249}Cf in the form of a layer with a thickness of (2.4 ± 0.3) μm the method of thermal reduction of oxide with lanthanum was used. Samples of ^{238}Pu in the form of a film with a layer thickness between 1 and 40 μm and those of ^{239}Pu with a layer thickness between 15 and 40 μm for X-ray structure analysis were prepared by direct high temperature evaporation and condensation of metal onto substrate. For production of ^{241}Am sources a gram sample of plutonium was used containing about 18.5% of americium at the time of production. Thermal decomposition of Pt_5Am intermetallic (262 mg mass) in vacuum was used to produce metal americium with about 80% yield. Resistivity of metal californium (^{249}Cf) films with a thickness of (0.8 ± 0.1) and

(0.1 ± 0.01) μm was measured using the potentiometric method. Initial resistivity values of samples 1 and 2 (5-6 hours after their production) made up (570 ± 80) and (9100 ± 1200) μΩ.cm, respectively. The resistivity of both film samples was found to decrease exponentially with increasing temperature. Negative temperature coefficient of resistivity achieved - (450 ± 22)x10^{-6} K^{-1} in the temperature range 77 - 300 K. The analysis of literature data on metal californium structure allowed an observation of a tendency to form the preferred DHCP structure with increasing sample mass. And, on the contrary, the smaller is the quantity of metal californium under examination, the higher is the probability of interpretation of X-ray diffraction analysis results based on FCC and HCP structures. The performed study on the effect of high specific activity on the crystal structure of plutonium isotope (^{238}Pu) thin layers showed an appearance of non-equilibrium (at room temperature) phases. However, many aspects of this observable phenomenon have not been investigated yet experimentally and comprehended theoretically.

TAN.P15 (Id: 118)
RADIOGRAPHIC EXAMINATION OF CURIUM ALLOYS WITH COBALT, IRON, AND CARBON

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Over many years JSC "SSC RIAR" has been performing activities on the production and investigation of metals of transplutonium elements (TPE), their alloys and compounds. This paper presents the results of production and radiographic examination of micro-samples of ^{244}Cm compounds with ion, cobalt and carbon, namely an identification of crystal lattices of the compounds obtained and calculation of crystal lattice parameters, study on the effect of intensive α-decay on crystal structures of intermetallides and carbides. Samples of the Cm-Co, Cm-Fe and Cm-C systems were prepared by high temperature condensation of metal curium vapor onto corresponding substrates. In the Cm-C sample such a "substrate" represents a thin amorphous carbon layer applied preliminary onto a flat iridium plate. The radiographic examination results of the produced compounds are presented in the table below.

Table. **Crystal lattice parameters of phases detected during investigation of the Cm-Fe, Cm-Co and Cm-C systems**

System	Phase	Lattice	Lattice parameters		
			a, Å	c, Å	V,Å^3
Cm-Co	°$Co_{17}Cm_2$	Hexagonal (P63/mcm)	8.378(7)	8.070(5)	491(1)
	Co_5Cm	Hexagonal (P6/mmm)	4.88(1)	4.08(4)	84(1)
	Co_2Cm	Cubic (Fd3m)	7.242(2)	-	-
Cm-Fe	$Fe_{17}Cm_2$	Hexagonal (P63/mmc)	8.406(3)	8.122(2)	497.0(5)
	Fe_2Cm	Cubic (Fd3m)	7.213(2)	-	-
Cm-C	Cm_2C_3	Cubic ( )	8.3904(5)	-	-
	Cm_3C	FCC	5.172(2)	-	-

Note. In the column "Lattice" a lattice spatial group is indicated in brackets. V - is volume of a crystal lattice elementary cell. Definition errors of the last character are given in brackets after the lattice parameter values. In the Cm-Co system three intermetallic compounds were detected: $Co_{17}Cm_2$ (hexagonal lattice of spatial group P63/mcm), Co_5Cm (hexagonal lattice of spatial group P6/mmm) and intermetallide Co_2Cm (cubic lattice of spatial group Fd3m). Cadmium did not show any solubility in α- and β-CO at room and elevated temperature. In the Cm-Fe system two intermetallic compounds were detected: $Fe_{17}Cm_2$ (hexagonal lattice of spatial group P63/mcm) and Fe_2Cm (cubic lattice of spatial group Fd3m). This system did not show mutual solubility of its components at room temperature. The effect of high α-activity of ^{244}Cm nuclide on the crystal structure of intermetallide Fe_2Cm was demonstrated. In the Cm-C system carbides Cm_2C_3 and Cm_3C with a cubic lattice were detected, which were isostructural with regard to carbides Am_2C_3 and Sm_3C. Crystal lattice parameters of the curium carbides were calculated and data on their x-ray amorphization induced by intensive ^{244}Cm decay were obtained.

TAN.P16 (Id: 119)
FORMATION OF RUTHENIUM ALLOYS WITH CURIUM AND TECHNETIUM

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Presented are the results of production and x-ray examination of ruthenium compounds with curium and technetium. ^{244}Cm alloy microsample was produced by the high-temperature condensation of curium vapor onto the ruthenium substrate and then analyzed by the x-ray diffractometric method. Two intermetallides were detected in the sample: Ru_2Cm with a hexagonal lattice of the Zn_2Mg type with parameters a = 5.279(1) Å, c = 8.812(3) Å; Ru_3Cm with a cubic lattice of the Cu_3Au type and a = 4.151(2) Å. Data were obtained on the x-ray amorphization of lattices of both intermetallides under intensive α-decay of curium-244. The study on the formation of ruthenium alloys with technetium is carried out as part of investigations of technetium transmutation. This process can be implemented in the high-flux reactor SM-3 (Dimitrovgrad, Russia) at an acceptable rate. For the subsequent separation of ruthenium from irradiated samples data are required on the phase relationship in the technetium-ruthenium system under irradiation conditions. This paper describes the x-ray diffractometric examination of samples of the original metal technetium and also those irradiated in the reactor up to a 19, 45 and 70% ruthenium accumulation. The study demonstrates that as a result of the ruthenium accumulation homogeneous solid ruthenium-technetium solutions with a hexagonal close-packed (HCP) structure are formed. Crystal lattice parameters a and c regularly decrease with increasing ruthenium concentration. The existence of a sufficiently strong dependence between the crystal lattice parameters and amount of ruthenium accumulated under irradiation allows the definition of ruthenium content in the irradiated technetium by using the x-ray diffractometric method.

TAN.P17 (Id: 139)
AMERICIUM ALLOYS WITH GOLD AND COPPER

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Over many years JSC "SSC RIAR" has been performing activities on the production and investigation of metals of transplutonium elements (TPE), their alloys and compounds. This paper presents the results of production and X-ray examination of micro-samples of ^{241}Am compounds with gold and copper, i.e. identification of crystal structures of the compounds obtained and calculation of crystal lattice parameters, study of the effect of α-decay on the intermetallide crystal structures. Samples of Am-Au and Am-Cu systems were prepared by high temperature condensation of metal americium vapor onto appropriate substrates. The X-ray examination results of the compounds produced are shown in the table below.

Table. **Crystal lattice parameters of phases detected during investigation of Am-Au and Am-Cu system**

System	Phase	Syngony (space group)	Lattice parameters			
			a, Å	b, Å	c, Å	V, Å^3
Am-Au	Au_6Am	Tetragonal (P42/ncm)	10.389(7)	-	9.703(7)	1047.4(2)
	AuAm	Orthorhombic (Pnma)	7.402(2)	4.564(1)	5.826(1)	196.8(1)
	AuAm	Cubic (Fm3m)	4.784(2)	-	-	-
Am-Cu	Cu_5Am	Hexagonal (P6/mmm)	4.958(1)	-	4.175(2)	88.88(5)

Note. V - is volume of a crystal lattice elementary cell. Definition errors of the last character are given in brackets after the lattice parameter values. The investigated systems did not show mutual solubility of their components at room temperature. In the Am-Au system three intermetallic compounds were found: Au_6Am (of Au_6Sm structural type), AuAm (of CuCe structural type) and intermetallide AuAm (of CsCl structural type). In the Am-Cu system intermetallide Cu_5Am (Cu_7Am) (of Cu_6Ca structural type) was detected. The effect of α-activity of nuclide ^{241}Am on crystal structure of the obtained intermetallide was investigated.

TAN.P18 (Id: 272)
HIGH-RESOLUTION X RAY STUDY OF ELECTRONIC STATE OF IONS WITH NON-STABILITY NUCLEI IN SOLIDS

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The communication presents foundations and results of study of the electronic state of ions of iron, lanthan and actinium groups elements in solids. Original experimental method of the high resolution X ray spectroscopy using X ray fluorescence or synchrotron irradiation will be discussed in this report. It is well known that the energy of X ray lines of ions depends on the electronic state (valence) and environment of ions [1,2]. Results of study of the valence shift of X ray lines upon change of electronic state of nd- and nf- ions in oxides, fluorides and other compounds before and after irradiation or thermal treatment will be presented in this report. The main attention will be focused on discussion of separate results and possibility for investigation of valence shift and X ray lines profile of the ions with non-stable nuclei. By this way we can obtain information on the electronic state of the unstable ions, surroundings of the nl ions and point defects of the solids. Using X ray microanalysor as source of fluorescent irradiation and original two-crystal spectrometer the tested shape of X ray lines is less than 5 MeV and minimum of ions concentration needed for the determination of the ions valence change may be close to 10^{-2} wt % [3].

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5. RADIONUCLIDES IN THE ENVIRONMENT, RADIOECOLOGY (REG)

Lectures

REG.L01 (Id: 256)
IMPACT OF THE CHERNOBYL ACCIDENT ON NORWAY: LESSONS LEARNT

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Norway was one of the European countries most seriously affected by the Chernobyl-accident. According to a nationwide survey in 1986 the total deposition of ^{137}Cs in the country was estimated at 2300 ± 200 TBq and about 10 % of the territory received more than 20 kBq m^{-2} of this radionuclide. This has resulted in significant exposure of wildlife as well as domestic animals grazing in forest and mountain areas and corresponding contamination of meat and milk intended for human consumption. Reindeer were particularly vulnerable due to its lichen feed during large parts of the year, but also sheep (and goats) frequently showed ^{137}Cs activity levels far above the upper limits defined for human consumption. Prognoses based upon on-going monitoring programmes indicate that there will still be problems with high concentrations of ^{137}Cs in animals on rough grazing in Norway for 10-20 years to come. During the first few years after the accident radioecological research had favourable conditions and biologists, chemists, physicists, veterinarians and agricultural researchers were working together to learn about the behaviour of ^{137}Cs in terrestrial and aquatic foodchains. Emphasis was placed on its behaviour in natural soils, uptake in forage, and transfer to animals. Transfer of Cs from soil was studied for a great number of naturally growing plants and fungi, and some mushroom species were found to be extreme Cs accumulators leading to problems in years with abundant mushroom growth. To avoid discarding meat and milk, considerable efforts were done to develop countermeasures reducing Cs activity levels in domestic animals, the most successful being: (1) use of Prussian blue (Gieze salt) to reduce the uptake of ^{137}Cs, (2) providing clean feed a few weeks before slaughter in connection with live monitoring of animals, and solely for reindeer (3) changing slaughter time from February to September, reducing the influence of lichen feeding, which is most important during winter. Prior to the Chernobyl accident food chain transfer was discussed in terms of transfer factors based just on measurements of activity concentrations in the media in question. The research in Norway and elsewhere after Chernobyl has emphasized the importance of chemical speciation of the radionuclides for their behavior e.g. in soil, water, and sediments. The mobility of Cs ions in the boreal soils of Scandinavia with high organic matter content has shown to be much greater than in the previously studied agricultural soils with high content of mineral matter including clay minerals fixing Cs ions strongly. Thus the uptake of Cs in plants from these soils is much greater than previously experienced, and strongly dependent on the chemistry of the soil solution. Particularly high mobility of ^{137}Cs is observed in southernmost Norway where the soils are more acidified than elsewhere. Results from repeated surveys of natural surface soils showed that the decline of ^{137}Cs was greater in coastal regions than in areas farther inland, probably attributable to a greater deposition of Mg^{2+} and Na^{+} in the former areas, replacing Cs ions adsorbed to soil particle surfaces. This effect appeared to be particularly strong near the southern coast where deposition of NH_4^+ from transboundary pollution is evident in addition to the marine cations. The effect of precipitation chemistry on the ^{137}Cs mobility however is found to decline with time, indicating that the ^{137}Cs cations are gradually moving to sites where they are more strongly bound. Experiments where identical soil columns, containing ^{137}Cs from Chernobyl and freshly added ^{134}Cs, are exposed to precipitation of different quality in amounts corresponding to ten years of deposition, largely confirm the conclusions from the repeated soil surveys.

REG.L02 (Id: 112)
TRITIUM IN THE RAINWATER AROUND THE NUCLEAR POWER PLANT OF PAKS, HUNGARY

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Tritium in the environment has natural and artificial sources. The formers are the nuclear reactions in the upper troposphere and the lower stratosphere, the latter are the various nuclear facilities (fission and fusion reactors, eTc.). In order to investigate the two phenomena separately it is necessary to know the effect of artificial tritium emitters to their surroundings with respect to distance from the source. In the presented study rainwater was collected around the nuclear power plant of Paks, Hungary, by means of special rainwater collectors. The sampling distances were 400 and 800 m from the centre of the plant. The samples were analyzed for tritium with liquid scintillation counting and some samples with the ^{3}He ingrowth method. Results show that the trace of the tritium plume from the plant is clearly detectable in the rainwater. However, the maximum activity of the rainwater is only around 10 and 5 Bq/L at 400 and 800 m distances, respectively. A Gaussian plume model is fitted to the measured tritium and meteorological data and shows that the influence of the plant on the tritium in rainwater is completely negligible over some kilometers distance.

REG.L03 (Id: 331)

POLONIUM (^{210}Po), URANIUM (^{234}U, ^{238}U) AND PLUTONIUM (^{238}Pu, $^{239+240}$Pu) IN THE BIGGEST POLISH RIVERS

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Radionuclides existing in the environment, both natural and artificial, are accumulated in plants and animals and transferred through the trophic chain. They are also transported with river from their caTchments' areas. The activities of polonium ^{210}Po, uranium (^{234}U, ^{238}U) and plutonium (^{238}Pu, $^{239+240}$Pu) were measured in the biggest Polish rivers with tributaries - the Vistula and the Oder - and 3 small Pomeranian rivers. The Vistula unfiltered water samples were collected from November 2002 to November 2003, the Oder and the Pomeranian samples were collected from October 2003 to July 2004 and all were measured using the α spectrometer. The highest quantity of ^{210}Po was transported from the Vistula caTchments area to the Baltic Sea in spring and the lowest in summer, while the Oder carries the highest amounts during spring time. Annually the southern Baltic Sea is enriched in 94.5 GBq of ^{210}Po (73.7 GBq from the Vistula, 14.8 GBq from the Oder and 6.0 GBq from the Pomeranian rivers). In the study, the highest activities of uranium ^{234}U and ^{238}U in the Vistula River samples was observed in spring, the lowest in summer. The activity of uranium in the Oder was different in all analyzed seasons, the lowest however in summer. In all analyzed river samples uranium isotopes ^{234}U and ^{238}U are not in the radioactive equilibrium state and the values of ^{234}U/^{238}U activity ratio lie between 1.03-1.84. Annually the southern Baltic Sea is enriched in about 750 GBq of ^{234}U and ^{238}U from all analyzed rivers. According to the Vistula River the highest quantity of plutonium ^{238}Pu and $^{239+240}$Pu were transported to Baltic See in spring and the lowest in summer. The highest activities of plutonium ^{238}Pu and $^{239+240}$Pu in the Oder River samples was observed in winter, the lowest in summer. Annually the Vistula River, the Oder River and the Pomeranian rivers enrich the Baltic Sea in 22.2 MBq ^{238}Pu and 137.6 $^{239+240}$Pu.

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REG.L04 (Id: 169)

EXPERIMENTAL STUDY AND MODELLING OF ^{137}Cs SORPTION BEHAVIOUR IN THE BALTIC SEA AND THE CURONIAN LAGOON

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Sorption-desorption behaviour of Cs, Pu and Am in the system was studied using data obtained from different sampling campaigns in the Baltic Sea and the Curonian Lagoon in 1999-2009 as well as from short- and long-term kinetic tracer experiments using natural sea or river water and bottom sediments collected in the Curonian Lagoon or the Baltic Sea. Sorption experiments were carried out with particles of various sizes from 0.2 to 50 μm. Samples of suspended particles and bottom sediments collected during these sampling campaigns were fractionated according to the size. Association of ^{137}Cs with solid phase was studied using sequential extraction. In addition, the in situ distribution of ^{137}Cs between water and suspended particles of various sizes was measured and expressed as K_d. K_d values for suspended particles of > 0.2 μm size ranged from 1000 to 5500 mL/g in the near shore zone of the Baltic Sea and from 1400 to 20000 mL/g in the Curonian Lagoon. The part of ^{137}Cs bound to 0.2-1 μm particles changed from 10% in the near shore waters to about 80% in the open sea. For all studied radionuclides Cs, Pu and Am an increase in mass activities with decrease of bottom sediment particle sizes was determined. Data obtained from tracer kinetic sorption experiments with ^{134}Cs and bottom sediment fractions of different grain size were used for finding a suitable kinetic sorption model, kinetic constants and the corresponding equilibrium K_d values. It has been found that the modelled data best conform with the mechanism of ion diffusion through the so-called inert layer on the surface of the sediment particles. Kinetic sortion experiments with Pu(V) and Pu(IV) at trace initial concentrations were performed with the natural seawater and bottom sediments with the aim of better understanding of plutonium behaviour in the Baltic Sea. Solvent extraction techniques (using TTA, HDEHP, DBM and PMBP) were employed to characterize the oxidation states of the formed plutonium species in the liquid and solid phases. Modelling is in progress.

REG.L05 (Id: 11)
NATURAL RADIONUCLIDES IN SEDIMENTS AND ROCKS FROM ADRIATIC SEA

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During the International Scientific Cruise to Adriatic and Ionian Seas organised by the International Atomic Energy Agency (IAEA), sediment and rock samples were collected at different locations. Sediments were sampled with grab corer at six locations in the middle and south Adriatic Sea. Rocks were collected on three islands (Palagruža, Brusnik, Jabuka) in the Adriatic Sea. In the samples, natural radionuclides ^{238}U, ^{234}U, ^{232}Th, ^{230}Th and ^{226}Ra were determined. Samples were first dried, crushed and homogenised. After that, radiochemical separation procedures were applied. After radiochemical separation, samples were measured by α spectrometry system equipped with PIPS (passivated implanted planar silicon) detectors. Activity concentrations of natural radionuclides in samples ranged from 11.7 to 27.0 Bq/kg for ^{238}U, from 7.9 to 28.1 Bq/kg for ^{234}U, from 13.2 to 31.9 Bq/kg for ^{232}Th, from 17.1 to 40.9 Bq/kg for ^{230}Th and from 8.3 to 52.3 Bq/kg for ^{226}Ra. In the presentation, the obtained values are discussed in detail and compared with results of similar investigations carried out elsewhere.

REG.L06 (Id: 345)
RECENT TRENDS IN RADIOMETRICS AND MASS SPECTROMETRY TECHNOLOGIES - SYNERGY IN ENVIRONMENTAL ANALYSES

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The dominant development in the radiometrics techniques has been the utilisation of large HPGe detectors in underground laboratories with anti-cosmic or anti-Compton shielding for the analysis of short and medium-lived radionuclides in the environment. They have included applications of high efficiency HPGe detectors (up to 200% relative efficiency to a 75 mm diameter, 75 mm long NaI(Tl) detector), often operating at least a few tens of metres underground, where the nucleonic component of cosmic rays is reduced by several orders of magnitude. In the mass spectrometry sector, applications of Inductively Coupled Plasma Mass Spectrometry (ICPMS) and Accelerator Mass Spectrometry (AMS) for the analysis of long-lived radionuclides in the environment are the most important recent achievements. These developments in both sectors did not only considerably decrease the detection limits for several radionuclides (up to several orders of magnitude), but they also enable to decrease sample volumes so that sampling, e.g., of the water column or sediments can be much easier and more effective. A comparison of radiometrics and mass spectrometry results for the analysis of radionuclides in the environment shows a reasonable agreement - within quoted uncertainties, for wide range of activities and different sample matrices analysed. ^{137}Cs, ^{3}H, ^{14}C, ^{129}I and Pu isotopes water profiles recently obtained for the Indian Ocean will be discussed in detail.

REG.L07 (Id: 270)
RADIONUCLIDES IN DRINKING WATER - AN OVERVIEW: THE EU DIRECTIVE, ANALYSIS, THE AUSTRIAN STANDARD, GEOGRAPHICAL DISTRIBUTION, REMOVAL AND WASTE PROBLEMS

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Drinking water is the most important food, therefore special care has to be taken for its purity. The European Union has issued a Directive on the Quality of Drinking Water, which also contains a chapter on radionuclides. The details will be discussed. With rare exceptions only Naturally Ocurring Radioactive Material (NORM) is of importance for the dose from drinking water. NORM usually cannot be determined by putting a sample on the Ge-detector. Radiochemistry is needed. New methods have been introduced and the traditional measurement of ^{222}Rn and ^{226}Ra has been extended to other radionuclides like ^{228}Ra, ^{210}Po and ^{210}Pb. The Austrian standard on compliance with the EU indicative dose will be presented and recommended to be used with country specific adaptions. In this context the geographical distribution of radionuclide concentrations in drinking water are of importance and examples will be given. Finally the question of purification of water arises. The results of a EU-project on removal of NORM (TENAWA) will be presented as well as the logical question of (radioactive) waste from the purification.

REG.L08 (Id: 55)
NATURAL RADIONUCLIDE CONCENTRATIONS IN AUSTRIAN MINERAL WATERS

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As an EU country, Austria has to meet the European Community guideline concerning waters for consumption; here a Total Indicative Dose of 0.10 mSv per year for all nuclides with the exception of tritium, ^{40}K, radon and radon progeny, is fixed. In our investigation we measured the radium isotopes ^{226}Ra and ^{228}Ra, the uranium isotopes ^{248}U and ^{234}U, as well as ^{210}Pb and ^{210}Po; although the latter two are radon daughters and therefore exempted from the guideline, they can contribute considerably to the committed dose due to their higher dose conversion factors. 1-1.5 L samples of brands from all over Austria were collected from different stores. Radium isotopes were separated by filtering the acidified and degassed mineral water through a Ra extraction disk containing an ion exchange resin [1]. Radium was eluted from the filter with EDTA. The EDTA solution was mixed with a scintillation cocktail and then counted with a low-level counter (Quantulus 1220) using pulse-shape analysis. This procedure collects also the ^{210}Pb, which can be seen in the β-spectrum close to the ^{228}Ra peak. ^{210}Po was spontaneously deposited from the remaining solution onto Cu planchets together with a ^{208}Po tracer and measured α-spectrometrically with a PIPS detector [2]. After adding ^{232}U as a spike uranium was separated by anionic exchange and also measured α-spectrometrically after microprecipitation with neodymium fluoride [3]. From our measured activity concentrations we will give an estimate of the radiological impact of mineral water on consumers. Generally, the calculated annual doses to the adult members of the public are far below the Total Indicative Dose of 0.1 mSv per year.

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REG.L09 (Id: 47)
RADIOLOGICAL CHARACTERIZATION OF DRINKING AND MINERAL WATERS IN SLOVENIA

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It is well known that water contains dissolved radionuclides from uranium and thorium decay chains. Their concentrations are variable and depend on the nature of the aquifer rock types and the prevailing lithology. Quantitative information about the activity concentrations of critical α emitting radionuclides in the food and drink is important in the study of cumulative radiation effects on human beings. Measurements of their levels in drinking water are therefore important to estimate the potential exposure of the public. Due to postulation that the dose coefficient is always related to a specific radionuclide the aim of our study was to determine the activity concentrations of ^{238}U, ^{234}U, ^{226}Ra and ^{210}Po in drinking and mineral waters from different geological or lithological background areas all over the country. For determination of investigated radionuclides α-particle spectrometry was applied. On the basis of radionuclide activity concentration the contribution of each particular radionuclide to internal radiation doses from drinking and mineral water for different groups of people (children, adults) was assessed.

REG.L10 (Id: 276)
RADIATION AND ENVIRONMENTAL MONITORING AT THE SITES FOR THE RW AND SNF TEMPORARY STORAGE IN RUSSIA

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In the light of the operation life termination of the Navy nuclear submarines and their technical service vessels, and because of international obligations of the Russian Federation to reduce armaments, the intensive decommissioning has started since the latter 1980-s. The decommissioning rate was higher than these submarine dismantlement rate limited by the available abilities of the industrial infrastructure. Therefore, radioactive wastes were accumulated at the sites for the RW and SNF temporary storage (STS), like a shot. From this perspective, ex-Navy shore technical bases located in Murmansk region (Andreeva Bay and Gremikha village) and at the Primorsk Territory (Sysoeva Inlet) are the subjects of the highest attention. To gather full information on the current radiation situation, independent in relation to the regulatory bodies, the FMBC specialists carried out radiation and hygienic monitoring. It included dynamic observation both of the radiation-and-hygienic situation parameters and doses to the public living close to the radiation hazardous facilities - STS in Murmansk region and Primorsk Territory. During the expedition travels, samples of environmental media, local foodstuffs and drinking water have been collected. Radiochemical analysis and γ-spectrometry methods were used in the sample examination. The findings of radiation and environmental monitoring confirm considerable exceeding (in comparison with normal background values) of man-made radionuclide contents in seawater, seaweeds, bottom sediments, vegetation and soil at local parts of the coastal stripe of the STS health protection zone. The radionuclide sorption examination in soil and ground water permits to assume effective migration from the contaminated areas via ground water and following radioactivity intake by the marine media at the off-shore water area. Taking into account further contamination of the STS area, observation of the radiation situation dynamic change should be continued both under regular operation mode and during the SNF and RW removal operations.

REG.L11 (Id: 266)
MODEL COMPOUNDS TO UNDERSTAND BACTERIAL SURFACE FUNCTIONALITY

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Of the many factors that influence the transport of actinides in the environment, microorganisms remain among the least understood and most difficult to study. Through a multitude of interactions, bacteria can play a significant role in both the environmental mobilization and immobilization of actinides. This presentation will give an overview of the current status of the field that will be illustrated with example from the literature and with our studies to understand these interactions between a bacterial surface and heavy metal complexation. We are studying a class of single and multifunctional polyelectrolytes as model systems such as propionic acid, 3,3,3-trifluoropropionic acid, phosphoenol pyruvate (PEP), phosphonoacetic acid (PAA), and 3-phosphonopropionic acid (PPA). PEP, PAA, and PPA possess two bifunctional aliphatic organic ligands of different chain lengths, containing each one carboxyl and one phosphate functional group. We are reporting our complexation study of Cm(III) and Eu(III) by time-resolved laser fluorescence spectroscopy (TRLFS) with these ligands. The interactions between species of curium(III)/europium(III) and the various model ligands were investigated as a function of ligand concentration and pH. Thermodynamic stability constants and selected density functional calculations describing the energetically most likely complex configurations will be discussed.

REG.L12 (Id: 102)
COMPLEXATION OF U(VI) WITH NITROGEN AND PHOSPHOROUS CONTAINING LIGANDS IN AQUEOUS SOLUTION

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The long-term safety assessment of nuclear waste disposals requires detailed knowledge of the transport and interaction behavior of actinides within the technical, geotechnical and geological barriers. In the case of accidental release of radionuclides into the surrounding environment, their migration behavior can be effectively influenced by the ambient conditions like pH, temperature, ionic strength and especially the presence of inorganic and organic complexing ligands, e.g. bioligands like humic acids, amino acids or phospholipids. Due to their high complexing capacity towards metal ions these ligands may influence the mobility of actinides. We have studied the U(VI) complexation with nitrogen and phosphorous containing model ligands in order to simulate the functionalities of bioligands. The aim of this work is to determine the influence of various nitrogen and phosphorous containing functional groups on the U(VI) complexation and to evaluate their contribution in comparison to oxygen-containing functional groups. As nitrogen-containing ligands we used the biologically important substances anthranilic acid (AA) and nicotinic acid (NA). Furthermore, phenylphosphonic acid (PPA) was used as aromatic phosphorous-containing ligand. The complexation studies in aqueous solution were performed as a function of pH using time-resolved laser-induced fluorescence spectroscopy (TRLFS). All ligands show a complex formation with U(VI) under the given experimental conditions ([U(VI)] = 0.05 mM, [ligand] = 0 - 0.5 mM, pH 2-4, I = 0.1 M, T = 25 °C). The binding of U(VI) by AA and NA takes place via the carboxylic group of the ligands. It can be concluded that oxygen containing functional groups dominate the U(VI) complexation in the investigated pH range. The nitrogen functionalities of the ligands play only a subordinate role. AA forms a 1:1 complex, with a corresponding stability constant of $\log \beta_{11} = 3.14 \pm 0.17$ [1]. The formation of 1:1 and 1:2 complexes was detected for the U(VI)-nicotinate system ($\log \beta_{11} = 3.73 \pm 0.30$, $\log \beta_{12} = 7.46 \pm 0.17$ [1]) as well as for the U(VI)-phenylphosphonate system ($\log \beta_{11} = 3.58 \pm 0.17$, $\log \beta_{12} = 6.81 \pm 0.10$). At high U(VI) concentrations (1 mM), a precipitation of the formed complexes was observed for the U(VI) complexation by AA and PPA. The precipitates were analyzed with FT-IR spectroscopy and X-ray diffraction. To study the influence of different functional groups on the complexation of actinides in different oxidation states, comparable complexation studies are performed with Am(III).

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REG.L13 (Id: 179)
MOLECULAR CHARACTERIZATION OF THE SPECIATION OF URANIUM ASSOCIATED WITH THE MARINE BACTERIUM IDIOMARINA LOIHIENSIS MAH

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The present study describes the speciation of uranium associated with Idiomarina loihiensis MAH1, a bacterial strain with biomineralization potential abilities isolated from the Alboran Sea in the west side of the Mediterranean Sea. The interactions of uranium with this marine bacterium was studied both in $NaClO_4$ solution as well as in sea water using a multidisciplinary approach combining X-ray Absorption Spectroscopy (XAS), Transmission Electron Microscopy

(TEM), Energy Dispersive X-ray (EDX) analysis and Time-Resolve Laser-Induced Fluorescence Spectroscopy (TRLFS). The results indicated that the speciation of uranium associated with the cells of the strain studied is highly dependent on the speciation of the background electrolyte used ($NaClO_4$ solution and sea water). In $NaClO_4$ solution (at U concentration of 0.5 mM, pH 4.3), EXAFS spectroscopy analysis indicated that the bacterial cells coordinated U through phosphate groups in a monodentate fashion mode and carboxyl groups in a bidentate binding mode. TRLFS results corroborate the EXAFS analysis showing the main implication of phosphate groups in the coordination of U. The uranium bacterial complexes were located within the extracellular polysaccharides (EPS), at the cell wall and some accumulates were observed intracellularly as was demonstrated by TEM analysis. However, in sea water and at U concentration of $5x10^{-4}$ M (environmentally relevant concentration), TRLFS analysis indicated that the marine bacterium precipitate this radionuclide as U carbonate mineral phases. In addition, a part of the metal was coordinated to phosphate groups. TEM analysis showed that the accumulated U was located only at the cell surface as electro dense precipitates. The results of this study will help to understand the role of microbial process on the transport and mobility of radionuclides in the Alboran Sea as it is the only connection between the Mediterranean Sea and the Atlantic Ocean, and where the transportation of radioactive waste and the traffic of nuclear submarines are very intense.

REG.L14 (Id: 13)

POLONIUM-210 AS A TRACER OF ENERGY TRANSFER IN MARINE FOOD CHAINS

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Generally, in marine biota ^{210}Po concentrations are higher in comparison with those in terrestrial organisms. As a consequence, much attention has been paid to polonium-210 (^{210}Po) and its precursor lead-210 (^{210}Pb), the two radionuclides belonging to the natural uranium series. These radionuclides give a major contribution to the internal radiation dose received by man mainly through ingestion of seafood. High ^{210}Po concentrations in internal organs of some crustacean, molluscs, and fish motivated research on the potential biological effects originated by high radiation doses that may be experienced in some marine species. Analyses of ^{210}Po and ^{210}Pb were made in a plethora of marine species from plankton to whales and from the sea shore organisms to those living in abyssal depths. Concentrations of those radionuclides ranged from very low values, of about 0.5 Bq kg^{-1} (wet wt.) in jellyfish, to very high values of about $3x10^4$ Bq kg^{-1} (wet wt.) in the gut walls of sardines, with the general pattern of ^{210}Po>^{210}Pb. ^{210}Po and ^{210}Pb in marine organisms are primarily absorbed from water and concentrated in phyto- and microplankton. Thereafter, these radionuclides are transferred to plankton consumers (herbivores) and to other trophic levels with ingested food, displaying higher yield of ^{210}Po transfer than of ^{210}Pb transfer along marine food chains. Investigation of epipelagic, mesopelagic, bathypelagic and abyssobenthic organisms revealed that ^{210}Po is transferred in marine food webs with transfer coefficients similar to those of energy transfer. ^{210}Po seems to represent the protein transfer in the food chains.

REG.L15 (Id: 32)

IMPACT OF U-MILL TAILINGS OF THE FORMER URANIUM MINE AT ŽIROVSKI VRH (SLOVENIA) ON RADIONUCLIDE ACCUMULATION BY WETLAND PLANTS

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Transfer of radionuclides into the environment is a common phenomenon and due to its potential harmful effect it is of particular scientific interest in the vicinity of uranium mines. Radionuclides, especially uranium and thorium decay products are discharged with U-mill tailings into the soil and water and consequently into vegetation where they accumulate. Radionuclide soil-to-plant transfer was observed in many studies. Uranium mine at Žirovski Vrh in Slovenia, which operated from 1985-1990, processed about 600,000 tons of U-ore. U-milling and mining tailings were deposited at the Boršt and Jazbec sites. Plants that grow in the vicinity of U-mill tailings may accumulate radionuclides in their tissues and thus represent a possible transfer of radionuclides into the food chain. These are so called metal-accumulating plants that developed metal resistance mechanism and may grow on metal-contaminated or metalliferous soil. Plants, such us marsh marigold (Caltha palustris), soft rush (Juncus effusus) and common reed (Phragmites australis) are known as accumulator plants, because they accumulate toxic metals in their above-ground tissues. Other plants, especially grasses, may also grow on metal contaminated soils but do not accumulate metals. Preliminary results of radionuclide accumulation by such plants growing in the contaminated area are presented. A common reed that was grown on the uranium-mill tailings accumulated 0.01 Bq/g d.w. and 0.002 Bq/g d.w. of uranium in leaves and stems, respectively. In contribution, activity concentrations of other nuclides from uranium and thorium decay chains in other plants are also presented and discussed.

REG.L16 (Id: 90)

THE SORPTION PROCESSES OF NP(V) AND U(VI) ONTO METAL OXIDE PHASES. THE FORMATION OF SORPTION COMPLEXES AND MECHANISTIC ASPECTS STUDIED BY IN SITU ATR FT-IR SPECTROSCOPY

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The migration behaviour of actinide ions in the geosphere is generally influenced by sorption processes in aqueous media. These processes occurring at solid/liquid interfaces can be monitored by in situ Attenuated Total Reflection Fourier-transform Infrared (ATR FT-IR) spectroscopy providing structural information on a molecular level.[1] In this work, the sorption of neptunium(V) onto metal oxides was investigated for the first time by ATR FT-IR spectroscopy. From the Np(V) sorption studies on the metal oxides, stable surface species of NpO_2^+ are derived. The type of the sorbed species can be elucidated by a spectral shift (~ 30 cm^{-1}) to lower wavenumbers of the antisymmetric streTching vibration υ3(NpO_2^+) compared to the aqueous species suggesting an inner-sphere complexation. Outer-sphere complexation is found to play a minor role due to the pH independence of the sorption species throughout the pH range 4 - 7.6. The comparative spectroscopic experiments of Np(V) sorption onto TiO_2, SiO_2 and ZnO indicate structurally similar bidentate surface complexes.[2] A detailed insight into the molecular mechanisms occurring during the formation of ternary actinide complexes at the solid/liquid interface is provided upon formation of uranyl carbonato complexes on ferrihydrite (Fh). The influence of the presence of atmospheric carbon dioxide during the sorption processes of the actinide ions was studied by sorption experiments which were carried out under inert gas conditions and in an ambient atmosphere. From the results, the formation of bidentate [Fh•••UO_2•••O_2CO] complexes can be derived under mildly acid conditions which is in agreement from recent EXAFS results obtained from baTch samples.[3] Furthermore, experiments with ^{13}C-labelled carbonate provide an unequivocal assignment of the spectral features reflecting the structural alterations of the carbonate ions upon sorption of uranyl cations onto Fh. From these assignments, the formation of the ternary uranyl complexes can be described at a molecular level.

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REG.L17 (Id: 194)

URANIUM SORPTION ON VARIOUS FORMS OF TiO_2 - INFLUENCE OF SURFACE AREA, SURFACE CHARGE AND IMPURITIES

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Titanium dioxide has properties that make it an excellent substrate for experimental study and theoretical development of adsorption models, including negligible solubility and a near neutral point of zero charge [1]. A number of different forms of Ti-oxide have been used in experimental studies, including hydrous Ti-oxide, anatase, rutile and various commercially available samples that contain a mixture of anatase and rutile. The aim of our work is to investigate uranium sorption phenomena and the influence of surface area, surface charge and impurities for a range of thoroughly characterised Ti-oxide surfaces. We have undertaken uranium(VI) sorption studies on a number of commercially available Ti oxides, some of which were aggressively pre-treated to remove inherent impurities. Characterisations performed on the various Ti-oxides comprised a range of chemical and physical methods including XRD, XRF, ATR FT-IR, chemical assays, BET determinations, and electroacoustic measurements. The sorption of U on these Ti oxides was studied by a baTch sorption method and the effect of pH, ionic strength, mass loading, and U concentration on uranium sorption was also investigated for several of these Ti-oxides. We found that the sorption of uranium (VI) on these Ti-oxides was extremely strong and much greater than many other common environmental sorbents on a surface area basis. Aggressive pre-treatment of one Ti-oxide significantly altered its isoelectric point, but did not appear to significantly impact its sorption behaviour. Differences in sorption behaviour between the various Ti-oxides were related to the surface area of these materials. The data provide insights into the effect of different source materials and surface properties on radionuclide sorption, and will be useful in assessing data obtained in diverse experimental studies involving Ti oxides.

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REG.L18 (Id: 28)
TRANSPORT OF ^{125}I$^-$, ^{137}Cs$^+$ AND ^{85}Sr^{2+} IN GRANITOIDIC ROCKS AND SOIL

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Transport of ^{125}I$^-$, ^{137}Cs$^+$ and ^{85}Sr^{2+} radionuclides in crushed granitoidic rocks and homogenized soil was studied. Two simple methods for calculation of transport parameters of these radionuclides in flow column experiments with groundwater (GW) as transport medium have been described. The first method is based on the assumption of a reversible linear sorption of reactive solutes (radionuclides) on solid phase (with constant distribution, K_d, and retardation, R_{exp}, coefficients), whereas the second one is based on the assumption of a reversible non-linear sorption (characterized with non-linear sorption isotherm, i.e. with non-constant K_d and R_{exp}). Both methods use the experimental breakthrough curves (BTCs), which are constructed using the measured activities at the outlet from the column. The BTCs are fitted with the integrated form of the simple 1-D advection-dispersion equation (ADE) expressed analytically for pulse application of radiotracer to the liquid phase (GW) before entering into columns. In case of the first method, the integrated form of ADE is modified by two correction coefficients, namely, the peak position and peak height coefficients by means of which the very good agreement between experimental and calculated data is usually obtained. The second method is more sophisticated because not only the calculation of the values of retardation coefficients changing during transport is needed, but also the Freundlich equation parameters of non-linear isotherm have to be sought. Both methods were tested in the evaluation of the transport parameters of a given radionuclides in beds of diorite, gabbro, granite and tonalite granitoides and clayey loamy sandy soils. The results of two different approaches have been compared.

REG.L19 (Id: 62)
SORPTION BEHAVIOUR OF CONTAMINANTS IN SOILS: PH AND HUMIC ACID EFFECTS ON SORPTION OF ON CEC OF CLAY SOILS AND MINERALS, AND THE MOBILITY OF Cd, Cs, Ni AND Sr

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The aim of this study is to use predictive modelling to understand and calculate the sorption behaviour of radionuclide contaminants in complex environments such as clay-based soils. The effects of natural organic matter (NOM) on the cation exchange capacities (CEC) of different characterised clay soils (St Bees and Mercia Mudstones, Boom clay and London clay) and their constituent clay minerals will be determined by "doping" the samples with varying amounts of purified humic acid (not exceeding 15%) and measuring the changes in CEC. This will be followed by an assessment of how NOM affects the mobility of radionuclides (^{109}Cd, ^{137}Cs, ^{63}Ni and ^{85}Sr) through the materials using column and baTch techniques. The experiments will be repeated with each of the constituent clay minerals (montmorillonite, kaolinite, illite, chlorite and bentonite) present in the soils using baTch techniques, and the results obtained used to improve the understanding of the separate effects of each mineral on the overall sorption behaviour of the soils. This will lead to testing to determine if the additivity principle holds for these systems. Radiometric techniques and inductively coupled plasma optical emission spectroscopy (ICP-OES) will be employed in the quantification of radionuclide speciation between the phases, and the samples will be characterised by SEM and XRD with the assistance of BGS.

REG.L20 (Id: 98)
VERTICAL AND HORIZONTAL DISTRIBUTION OF ^{137}Cs IN ZONE OF CHERNOBYL CONTAMINATION IN RUSSIA

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Peculiarity of vertical migration of ^{137}Cs was investigated in zones of Chernobyl contamination (Bryanskaya, Orlovskaya, Tulskaya and Kaluzhskaya districts). Vertical distribution of ^{137}Cs on inviolate localities characterized by minimum penetration of the peak of maximal concentration into the depth of about 2-4 cm in dependence of type of soil and intensity of processes of bioturbation. Practically all quantity of ^{137}Cs in inviolate soils is accumulated in the top-most 0-15 cm. The waterlogged soils as well as acidic soils of the pine forests are exclusion of this rule, where ^{137}Cs may migrate into bigger depth. For example, the sandy soils without feature of podzolic processes contain 97% of the radionuclide in the first 5 cm. In dernovo-podzols the first 5 cm contain only about 60% of ^{137}Cs and a second peak of vertical distribution of ^{137}Cs was observed, related with accumulation of radionuclide in the illuvial horizont. Vertical distribution of ^{137}Cs in arable soils and adjacent slopes of dry valleys is formed greatly by processes of erosion and accumulation. Alternation zones of previous erosion and accumulation inside of the arable slope have been observed. On the flood areas of small rivers accumulation of the radionuclide is discovered on the billow near the river-channel, peak of vertical distribution of ^{137}Cs is in the depth of 5-10 cm. In marshy soils a decrease of ^{137}Cs in 0-25 cm depth relatively of soils of higher geomorphological positions is usually observed, probably because of the penetration of the radionuclide into a greater depth. But, under periodically wet conditions of flood area depressions it can form local spots of contamination 5 times higher than in soils of adjacent dry hills.

REG.L21 (Id: 347)

MIGRATION ABILITY OF PLUTONIUM AND AMERICIUM IN THE SOILS OF POLESSIE STATE RADIATION-ECOLOGICAL RESERVE

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One of the most important factors determining the radioecological situation in the terrestrial ecosystems is the radionuclide species in a soil medium. Radionuclide forms determine the processes of their entrance into the soil solutions, redistribution in soils, migration to the surface, ground and underground waters and spreading outside of the contaminated area. The present work is devoted to investigation of physicochemical forms and migration ability of plutonium and americium in soils of Polessie State Radiation-Ecological Reserve (PSRER), where located the main part of α-emitting radionuclides of Chernobyl origin. The objects of investigation were mineral and organic soils sampled in 2008 with the step of 5 cm to the depth of 25-30 cm. The forms of plutonium and americium distinguishing by association with the different components of soil and by potential for migration in the soil medium were studied using the method of sequential selective extraction according the modified Tessier scheme. Activities of ^{238}Pu, ^{239,240}Pu and ^{241}Am in the samples were determined by the method of radiochemical analysis with α-spectrometer radionuclide identification. A vertical radionuclide distribution in soils and the total radionuclide reserves in the soil profiles were established. Intensities of vertical radionuclides' migration in the soils were estimated. It was shown that the main part of plutonium and americium is in the 0-20 cm soil layer, more often in 0-10 cm layer. Location of the radionuclide weighted mean quantity in the soils is at the depth of 3-15 cm from the soil surface. The average rate of vertical radionuclide migration of this quantity varies from 0.15 to 0.7 cm y^{-1} and is practically the same for plutonium and americium. The main part of plutonium and americium in soils is in immobile forms. Radionuclide portions in water soluble and reversibly bound forms do not exceed 9.4 % of radionuclide content in the soil. In mineral soil samples, the radionuclide portion in these fractions exceeds the corresponding portion in organic ones. In both mineral and organic soils, the portion of mobile americium is higher than plutonium. The portion of biological available forms of plutonium and americium is 2.7-29 % of total radionuclide content in the soils. The higher portion of biological available forms is characteristic for mineral soil (14-29 %) as compared with that in organic one (2.7-18 %). The reserves of mobile and biologically available radionuclide forms increase with the depth of soils. The increase of radionuclide portions in mobile and biologically available forms promotes the radionuclide entrance into the soil solution and enhancing the intensity of migration processes in the soil-plant system. The main control factors of radionuclide migration in soils under consideration are water regime and presence of radionuclide in composition of fuel particles. These factors could be more affective than radionuclide solubility in the soil waters because of some part of radionuclide transfer with the particles.

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Posters

REG.P01 (Id: 8)

TRANSLOCATION OF RADIOACTIVITY FROM SUBSTRATE TO MACROMYCETES IN SOME MINING AREAS

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The present study follows the extent of translocation of radioactivity from substrates of uranium and sulphur mining areas to the macromycetes spontaneously occurred during 2008-2009. To this purpose, radioactivity (gross α+β, ^{226}Ra and ^{137}Cs) of both macromycetes and their substrates was measured. The results obtained were confirmed by FT-IR spectroscopy, evidencing the presence of characteristic bands at about of 910 cm^{-1}, corresponding to the asymmetric streTching vibration of the uranyl unit and to the interaction between the UO_2^{2+} ions and the group belonging to various cellular components.

REG.P02 (Id: 14)

RADIOACTIVITY IN SOILS AND HORTICULTURE PRODUCTS NEAR OLD URANIUM MINING AND MILLING SITES

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Uranium in Portugal was mined mainly in small mines in the Centre-North of the country and lasted for most of the 20th century. Uranium production was discontinued in 2001, but most of the former mining and milling sites were not remediated yet and mining and milling waste heaps remain in surface exposed to weathering. In villages near old uranium mines, soils are used for horticulture production and to provide pasture for livestock grazing. Soil samples as well as agriculture products from the areas around former uranium mines were analyzed for α emitting radionuclides. Samples from other areas far from uranium mining sites were also included in the survey as reference sites. Results of uranium series radionuclides showed that soils of regions with different geology may contain very different baseline radionuclide concentrations. Soils near uranium mining waste piles in several cases displayed enhanced radionuclide concentrations resulting from soil contamination by mining waste. Concentrations of radionuclides in soils and vegetables of reference areas displayed values similar to those near uranium mining sites in the same geological province, indicating that enhancement of radionuclides in the terrestrial food chain, with the exception of ^{226}Ra, is generally not high. Concentrations in soils and horticulture products of these uranium areas in the uranium producing region were generally two orders of magnitude higher than baseline concentrations measured in soils and vegetables in a sedimentary region of the South of Portugal.

REG.P03 (Id: 21)

METHODS AND RESULTS OF THE MAIN NEGATIVE FACTORS OF KOSHKAR-ATA TAILING POND WITH IMPACTS ON ENVIRONMENT

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Ecological situation of Mangystau oblast (Kazakhstan) started aggravating in 1990s with exploitation of uranium ore, oils, mineral raw materials and building of chemical industry in Mangyshlak without paying due attention to environmental issues. According to available data the worsening is connected to number of ecologically unfavorable problems. The KOSHKAR-ATA tailing pond, representing the drain-free settling pool for industrial, toxic, chemical and radioactive wastes, as well as for ordinary domestic drains, is 5 km northward to Aktau (Mangystau oblast), which is situated on the shore of the Caspian sea. Industrial, toxic and radioactive waste, solid sediments of unpurified ordinary domestic drains from a part of the Aktau dwelling region have been drained into the tailing pond since 1965 and have been stored there up to now. Thus, continuous and long-term contamination of the lands around the KOSHKAR-ATA tailing pond and their vegetation unambiguously forces to consider regional ecological situation as critical. That is why it is very important to take immediate measures in order to reduce sequences, eliminate the reasons and sources of contamination and restore natural characteristics of the environment. Field investigations are performed in the scope that assures detailed radiometric surveying of the shore zone of the KOSHKAR-ATA tailing pond and its coast. The field investigations included: mapping of current outline of the coast-line/water edge of the toxic waste storage; footpath γ-surveying of the shore exposed surface (EDR was measured in 150 points) and mapping of the γ-field distribution; assessment of the volume activity for radon over the shore exposed surface of the tailing pond; detailed γ-surveying in the vicinity of surface burial of radioactive waste (RW); monitoring of aerosols at 10 stations; species identification and taking of samples of vegetation at the waste storage in places where soil fill had been previously performed; estimation of the radionuclide accumulation rate by vegetation and their further wind transfer. Analysis showed that there is an increased content of radionuclides as thorium-234, radium-226, lead-214, bismuth-214, lead-210, uranium-235, thorium-227 in bottom sediment samples. Presence of various forms was studied for isotopes of thorium, radium, ^{210}Pb, ^{238}U. Lead-210 is considered as one of the most biologically hazardous natural radionuclides, even more radio-toxic than ^{90}Sr. Considerable part of ^{210}Pb (up to 53%) is associated with acid-soluble form what demonstrated its high migration abilities. Mobility series for ^{210}Pb (in %) is as follows: acid-soluble forms (40.2%) > exchange forms (11.7%) > water soluble forms (0.4%). Analysis of obtained results shows that there are higher contents of lead-210 in samples taken within the sectors; this is particularly so for the sector "Caspii."

REG.P04 (Id: 29)

DETERMINATION OF CATION EXCHANGE CAPACITY OF FUCOIDIC SANDS FOR Cs^+ AND Sr^{2+} UNDER DYNAMIC COLUMN CONDITIONS

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In the framework of the development of remediation scenario of underground areas affected by chemical mining of uranium in the locality of Stráž pod Ralskem a complex geochemical and transport model is proposed, which comprises interaction of rocks and groundwater affected by acid leaching. The chemical remediation processes also require the knowledge of the properties of the affected rocks. One of the important rock constituents are fucoidic sands constituting significant part of the cenoman aquifer. Therefore, it is of great importance to determine, among others, their cationic exchange capacity (CEC) toward univalent and divalent cations. In this paper results on the study of sorption and desorption behavior of Cs^+ and Sr^{2+} on column of fucoidic sands under dynamic flow conditions are presented and their CEC for these two cations are determined. The determination of CEC is based on the construction of respective breakthrough curves using ^{137}Cs and ^{85}Sr radionuclides as isotopic indicators in laboratory experiments. The samples were taken from several parts of the bore hole in the area of interest. Undisturbed cores of 5 cm in diameter and 10 cm long were put in the glass columns and the cores were perfectly tightened using acrylate resin. For the study of sorption/desorption cycle the so-called background groundwater was applied. The concentration of 10^{-6} mol/dm^3 of Cs^+ and Sr^{2+} in liquid phase individually was established using neutral salts of $CsNO_3$ and $Sr(NO_3)_2$, resp. The groundwater was introduced at the bottom of the columns by a multi-head peristaltic pump at about 4 cm^3/h of a constant flow-rate. The results show that the CEC capacity of the investigated fucoidic sands for ^{137}Cs and ^{85}Sr is 0.1-1.5 µmol/100g and 0.05-0.5 µmol/100g, respectively, in dependence on the evaluation of corresponding breakthrough curves. Some differences in the behavior of the cores during the experiments have also been explained.

REG.P05 (Id: 36)

ORIGIN OF ^{129}I IN THE ENVIRONMENT OF SLOVENIA

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^{129}I is considered as a global pollutant and its role as a global tracer to follow the dissemination of radionuclides from a source point such as nuclear fuel reprocessing plants increases. The natural ^{129}I/^{127}I isotopic ratio of the order of 10^{-12} was significantly influenced by releases of anthropogenic ^{129}I to the environment. The ratio of ^{129}I/^{127}I in the marine environment has increased to 10^{-11} - 10^{-10}, and in the terrestrial environment to 10^{-9} - 10^{-7}, even 10^{-6} in the vicinity of nuclear fuel reprocessing plants. Anthropogenic ^{129}I predominates in the biosphere, soil and in upper layers of the oceans, therefore it can be expected that the isotopic ratio ^{129}I/^{127}I is increasing in these compartments of the ecosystem. At the Josef Stefan Institute we have developed a radiochemical neutron activation analysis method (RNAA) for the determination of ^{129}I in environmental samples and the detection limit of the method is $5x10^{-14}$ g ^{129}I/g of solid sample. The developed method was used to trace ^{129}I in the environment of Slovenia. From marine environment seawater, alga, thorny oyster and sediment samples were analyzed and from terrestrial environment precipitation, soil and pine needles. The results found were in the range from 7.2 to 120.8 x 10^{-7} Bq/kg for precipitation, up to 75 x 10^{-4} Bq/kg for soil, from 0.1 to 2.12 x 10^{-4} Bq/kg for seawater, from 7 to 62 x 10^{-4} Bq/kg for thorny oyster and from 17 to 73 x 10^{-4} Bq/kg for brown alga Fucus virsoides (Donati) J. Agardh., which is endemic to the Adriatic Sea. The content of ^{129}I in marine sediment and pine needles samples analyzed till now was below the detection limit of the method. The data of this study represent a survey of ^{129}I in the environment of Slovenia. The most likely source of ^{129}I are nuclear fuel reprocessing plants in La Hague and Sellafield, which are known to be the major sources of ^{129}I in the environment of North Europe. ^{129}I is transferred to the atmosphere and washed out to the marine and terrestrial environment of Slovenia by precipitation, so it is of atmospheric-precipitation origin.

REG.P06 (Id: 37)
BACKGROUND LEVEL OF RADIOACTIVE ISOTOPES AROUND A L/ILW DISPOSAL FACILITY BEFORE IT STARTED OPERATION AT BÁTAAPÁTI, HUNGARY

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The new Hungarian National Radioactive Waste Disposal Facility was established in the granite of the Mórágy Block Mountains to store low and intermediate level radioactive waste (L/ILW) originating from the Paks Nuclear Power Plant. The safe operation of the radioactive waste disposal facilities requires a long-term environmental monitoring to gain information about the radiation properties of the site together with the radiation exposure of the personnel and the environmental media. Before the start of the operation of the facility the environmental characteristics of the site and its vicinity, i.e. the so-called zero level was determined. The determination of the zero level of the nuclear facilities is inevitable as the evaluation of the measurement data in the course of the operation should be related mainly to this zero level. In the course of the measurements the monitoring of the environmental elements, i.e. air, soil, water (springs, streams, precipitation), fall-out, wash-out, plant and animal samples, was performed from several points of view. From the samples collected radiocarbon, tritium, strontium-90, γ emitters, gross α, gross β and field γ measurements were carried out. The results are obtained before of the operation of the facility, they can approximately be considered as the Hungarian background data.

REG.P07 (Id: 38)
TESTING AUTOMATIC GROUNDWATER SAMPLING UNIT BY THE ISOTOPE ANALYTICAL AND DISSOLVED ION TESTS

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Automatic water sampling unit was developed in Herteleni Laboratory of Environmental Studies of MTA ATOMKI for monitoring the radioactive emission from nuclear facilities into the groundwater. The efficiency of existing and renewed geometry units and the reproducibility of survey data have been examined in the course of this work. A testing method was developed for this purpose, and ion binding efficiencies of ion exchange resins were analysed for different ion concentrations. These efficiencies have to be taken into consideration when we estimate the amount of the contamination transferred into the groundwater on the basis of the proportion of ions gained back from the resin. The model tests were executed under controlled laboratory circumstances. These circumstances were tried to be formed into facts true to nature. It has been found during the chain of tests that the sampling unit is suitable for well reproducible sampling. It can be stated that all tested geometrical lay-outs are usable and work with proper efficiency in small/low range of concentration as well. Taking the different efficiencies into consideration the activities can be corrected for every element if necessary. A correcting factor should be introduced during the ^{14}C anion exchanging sampling because samples taken by exchanging have systematically lower radiocarbon content than the actual one. We are working on continuing the tests. The γ activity measurement of existing gained back cation samples and giving the exact value of ^{14}C correction can give more reliable picture and direction to developing existing and possibly new systems.

REG.P08 (Id: 50)
RADIOCESIUM ACTIVITY CONCENTRATION MEASUREMENTS IN GAME MEAT AS A TOOL FOR MONITORING RADIOACTIVE CONTAMINATION IN WILDLIFE AND ENSURING RADIOLOGICAL SAFETY FOR CONSUMERS

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An increased level of cesium radioisotopes was noted in many foodstuffs after the Chernobyl disaster. The radiocesium activity concentrations may remain high in products such as game meat, wild mushrooms, and forest berries. Therefore, an official monitoring of radiocesium in game meat was established by Polish Veterinary Inspection. Generally, consumption of game meat in Poland is low. However, in hunters' families this sort of food can contribute a large portion of meat to a diet. Moreover, some quantities of Polish game meat are exported to many countries. Meat samples of wild boar, red deer, and roe deer were taken at game processing facilities by veterinary inspection officers and then transported to laboratories for analyses. Radiocesium activity concentrations were determined by γ-ray spectrometry using scintillation (NaI(Tl)) detectors. The ^{134}Cs activity concentrations were negligible (below MDA) in all the samples analysed. Generally, the ^{137}Cs activity concentrations varied between and among the animal species even in the same region ranged from MDA values to some hundreds Bq/kg. The effective dose, which general population in Poland received from consuming game meat, may be considered very low with the exception of hunters' families. Assuming the mean annual game consumption of 20 kg in hunters' families and the mean effective dose of 0.23 μSv per kg (two year studies), an individual in the most exposed group may receive the effective dose of 4.6 μSv. In conclusion, consumption of local game meat is safe in terms of contamination with radiocesium and radiological protection.

REG.P09 (Id: 65)
DETERMINATION OF STRONTIUM-90 IN FOODSTUFF

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The Nuclear and Technological Institute (ITN) has the legal responsibility to carry out the environmental radiological monitoring at a national level. This survey was planned to provide relevant information on radioactivity levels in different components of the ecosystem (atmosphere, aquatic and terrestrial environments). As concerns the terrestrial environment the strontium-90 (^{90}Sr) concentrations in food samples have been determined by analyzing milk, complete meals, meat and vegetables. Strontium-90 (^{90}Sr) is a radioactive isotope of strontium, with a long half life (28.8 years) that could be taken by man through food chain transfer due to its biochemical similarity with calcium. A method based on the separation of ^{90}Sr by extraction chromatography and β determination by Liquid Scintillation Counting technique was used for strontium analysis in food samples. The trials were carried out using 7g of ash samples, and then dissolved in concentrated nitric acid. Following additional precipitation steps the sample is loaded onto a column with 3 grams of commercial Sr-resin. This resin is specific to Sr ions and enables a rapid and simple separation of strontium from calcium, potassium and many other elements. The column is rinsed with 8M HNO_3 and 3M HNO_3 and after this two washing steps, the strontium retained in the column is stripped at the end with 0.05M HNO_3. The strontium solution eluted is collected in a beaker and the chemical yield was determined by gravimetric method. The strontium precipitate is dissolved in 8 mL of 0.1M HCl and added to 12 mL of the Ultima Gold LLT scintillation cocktail. The ^{90}Sr activity was determined after ^{90}Y ingrowths in the channel region 250-800 keV using a counting time of 120 minutes. Validation trials to evaluate the accuracy and the precision of the method were performed using ^{90}Sr spiked samples. The method has been applied to the ^{90}Sr determination of complete meals, meat and vegetables. Results of validation trials and food analysis will be presented and discussed in this paper.

REG.P10 (Id: 68)
AGE DATING OF THE HOT SPRING WATERS IN KOREA USING NATURAL RADIONUCLIDES

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The use of radioactive isotopes as a groundwater-dating tool plays an important role in assessing the dynamics of groundwater systems, essential for the characterization of water resources and planning its exploitation. In hydrogeological studies the application of radioisotopes with a short half-life (below 100 year) is limited in dating old groundwater. However, they can be extremely helpful in solving another type of questions such as the identification of mixing between old and young groundwater systems. Within the environmental radioisotopes, ^{3}H cannot be detected in waters more than approximately 50-60 years old due to the short half-life ($t_{1/2}$ = 12.32 year). Among the radioactive isotopes with a half-life greater than 1 ka, ^{14}C ($t_{1/2}$ = 5730 year) represents the most important tool in groundwater dating. This radioisotope is present in the atmosphere, soil, aquifer matrix, eTc. Using these two radioisotopes, the age of the hot spring waters in Korea were estimated. Also some other natural radioisotopes, Ra and U, were analyzed to know the characteristics of the hot spring waters. Most hot spring waters are found in granite region in Korea and the Mesozoic granites at the southern part in the Korean Peninsula are divided into two groups, Jurassic Daebo-granite and Cretaceous Bulguksa granite. High temperature hot springs with more than 40 °C occur in the above granite areas. Therefore, fifteen hot spring waters were collected and the contents of ^{3}H, ^{14}C and other radionuclides were determined. Tritium concentrations in hot spring waters were very low, therefore, they were concentrated using the Ni-Ni electrolytic enrichment method. They were analyzed using low background liquid scintillation counter. Dissolved carbonate was precipitated with $BaNO_3$ to $BaCO_3$ form and it reacted with phosphoric acid to produce CO_2. Finally, CO_2 was converted to graphite. After then, it was analyzed using AMS. In most hot spring samples tritium could not be detected and its concentrations ranged <0.5-1.31 TU. And ^{14}C contents ranged 2.62 - 94.13 pMC(%). From the ^{3}H and ^{14}C analysis, we found that some hot springs are mixed with recent groundwater and that hot spring water aged from 490 years to 33680 years.

REG.P11 (Id: 79)
USE OF SELECTED CHELATING AGENTS TO MOBILIZE RADIOCESIUM FROM THE BODY

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Examinations were carried out to compare the effects of Prussian Blue (PB), Ca-gluconate, and Na-citrate on removal of radiocesium from male Wistar rats. The animals were randomly divided into 5 groups of 10 animals each. Radiocesium was administered by intragastric intubation to all tested rats for 5 consecutive days with a daily dose of 20 kBq. Animals in group 1 (the controls) were administered only cesium-137 chloride whereas animals in groups 2 to 5 were subjected to the same radiocesium schedule as rats in group 1 and then treated as follows: groups 2 and 3 were given PB by gastric intubation at a dose of 20 mg in 0.5 mL of distilled water 1 hr or 1 hr and 5 hr, respectively, after cesium-137 contamination whereas rats in group 4 after each PB treatment were subsequently injected ip for 5 consecutive days with a daily dose of 20 mg of sodium citrate and calcium gluconate in 0.5 mL of distilled water. Rats in group 5 were treated similarly to rats in group 4 but without PB administration. Animals were killed 6 days after termination of the experimental protocol. The whole-body retention of cesium-137 was measured in a well type scintillation counter. Mean values were compared by Student's t-test. Prussian Blue, Ca-gluconate and Na-citrate failed to produce any unfavourable effects on body weight gains and organ to body ratios of the liver, kidneys, heart, and testes in all rats tested. Control rats retained 71.2% of administered radiocesium in the whole-body. Fivefold and tenfold administration of PB or fivefold treatment with PB plus 5 injections of Na-citrate and Ca-gluconate decreased significantly the whole-body retention of radiocesium to 49.7%, 38.4%, and 43.2%, respectively. On the other hand, fivefold treatment with Na-citrate and Ca-gluconate (witout PB treatment) failed to decrease radiocesium retention in comparison to that in the controls. The present results showing a high efficiency of Prussian Blue in removal of radiocesium from animal bodies are in accordance with earlier reports of others. On the other hand, no effects of Ca-gluconate and Na-citrate injection on radiocasium mobilization from the body were in contrast to the opinion that citrate and gluconate enhance the removal of radiocesium from animals.

REG.P12 (Id: 89)

SPECIATION OF ^{129}I AND ^{127}I IN SEAWATER PROFILES FROM THE KATTEGAT AND BALTIC SEA

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Long physical half life, long residence time in the marine environment and continuous releases from nuclear fuel reprocessing plants make ^{129}I a suitable tracer for the study of marine geochemical cycle of stable iodine and conversion mechanism of iodine species by speciation analysis of ^{129}I and ^{127}I. Up to date there are few data on chemical speciation of ^{129}I in seawater and no ^{129}I chemical speciation analyses in seawater profile were reported. The converting mechanism of iodine chemical species is still unclear. In the present work, depth profile samples of seawater collected from the Kattegat Belt Sea and Baltic Sea are analyzed for ^{129}I, ^{127}I as iodide, iodate by using anion exchange chromatography coupled with accelerator mass spectrometry (AMS) and inductively coupled plasma mass spectrometry (ICP-MS), respectively. A relatively high concentration of ^{129}I (2-18x10^{10} atom/L) was found in the Kattegat comparing with that in the Baltic Sea (<2 x 10^{10} atom/L). The results show that in the Baltic Sea the concentrations of ^{129}I, ^{127}I increase with depth and salinity, while the same trend does not occur in the Kattegat Basin. In Baltic Sea the predominant species of iodine is iodide while in the Kattegat the ratio ^{129}I$^-$/^{129}IO$_3^-$ is 1-2. Along the water profile, the atomic ratios of ^{129}I$^-$/^{127}I$^-$ and ^{129}IO$_3^-$/^{127}IO$_3^-$ decrease from the northern Kattegat to the central Baltic Sea. Speciation analysis of ^{129}I in seawater profile is indispensable for a better understanding of geochemical cycle of stable iodine and the converting mechanism of iodine species in the marine environment.

REG.P13 (Id: 93)

RADIATION MONITORING METHOD FEATURES OF CONTAMINATION AREAS AGEING IN LANDSCAPE

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The aim of this work is to generalize experimental research data received in zone of Chernobyl trace in 2007 in Bryansk region in order to distinguish automorph, transit and accumulation areas. Variability of ^{137}Cs levels is shown at the 3 experimental sites.

REG.P14 (Id: 96)

DISTRIBUTION OF NATURAL RADIONUCLIDES AND ^{137}Cs IN THE SEDIMENTS OF A MEDITERRANEAN FJORD-LIKE EMBAYMENT, AMVRAKIKOS GULF, GREECE

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Five small sediment cores were collected from the seafloor of Amvrakikos Gulf for the study of the spatial and downcore distribution of natural radionuclides and ^{137}Cs in the sediments. Amvrakikos Gulf is a shallow (< 65m) marine embayment lying on the west coast of Greece having a surface of some 405 km^2 and a coastline length of 256 km. It is separated from the Ionian Sea by a beach barrier and is connected to the sea through a narrow channel, 600 m wide and less than 8 m deep. Amvrakikos Gulf receives the freshwater inputs of the Arachthos (2202x10^6 m^3 yr^{-1}) and Louros (609x10^6 m^3 yr^{-1}) rivers at its northern shoreline. The Gulf is characterized by a general two-layer type of stratification in the water column and is considered as the only Mediterranean fjord. Sediment samples were analysed for specific activities of ^{238}U, ^{232}Th, ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs together with grain size and C$_{org}$. ^{238}U and ^{226}Ra concentrations range from 37.2 to 124.0 and 8.5 to 27.2 Bq kg^{-1} with an average value of 62.1 and 17.9 Bq kg^{-1}, respectively. The concentrations of ^{232}Th and ^{40}K range from 11.2 to 46.5 Bq kg^{-1} and 238 to 892 Bq kg^{-1} with an average value of 36.6 and 710 Bq kg^{-1}, respectively.

The activities of ^{232}Th and ^{40}K reported in the present study are comparable to those of the world average. On the contrary, the reported ^{238}U activities are significantly higher than those of the world average (UNSCEAR, 2000). The value of the ^{226}Ra/^{238}U ratio ranges from 0.09 to 0.66 with an average value of 0.35 showing disequilibrium between the two radionuclides in all sediment samples that can be ascribed to the weathering of phosphorites of Epirus (at the northern margin of Amvrakikos Gulf) as well as to the high phosphate fertilizers inputs due to the intensive application of fertilizers in the surrounding agriculture areas. It is well known that phosphorites and phosphate fertilizers are characterized by high ^{238}U concentrations. The spatial distribution of ^{238}U, ^{226}Ra, ^{232}Th and ^{40}K, in the surface sediments of Amvrakikos Gulf shows a clear increasing eastward trend. Maximum activities were observed at the eastern deep basin, while minimum values were found at the western basin of the Gulf. The man-made radionuclide ^{137}Cs was found in measurable concentrations (5.2-20.2 Bq kg^{-1}) in all surface sediments (0-5 cm). The higher specific activities of ^{137}Cs were found in the deeper sediment layers (8-44 cm) than in the surface layer. The high content of the clay minerals and organic matter found in the sediments of Amvrakikos Gulf play very important role in the high level of ^{137}Cs. Moreover, the elevated specific activities of ^{137}Cs in the deeper sediment layers (44 cm) suggest a high sedimentation rate for Amvrakikos Gulf.

REG.P15 (Id: 105)

NATURAL RADIOACTIVITY IN TUNISIAN AND IN SOME IMPORTED PHOSPHATE FERTILIZERS USED IN TUNISIA

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The radioactivity concentrations of ^{226}Ra, ^{232}Th and ^{40}K in different phosphate fertilizers, triple superphosphate (TSP), diammonium phosphate (DAP), monoammonium phosphate (MAP) and in phosphoric acids produced and used in the agricultural soils in Tunisia were measured. Other imported and used phosphate fertilizers in the country consisting of some NPKs were also subject to radioactivity concentrations measurement. Gamma spectrometry using a high purity germanium (HPGe) detector was used for the measurement. It was found that generally the Tunisian fertilizers contain the highest activity concentrations with the exceptions of ^{40}K. TSP contains the highest concentration of ^{226}Ra (186.2±14.5 Bq/kg) and of ^{232}Th (33.7±3.5 Bq/kg). The highest amount of ^{40}K was found in one type of NPK (9969.5 Bq/kg). The radium equivalent index was calculated for all the analysed fertilizers and was found to be the highest in the imported NPKs samples (the maximum value was about 775 Bq/kg) because of their high content in ^{40}K. For the Tunisian fertilizers the maximum radium equivalent index was about 238 Bq/kg in TSP.

REG.P16 (Id: 107)

FURTHER RESULTS IN SEARCH OF TRANSURANIUM ISOTOPES IN EFFLUENTS DISCHARGED TO AIR FROM NUCLEAR POWER PLANTS

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In this work we present data on transuranium isotopes in effluents (quarterly, since 2006 half-yearly determined activity concentrations of ^{238}Pu, ^{239,240}Pu, ^{241}Am, ^{242}Cm and ^{244}Cm in discharged effluents, annual discharged activities of individual radionuclides) discharged to air from 7 stacks in 2004-2009. In the effluents discharged to air from one stack low-level activities of transuranium isotopes were present throughout the period under study. Transuranium isotopes had been discharged to air from this stack also in previous years since 1996 when defect in the cladding of a fuel element and consequent contamination of the primary circuit occurred. In the effluents discharged to air from another stack transuranium isotopes were present only in some monitoring periods in the years under study. We did not find transuranium isotopes in the effluents of the other stacks up to 2006. The transuranium isotopes in discharged effluents were registered in the second half-year of 2006. In other years (2007-8) especially low-level ^{241}Am was found in these effluents.

REG.P17 (Id: 116)

RADIOECOLOGICAL SITUATION IN AREAS OF URANIUM LEGACY OF UZBEKISTAN

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Central Asia countries were the main supplier of the uranium in former USSR and one of them was the Republic of Uzbekistan. The mining activity and treatment of uranium ores resulted in formation of large amounts of radioactive wastes making an exigent problem with securing radiation safety on the territories of uranium mines, tails and adjacent settlements. In this work, the results of measurements of the radon concentrations on the territories of former uranium mines and nearby populated sites are presented. Electronic units of RRA-01M (Russia) and PRM-145 (Slovenia), acting on semiconductor detector and scintillation cells, respectively, were used for radon measurements. Both units record α particles of daughter radionuclide ^{218}Po. For reliable estimation of the annual efficient doses (AED) of inhabitants and their lifetime fatal risks (LFR) a long time registration of radon α particles by nuclear solid track detectors CR-39 (Italy) has been carried out. Residences and working premises were selected on free basis and radon exposure with CR-39 was monitored for two and three months at summer and winter time. In settlement Yangiabad, volume activity of indoor radon was 60 - 100 Bk/m^3, values of AED of radon for inhabitants 1.7 - 2.8 mSv/year and LFR estimation at (1.3 - 2.3) x10^{-4}. In village Chorkesar in windless and matutinal period radon activity on territory of the mine reached 70-100 Bk/m^3 at a distance of 50-70 cm from ground. Indoor radon level are in the range from 60 to 450 Bk/m^3 and 1.7 - 12.6 mSv/year for AED, (1.3 - 10.5)x10^{-4} for LFR at summer and from 250 to 1200 Bk/m^3 and 7.0 - 33.6 mSv/year for AED, (5.6 - 27)x10^{-4} LFR at winter time.

REG.P18 (Id: 125)
STUDY OF ^{7}Be CONCENTRATION IN THE NORTH OF THE SPAIN

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From the Sun and interstellar spaces of the universe the primary cosmic radiation comes to the Earth. These cosmic rays interact with the nuclei of atmospheric constituents, producing an interaction cascade and secondary reaction and also produce a number of radioactive nuclei known as cosmogenic, ^{7}Be between others as a result of spallation reactions. In Bilbao (north of Spain) there is a high flow rate atmospheric sampler which belongs to the Radiological Surveillance Network of the Autonomous region of the Basque Country. Among the radionuclides present in the air aerosols ^{7}Be is found. The network is in operation since October 2001 and so a long record of ^{7}Be measurements is available. To study the behaviour of ^{7}Be in air, the recorded values have been considered as a succession of data what is usually known as a Time Series. It is made up of 4 components: trend, cyclical component, seasonal component and irregular component. The cyclical component is not fully developed because the studied period (2001 - 2009) is shorter than the solar cycle (eleven years period), which is the ultimate cause of that component. A trend and seasonal components have been determined and finally an irregular component has been observed. This is caused by occasional events, such as rain, which washes ^{7}Be out depositing it onto the ground. However, it has been found that the atmospheric concentration of ^{7}Be is negligibly affected by rain.

REG.P19 (Id: 152)
RADIOACTIVITY CONCENTRATION IN SOIL SAMPLES TAKEN FROM REGION OF BANJA LUKA (BOSNIA AND HERZEGOVINA)

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Surface soil samples (0-15 cm) were collected from the region of Banja Luka (Bosnia and Herzegovina), in order to evaluate their radioactivity. Concentrations of radionuclides in samples were determined by γ-ray spectrometer using HPGe detector. The obtained results of uranium and thorium series as well as potassium ^{40}K and ^{137}Cs activity concentrations are discussed. The radioactivity levels are compared with the results form other countries.

REG.P20 (Id: 158)
SPECIATION OF ^{129}I AND ^{127}I IN SOIL AND SEDIMENT SAMPLES

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Iodine is a biophilic tracer element with thirty-four isotopes including one long-lived isotope, ^{129}I (15.7 My), and one stable isotope, ^{127}I. Toxicity, mobility, bioavailability, bioactivity, and the uptake of iodine in environment are governed by its chemical speciation and the soil or sediment conditions. It is therefore important to identify and quantify different physicochemical contaminant species. In this work, a sequential extraction procedure combined with accelerator mass spectrometry (AMS), and inductively coupled plasma mass spectrometry (ICP-MS), respectively, was applied for investigation of ^{129}I and ^{127}I in different physicochemical forms in soil and sediment samples. The fractionation allowed identification of water soluble, exchangeable, carbonate, oxides, iodine bound humic acid, iodine bound fulvic acid and iodine bound humin forms. This is the first study to identify humic acid, fulvic acid and humin bound ^{129}I. A Danish soil sample, a soil reference material (IAEA-375), an anoxic sediment sample collected from Helvik Fjord (South Norway) and an oxic sediment sample collected from the Barents Sea (Russia) were investigated. The results of the work show that in the investigated samples ^{129}I bound to organic matter accounted for more than 50% of the total iodine. Approximately 90% of the ^{129}I is bound to organic matter and oxides, and 10% was found in the readily exchangeable fractions. Speciation analysis of ^{129}I in soil/sediments is indispensable for a better understanding of bioavailability, mobility, bioactivity, uptake and toxicity of iodine in the environment.

REG.P21 (Id: 168)

Cs, Pu AND Am SORPTION TO NATURAL CLAY AND THEIR VARIOUS COMPONENTS: LABORATORY EXPERIMENTS AND MODELING

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Natural clay and iron minerals of complex composition are reactive mineral phases which can affect retardation and transport of radionuclides in the environment. Therefore, it is important to understand and quantify sorption of radionuclides on them. Sorption of Cs, Pu(IV), Pu(V) and Am(III) at trace concentrations on three well characterized samples of Triassic clay as a function of pH and composition of solution and contact time was studied. Sorption experiments were also performed with three iron oxide minerals found in the clay mineral coatings and microorganisms (bacteria and fungi) isolated from the clay and groundwater samples with the aim of better understanding the effects of various natural clay components on the speciation and sorption behavior of radionuclides. The character of bonding of Cs, Pu and Am to the clay samples was analyzed by sequential extraction (SE). Solvent extraction techniques (using TTA, HDEHP, DBM and PMBP), ultrafiltration and α spectrometry as well as ICP-MS were employed to characterize the oxidation states of the plutonium species in liquid and solid phases. The results have indicated that iron oxide coatings play an important role in binding of Pu(IV) on the clay, whereas ion exchange and $CaCO_3$ were found to be responsible for the binding of Am(III). The coatings also affected Cs sorption from solutions of variable ionic strength. Sorption experiments with microorganisms indicated that bacteria and fungi can exert different effect on the sorption behavior of Pu. Kinetic data evaluated by kinetic models derived for six different control processes mostly indicated a sorption mechanism controlled by Pu or Am diffusion in the inert layer on the surface of clay. Corresponding kinetic coefficients were determined. Surface complexation modeling of the sorption dependence on pH using a two-site surface complexation model showed high affinity of all the minerals for $Pu(OH)_3^+$ at pH > 4. Iron oxide minerals also sorbed $Pu(OH)_4^0$: goethite at pH>8, hematite at pH = 7-9 and magnetite at pH > 5.5. Am^{3+} and $Am(NO_3)_2^+$ were found to be the most selectively sorbed species on all the minerals from $NaNO_3$ solutions in the pH range of 3.5-5, whereas $AmOH^{2+}$ and $Am(OH)_2^+$ were sorbed to a different extent on different minerals at pH 5-9. The stability constants obtained by modeling have indicated that the sorption affinity of the clay for Pu is higher than that of the iron oxides studied, while the sorption affinities for Am are mutually comparable. In addition, it has been found that Pu is much more selectively (i.e. strongly) bound than Am on all the minerals studied. This well corresponds with the higher exchangeability of Am found by SE and can explain its higher mobility in the environment.

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REG.P22 (Id: 188)

ANTROPOGENIC RADIOTRACERS ASSESSMENT AS A TOOL TO ENVIRONMENTAL COMPLIANCE RULES AND MANAGEMENT

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A framework, within which models and studies can be applied and results analyzed to develop coherent and logical environmental impact management methodologies for ionizing radiation, is essential. A number of components, which could form the basis for such a system, has to include the dilution factor for radioactive liquid effluent releases. The regulatory mechanism in Brazil established that the operational management concerning the radioactive liquid effluents release must take into consideration the characteristics of the discharged liquid, the knowledge of the dispersion conditions after discharge into the system receptor (sewage and environment), the characteristics description of the effluent recieving system environment and the estimate of the dose. In this study a strategic, fast and cheap methodology was tested to estimate the dilution factor in the release of liquid radioactive effluent at Instituto de Pesquisas Energéticas e Nucleares (IPEN). The radioisotopes ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs, generated in the routine operation of the Research Reactor IEA-R1, were used as radiotracers. In the generated liquid radioactive effluent, stored in a tank of 300 m^3, the radioisotopes initial concentration was determined. The tank flow rate output was estimated to 10.9 ± 0.9 m^3 h^{-1} for liquid controlled dispenser. A planned release of tank effluent stored was carried out. Simultaneously, a sample was taken upstream of the storage tank discharge point for monitoring the concentrations of ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs, respectively, in the mix sewage system discharge point E1 at IPEN. The initial concentration of the ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs was determined as 56881 ± 3255 Bq L^{-1}, 41.5 ± 2.9 Bq $^{-1}$, 1332 ± 11 Bq L^{-1}, 401 ± 8 Bq L^{-1} and 291 ± 7 Bq L^{-1}, respectively. The dilution factors, in the discharge point of the aqueous effluent, were found to be 4.3 and 7.4 for ^{3}H, 12 and 16.1 for ^{54}Mn, 12.6 and 14.2 for ^{60}Co, 12 and 27.9 for ^{65}Zn and 6.2 and 13.9 for ^{137}Cs, respectively. These determined values correspond to two consecutive days of planned release and diluted effluents sampling. The developed methodology was rapid and without additional environmental and financial costs, because the radiotracers ^{3}H, ^{54}Mn, ^{60}Co, ^{65}Zn and ^{137}Cs had already existed routinely in the effluent. The current goal in the world is the development and improvement of cleaner production technologies.

REG.P23 (Id: 202)
PLUTONIUM 241 - IMPORTANCE AND WAYS OF DETERMINATION

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Plutonium 241 is the only β emitting plutonium isotope formed by a nuclear reactor operation. In contrast to the other ones, its half life is relatively short (about 14 years). Emitting β radiation, it decays to another important radionuclide, ^{241}Am. Hence, the activity of americium 241 is still increasing in the area of Chernobyl, Ukraine. Nuclear fission leads to higher-order activities of the ^{241}Pu than of the other plutonium isotopes. On the other hand, the β energy of this radionuclide is only 20 keV, which, in addition with no γ radiation, falsely decreases its importance. For example, determination of isotopes activity ratio between ^{241}Pu and other isotopes can lead to the recognition of the isotope mixture source.

REG.P24 (Id: 226)
URANIUM AND LONG-LIVED DECAY PRODUCTS IN WATER OF THE MULDE RIVER

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The Mulde River, a tributary of the Elbe River in east-central Germany, is formed by the confluence of the Freiberger Mulde and the Zwickauer Mulde, both rising from the Erzgebirge. Running north, the two rivers merge near the small village of Sermuth to form the Vereinigte Mulde, which flows into the Elbe River near Dessau. The Erzgebirge or Ore Mountains are a mountain range, which is located at the border between Saxony / Germany and the Czech Republic. Due to its rich uranium deposits the area of the Zwickauer Mulde River became the world's third largest uranium supplier during the time of the Warsaw Pact. Today, clean-up of the legacy of the former mining and milling industry represents one of the largest remediation projects in progress. Being part of the project "Transport and availability of uranium and decay products in alluvial soils of the Mulde River", this work deals with the investigation of radionuclides in the water of the Mulde River. The radionuclide contents of water and sediment indicate the overall condition of a river, with the water showing a faster response to variations. 20 samples were collected along the course of the rivers Zwickauer Mulde and Vereinigte Mulde between the city of Aue in the Erzgebirge and the Saxon border. For comparison, further samples were taken at the headwaters of the Zwickauer Mulde, along the Freiberger Mulde and the Leine River (Hannover), respectively. A total of 26 samples of water were taken and analysed for uranium, polonium-210, and radium-226 content using α-spectroscopy, liquid scintillation counting, and mass spectroscopy, respectively. The work focuses on present contaminations deriving from tributary streams or river sediments and the detection of changes as a result of remediation effects. Own results are compared with results from earlier investigations, carried out from 1991 to 1993.

REG.P25 (Id: 242)
LEACHABILITY OF NATURAL RADIONUCLIDES (^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th AND ^{228}Ra) IN BRAZILIAN PHOSPHATE FERTILIZERS AND PHOSPHOGYPSUM

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The presence of natural radionuclides of the U and Th decay series in phosphate mineral ores and their redistribution in products, by-products and residue is well known. The most common phosphate fertilizers used in agriculture are single superphosphate, triple superphosphate, monoammonium phosphate and diammonium phosphate, together with phosphogypsum which is used as soil amendment. For a long-term safe application of these fertilizers it is important to study the bioavailability of the radionuclides to the environment. For this purpose, the evaluation of radionuclides concentration in the labile fraction is more suitable than the total concentration since this fraction is more available for the absorption by plants and water contamination. This paper aims to study the leachability of radionuclides (^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Ra) present in phosphate fertilizers and phosphogypsum, using a solution of EDTA as extracting agent. U and Th activity concentrations were measured by instrumental neutron activation analysis. ^{226}Ra, ^{210}Pb and ^{228}Ra were measured by gross α and β counting after radiochemical separation.

REG.P26 (Id: 250)
NATURAL AND ARTIFICIAL RADIOACTIVITY IN SOILS FROM ROMANIAN SITES DETERMINED BY γ-RAY SPECTROMETRY

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This paper presents the radioactivity levels of ^{137}Cs man-made and naturally occurring radionuclides (^{40}K, ^{235}U, ^{238}U-decay series, and ^{232}Th-decay series) in Romanian soils from several industrial sites, determined by low background γ-ray spectrometry. Agricultural soil samples were collected from the vicinity of the phosphate fertilizer plant at Turnu Magurele, as well as from non-ferrous and chemical industry sites at Copsa Mica and Pitesti. Control samples from agriculture and non-specific industry locations were considered (Crevedia and Magurele). Surface soil samples were collected from six locations with different degrees and types of industrial activity (Baia Mare, Copşa Mică, Deva, Galaţi, Oradea, Afumati), and a background site with relatively clean air in the Carpathian Mountains (Fundata). This method makes it possible to assess U, Th, and K concentrations in soil samples by measuring ^{238}U and ^{232}Th (in equilibrium with their radioactive daughters), and ^{40}K specific activities. Their values were found to be in agreement with those determined by Instrumental Neutron Activation Analysis (INAA), through neutron irradiation of soil samples and appropriate standards at TRIGA reactor of the Institute of Nuclear Researches in Pitesti. The radioactivity levels in the investigated soils were compared with Romanian norms for the environmental radioactivity and literature data.

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REG.P27 (Id: 251)
AIRCRAFT-CARRIED SAMPLING SYSTEM FOR AERIAL RADIOACTIVITY MONITORING

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The monitoring airborne radioactive particle became a prominent figure for early warning to form the basis of effective and successful prevention against disasters like the misfortune in Chernobyl. Air sampling and analyzing techniques are essential for early detecting nuclear activities and/or accidents in neighboring countries. Aircraft-borne sampling system is the best way to approach the aerial point of interest. In the earlier study, we designed passive type sampling system using high speed cruise of aircraft. The design concept was focused on maximizing the suction flow necessary for achieving high efficiency of radioactive particles sampling, while minimizing disturbance to the aircraft which carries the system. The optimization was performed with the computational fluid dynamics (CFD) code, FLUENT (FLUENT Inc., 2006). A rocket launcher of a fighter was remodeled into the sampling equipment. The sampler contained a cylindrical filter paper to collect radioactive dust and a charcoal filter to capture radioactive iodine. Replacing the samplers as the launchers at the fighter wings, the aircraft passed in the safety evaluation. Several sampling flights have been performed to provide against emergencies and to assess environmental background. Right after each flight, the filters were radio-analyzed in high purity germanium detectors. The long-half-life radionuclides likely to be made by nuclear experiments or accidents drew main attentions. Fortunately, no nuclide of concern was detected over minimum detectable activity.

REG.P28 (Id: 267)
INVESTIGATIONS ON SEEPAGE WATERS OF THE TEST SITE GESSENWIESE BY TRLFS

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The test field “Gessenwiese” was installed on a leaching heap at the former uranium mining area Ronneburg (western Thuringia) for investigations in acid mining drainage and in heavy metals retention, especially uranium [1]. The uranium speciation in seepage water of the Gessenwiese was determined by TRLFS. Time-resolved laser-induced fluorescence spectroscopy (TRLFS) posses some superior features, above all a very high sensitivity for fluorescent heavy metal ions. The predominance of TRLFS compared to other spectroscopic techniques, e.g. XRD and IR was showed in [2] in analyzing the speciation of U(VI) in a thin layer of an alteration product formed on depleted uranium. TRLFS analyses were carried out to compare it in a later stage with the uranium speciation in plants, which grow on that grassland and may take up uranium contaminated water. The uranium content in the seepage water samples was 75.1 and 291 ppb, respectively, and both samples delivered an evaluable fluorescence signal for TRLFS. The positions of the six peak maxima from these signals are in both water samples in concordance with data for uranium sulfate species published in the literature [3] at 477, 493, 513, 537, 562 and 591 nm. Moreover, the time-resolved fluorescence signals of both water samples possess a mono-exponential decay, indicating the presence of one main species. These two characteristics, i.e. positions of peak maxima and lifetimes revealed without doubt that the uranium speciation in the seepage water is dominated by uranium (VI) sulfate species. TRLFS measurements with plant compartments (e.g. roots, leaves, shoots) which grow in association with the seepage water will be carried out in future investigations. Samples could be obtained from the plant compartments by centrifugation as cell sap, or as solid milled plant compartment sample and subsequently analysed by TRLFS. The reactivity and toxicity of uranium depend on the speciation of heavy metals [4] and accordingly have to be considered as important possible risk factor as uranium may enter economic plants and eventually arrive in the food chain.

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REG.P29 (Id: 277)

RADIOECOLOGICAL CRITERIA AND NORMS DURING REMEDIATION OF SHORE INFRASTRUCTURE OF RUSSIAN NUCLEAR FLEET

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Remediation of sites and facilities belonging to the sites of temporary storage (STS) of spent nuclear fuel (SNF) and radioactive wastes (RW) at Andreeva Bay and Gremikha village on the Kola Peninsula is one of the regulatory functions of radiation protection. After termination of operation of the ex-Navy serviced facilities, their infrastructure degraded resulting in radioactive contamination of some parts of the site. As one end-goal of remediation is putting STS into ecologically safe conditions, elaboration of quantitative radiation-ecology criteria and norms for STS site and facility conditions is urgent. Remediation criteria and norms defining requirements for radiation protection of workers, public and limits of environmental contamination have been developed for three main options: conservation, conversion (partial or overall renovation) and liquidation in the form of:

- dose limits and dose constraints;
- levels of radioactive superficial contamination of workshops and equipment inside them;
- specific activity of radionuclides in marine media, including fish;
- activity concentration of radionuclides in ground waters on-site STS.

The Environmental radiation monitoring findings served as a basis for the associated databank arrangement. The experimental results received have been plotted on the electronic map of the place. Further steps relate to generation of full geographic information system, which permits to carry out the computer calculation of the main indexes for simulation and prognosis of radio-ecological situation with the purpose of the radiation safety regulation.

REG.P30 (Id: 304)

THE ESTIMATION OF RADON ACTIVITY IN MULTISTOREYED BUILDINGS OF INDUSTRIAL IRON-ORE REGION

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Some regions of Ukraine are radon-dangerous. Geologic peculiarities of Krivbass region, available mining tunnels, historical habits in house construction, increased mortality on malignant tumor of population, specify the actuality of “Radon” problem investigations in iron-ore region which should be considered in the estimation of radiation exposure to population. In our own research it has been determined that as a rule radon is accumulated in basement and on the ground floor of buildings. Though significantly higher doses of radon EEVA were registered in the upper stores of five-storied dwelling houses built in the period 1968-1970. Thus the levels of ^{222}Rn EEVA in five-storied dwellings were registered as follows: 33 - 113 Bq m^{-3} on the ground floor, 21-63 Bq m^{-3} on the first floor, 43-69 Bq m^{-3} on the second floor, 22-85 Bq m^{-3} on the third floor, and 67-93 Bq m^{-3} on the fourth floor. During the measurement the daily variation of radon content in air of the dwellings it was determined that the radon volume activity depends directly on ventilation regime in the dwelling. Thus the radon level was measured in two apartments of the fourth floor with two different ventilation regimes - with regular opening of the window and without this procedure. In the first case the family consisted of three employed adults. Radon level was measured in the kiTchen with regular ventilation by window. This level was 67 Bq m^{-3}. Effective dose of radon exposure was 2.9 mSv year^{-1}. Radon level was measured in the room with vacuum ventilation provided by the house design. This level was 93 Bq m^{-3}. In the second case the residents of the apartment were three employed adults. Radon level was measured in the room with vacuum ventilation provided by the house design. This level was 93 Bq m^{-3}. Effective dose of radon exposure was 4.0 mSv year^{-1}. In both cases exposure doses were significant in spite of additional ventilation. Enhanced radon concentration in the upper stores is caused by lack of ventilation. The rate of air near ventilation holes was < 0.1 m s^{-1}. This peculiarity appears during the period of heating the apartments, when windows are closed hermetically and the air inflow is lower. As a result the contingent of population exposured to radon increases.

REG.P31 (Id: 316)
FRACTIONATION OF Sr(II), Th(IV), U(VI) IN THE NATURAL WATER SAMPLES BY NANOCOMPOSITE TRACK MEMBRANES

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Knowledge of the speciation and partitioning of radionuclides and their natural analogs in nature water have great radioecological significance for all environmental systems and Ural region in particular. Distribution of interesting elements in geochemical fractions in water solutions is one of the more important points for understanding and modeling mechanism of radionuclide migration in natural water. In this work the method of ultrafiltration was used for the determination of fractionation composition of Sr(II), Th(IV), U(VI) in natural basin in the vicinity of the Mayak Production Association (South Urals, RF) [1]. Samples of natural water from the Malishevo Lake; Sosnovskoe Lake; Techa River, Shelkun Lake; Karasie Lake, Komarovo Pond, Cherviannoe Lake were investigated. Sampling of the natural water was carried out in spring (May) 2009 to determine radionuclides and investigate their physicochemical state (speciation). Samples selection and preparation techniques met the recommendations of the Public Health Ministry of the USSR. For the separation of each of the fractions we used the synthesized nanocomposite track membranes NCTMs with conducting titanium oxynitride coating to determine fractionation and chemical composition of suspensions and colloids in natural water. This NCTM were obtained from polymer track membrane by modification of the surface and volume of pore space by deposition inorganic high-strength titanium oxynitride layers with different thickness by the method of ion-plasma sputtering [1]. In this work we used membranes produced in Flerov Laboratory of Nuclear Reactions, JINR (Dubna, Russia) with initial pore size 100-3000 nm as a precursor to synthesize membranes with advanced chemical and mechanical properties. These coatings make it possible to simultaneously vary the size of the entrance pores within the necessary limits (from 100 nm to 1-10 nm) and the physicochemical characteristics of the final composite. NCTMs allowed not only to fractionate suspensions, colloids and molecular-ions fractions, but visualized and described natural nanodimensional objects. Using conducting NCTM we have carried out non-invasive chemical identification of biological and geochemical suspensions and colloid materials by means of EM and EDX methods. NCTM has been shown to separate about 10-20 % of Sr(II), Th(IV), U(VI) concentrations from natural water samples with the size >0.1 μm. These fractions consist of Mg-Si-Ca-Fe, $CaSO_4$, biological materials (diatomaceous algae). Our collaborators from IPAE UB RAS have investigated and identified the types of algae. It was shown that molecular-ions fractions of the above elements are in the range 80-90 % of their concentrations.

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REG.P32 (Id: 325)
ENVIRONMENTAL MONITORING OF NUCLEAR MATERIALS FOR SAFEGUARDS PURPOSES BEFORE DECOMMISSIONING OF NUCLEAR FACILITIES

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A comprehensive environmental monitoring and analysis is very important before the decommissioning of a nuclear facility to assure the database for reveal undeclared clandestine nuclear activities. Experiences in the process and sampling concerning decommissioning are not yet available in Hungary because the facilities have not still accomplished the end of their lifecycle. Decommissioning is the final phase in the lifecycle of a nuclear installation and is to be considered as part of a general strategy of environmental restoration after the final suspension of the industrial activities. The first research reactor in Hungary will need to be decommissioned over the next 10 years therefore the environmental monitoring will be necessary before the commencement of the process. The international regulation and the European Union also specify the environmental monitoring and an environmental impact study before the decommissioning procedure. The development of a comprehensive environmental monitoring system including sampling strategy and method development for the analysis of environmental samples, originating from the vicinity of the Hungarian nuclear facilities has been begun at the Institute of Isotopes. The developed method was tested by analysis of real samples. Environmental samples and its nuclear material content were analyzed in the near of a Hungarian nuclear facility.

REG.P33 (Id: 330)
PLUTONIUM SPECIATION IN THE SOUTHERN BALTIC SEA SEDIMENTS

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Wet and dry atmospheric fallout from nuclear weapon tests is one of the most important sources of plutonium in Baltic Sea. The other sources: plutonium releases from spent fuel facilities in Sellafield (UK) and Cap de la Hague (France) are less important. Since April 26, 1986 there is a new source of plutonium - Chernobyl plutonium, which should be taken under note in estimation of its radiological effects on the environment. In marine environment plutonium exists on III, IV, V and VI oxidation state and in the Baltic Sea 99% of total plutonium is deposited in sediments. In this study 6 different chemical forms of plutonium (the fraction dissolved in water, connected to carbonates, connected to oxides, complexed with organic matter, mineral acids soluble and the rest) in sediments from the delta of Vistula River, Gdańsk Bay, Gdańsk Deep and Bornholm Deep were determined. The distribution of $^{239+240}$Pu in analyzed sediments samples was not uniform and depended on its chemical form, depth and geomorphology of the sediments. The highest $^{239+240}$Pu concentrations were found in muddy sediments of Gdańsk Bay at 8-9 cm layer while the lowest, 10 times lower than in Gdańsk Bay, near the delta of Vistula River. The lowest plutonium concentrations were found in the lowest layers (below 22 cm). According to all analyzed fractions the biggest amount of $^{239+240}$Pu in all sediments was found in the fraction connected to carbonates: the delta of Vistula River - 34%, Gdańsk Bay - 42%, Bornholm Deep - 35%. In sediments of Gdańsk Deep plutonium was mostly connected to fraction soluble in mineral acids (40%). Significant $^{239+240}$Pu amount was also connected to organic matter (29%), but the lowest amount of plutonium was in fraction dissolved in water.

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REG.P34 (Id: 332)
NATURALLY EXISTING ^{210}Po IN HUMAN AND DOG HAIR SAMPLES

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More than 100 samples of human hair and 15 samples of dog fur were collected from Pomeranian area for polonium analysis. The ^{210}Po concentration in analyzed hair samples ranged from 0.33±0.02 to 12.84±0.80 mBq.g^{-1} dw (mean value is 3.41 mBq.g^{-1}). The highest ^{210}Po activity values were found in hair sample collected from men had rich fish diet and smoked about 20 cigarettes per day. The lowest ^{210}Po concentration was found in hair sample collected from a lady who did not consume fish and did not smoke cigarettes. The lowest amounts were also found in children's hair (0.43±0.03 and 0.84±0.08 mBq.g^{-1} dw) who did not eat much fish and lived at homes free of cigarettes. In the case of a child living at home where exposed at cigarettes this concentration was much higher - 4.02±0.37 mBq.g^{-1} dw. Among dogs fur the highest ^{210}Po concentration was found in 7 years old dachshund dog - 4.48±0.54 mBq.g^{-1} dw, but the lowest ^{210}Po concentration was found in 3 years old staffordshire bull terrier biTch - 1.96±0.26 mBq.g^{-1} dw.

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REG.P35 (Id: 333)
POLONIUM ^{210}Po AND URANIUM ^{234}U AND ^{238}U IN PHOSPHOGYPSUM AND SURFACE WATER AT WASTE DUMP IN WIŚLINKA FROM PHOSPHORIC FERTILIZERS INDUSTRY OF GDAŃSK AND THEIR ACCUMULATION IN PLANTS

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Phosphogypsum refers to the gypsum formed as a by-product of processing phosphate ore into fertilizer with sulfuric acid. They are radioactive due to the presence of naturally occurring uranium and radium in the phosphate ore and enriched in natural polonium ^{210}Po, while the uranium radionuclides (^{234}U and ^{238}U) tend to be in waters around waste dump. The migration and distribution of uranium and polonium radionuclides in analyzed water samples are not uniform. All analyzed phosphogypsum samples contained higher ^{210}Po activity in comparison with uranium radionuclides. The values of uranium and polonium concentration in water with immediate surroundings of waste heap are considerably higher than in the waters of the Martwa Wisła River. The activity ratio of ^{234}U/^{238}U is approximately about 1 in the phosphogypsum and between 0.92-9.99 in the water of retention reservoir and pumping station, while in the water from the Martwa Wisła River is slightly higher than 1. Polonium and uranium concentrations in water samples of the Martwa Wisła are relatively low in comparison with value in retention reservoir and pumping station near phosphogypsum waste heap. This suggests that radionuclides could be leached from dumping site to the surrounding environment. The major source of polonium and uranium in plants is wet and dry atmospheric fall containing the soil and the air dust from the phosphogypsum waste dump and also accumulation through the root system. The highest uranium and polonium concentrations were found in older part of grasses (yellow oat-grass, meadow foxtail, moneywort), turned for atmospheric fall for a long time.

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REG.P36 (Id: 335)
TEMPERATURE EFFECTS ON EMANATION OF RADON FROM ROCK TO WATER AND ON ITS PARTITION BETWEEN WATER AND AIR

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Emanation of radon (^{222}Rn) from rock and partition of radon between water and air are important parameters, so its evaluation is of significance in the study of radiation protection, environmental geochemistry, climate change and so on. In this study, emanation of radon from rock particles (1-2 mm) to water and partition of radon between water and air by temperature of 0, 10 and 20 °C have been determined using an equilibration partitioning in a closed system (EPICS) method. Specific activity of radon in the rock particles used here were 4.990 ± 20 Bq/kg. The results show that emanation of radon from rock and partition of radon on air increase with increasing the system temperature, however partition of radon on water slightly decreases with increasing temperature. In conclusion, radon emanation from bed rock or soil and partition of radon between water and air is controlled by temperature change, which in turn is also governed by climate change.

REG.P37 (Id: 337)
MICROBIAL ACTIVITY IMPACT ON THE CHANGE OF Pu OXIDATION STATES

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The potential of migration of Pu at contaminated sites and also from the waste repository depends on the Pu environmental behavior determined by its chemical speciations. Depending on the ambient conditions plutonium exists in several oxidation states due to the low differences in the oxidation - reduction potentials between the oxidation states [1]. The geochemical and physical characteristics of the environment (mineralogy, pH, chelating agents, redox conditions, temperature, and humidity) and microbial activity play an important role in plutonium behavior in various systems. Under appropriate conditions the activity of microorganisms could affect the chemical nature of plutonium due to changing redox condition and biotransformation of its oxidation states. The oxidation states control the solubility, complexation and thus the geochemical action of Pu [2]. The direct enzymatic or indirect nonenzymatic interaction of microbes with plutonium could considerably cause its solubility or immobilization because of the formation of stable complexes with metabolites and organic degradation products. This study is aimed at investigations on the change of plutonium oxidation states due to activity of aerobic bacteria and fungi. Both microorganisms and fungi were isolated and identified from samples collected from the low-level radioactive waste repository. Aerobic bacteria Bacillus mycoides, Rhodococcus lutes, unidentified Gram-bacteria, and fungi Paecillomyces lilacinus, Absidia sp., Gliocladium virens have been detected in dust. Oxidized and reduced Pu was tested by two radiochemical procedures [3,4]. One procedure (co-precipitation with NdF_3) only differentiates between reduced Pu [Pu (III) and Pu (IV)] as well as oxidized Pu [Pu (V) and Pu (VI)]. The second (extraction chromatography) procedure was designed to differentiate between Pu (III) and Pu (IV). The amount of reduced and oxidized Pu was determined by measuring α activity after radiochemical separation. The results of the experiment have shown that all bacteria and fungi can very slightly alter oxidation states of plutonium due to their microbial activity. No more than 8-9 % and 15 % of Pu (IV) was reduced to Pu (III) by bacteria during one hour and 24 hours of the interaction respectively. Pu by fungi was reduced no more than 1%.

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REG.P38 (Id: 340)
CURRENT STATUS OF GROSS α/β ACTIVITY ANALYSIS IN WATER SAMPLES: A SHORT OVERVIEW OF METHODS

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In the last few decades, increasing attention is paid to restrict exposure of the public to the naturally occurring radiation. The WHO (WHO, 2004) and the EU Council (EC, 1998; EC, 2010-in preparation) have determined the reference level of the effective dose received from drinking water consumption at 100 μSv y^{-1}. This value excludes the dose received from ^{3}H, ^{40}K, ^{222}Rn, and radon decay products, but it includes the other α- and β-emitting radionuclides. In general, gross α-β analysis is used as the first step of the radiological characterization of drinking waters as a screening method. Its main advantages are the relatively low costs, simplicity, and stability. In the third edition of WHO guidelines for drinking water quality (WHO, 2004) the recommended levels are 0.5 Bq L^{-1} for gross α and 1 Bq L^{-1} for gross β activities, respectively. If the measured values are below the reference levels of gross activity, the drinking water examined is acceptable for human consumption without any further action with respect to its radioactivity. Otherwise, nuclide specific analysis is required to determine the radionuclide content using more sophisticated, more expensive and time-consuming procedures. Due to the uncertainties of gross α and β measurements this method is often the subject of discussions and debates. The aim of this work is to collect information about recently used standard and routine methods concerning gross α and β activity determination in drinking waters in order to evaluate their possibilities. Sample preparation methods - e.g. evaporation, co-precipitation - and detection systems - e.g. gas flow proportional counting, liquid scintillation counting and scintillation counting - are compared on the ground of literature data. In the course of our work, the following parameters were analyzed: background, counting efficiency, interferences, sample capacity, minimal detectable activity, typical counting time, time demand of sample preparation. On one hand, the paper gives an overview of the recently used techniques and, on the other hand, tries to help finding a suitable gross α β radioanalytical tool adapted to the laboratory's demands. This work is part of our effort to develop a reliable method to determine reference values for gross α and β activity in water samples to be used for an interlaboratory comparison between EU monitoring laboratories in the near future.

REG.P39 (Id: 348)
STATE AND MIGRATORY ABILITY OF URANIUM AND RADIUM IN THE SOILS OF BELARUS

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Alpha-emitting uranium and radium are present practically in all ecosystems and together with their decay products play an important role in formation of internal radiation dose of population from natural radionuclides. This dose depends essentially not only on the content of radionuclides in environmental components, but also on the radionuclides' state and migratory ability in a soil medium. The aim of this work is the determination of uranium and radium species in soils and natural soil waters and evaluation of radionuclide migratory ability in different soils. The objects of investigation were samples of 0-10 cm layers of soils from different regions of Belarus. The natural system "solid phase - pore water of soil" have been used for determination of uranium and radium migratory active forms in the soil samples. The potentially mobile forms and potentially biologically available forms were determined using accordingly the model systems "soil - 1 mol/L solution of ammonium acetate" and "soil - 1 mol/L solution of HCl". Radionuclide species in soil waters were investigated by passing of the water samples through the filters of different pore size with subsequent separation of complex radionuclide forms of different electrostatic charge using the method of ion exchange chromatography. Radionuclide activity in the samples was determined by radiochemical analysis with radionuclide identification by α spectrometer SOLOIST U0450 with low-background detectors 576 A 600 RV. The specific activities of the soil samples are 4.8-22 Bq/kg for ^{238}U and 4.5-29 Bq/kg for ^{226}Ra. It was found that both ^{228}U (84-94%) and ^{226}Ra (77-91%) are mostly in the fixed forms (insoluble in 1 mol/L HCl). The radionuclide rate in migratory active forms for ^{238}U was 0.06-0.7 % and for ^{226}Ra 0.01-1.0 % of the total radionuclide content in soil samples. The rates of potentially mobile radionuclide forms are 3.1-9.3 % for ^{228}U and 1.8-4.5 % for ^{226}Ra but the radionuclide rates of potentially biologically available forms are respectively 6-16 % and 9-23 % of the corresponding radionuclide content in the soil samples. The main part of ^{228}U and ^{226}Ra of natural soil water samples (65-91 %) is in dissolved state or in composition of fine colloidal (less than 0.45 μm) particles. The radionuclides in this state are the most mobile in environment. In composition of this portion of natural water samples ^{226}Ra is mainly in the cationic form, but ^{228}U coexist in anionic, cationic and neutral complexes. In organic soil waters, the anionic uranium complexes prevail, but in mineral soil waters, the cationic and neutral complexes are the main portion of radionuclide. The received information allows estimating the uranium and radium rates in forms participating actively in radionuclide distribution in the soil medium as well as evaluating the radionuclide reserves in forms that can enter the soil solutions and participate in processes of biogeochemical radionuclide migration in ecosystems. The results of investigation are the basis for differentiation of soils according to migration ability of uranium and radium and for modeling of radionuclide transferring in the environment.

REG.P40 (Id: 349)
RADIOANALYTICAL INVESTIGATIONS OF URANIUM CONCENTRATIONS IN NATURAL SPRING, MINERAL, SPA AND DRINKING WATERS IN HUNGARY

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As many other countries in the Central European region, (e.g. Czech Republic, Slovakia, Romania) Hungary is rich in spring-, thermal-, and mineral waters as well. Recently the consumption of natural mineral- and spring waters comes to be more and more popular in a certain populations. It is well known some of these waters have elevated level of dissolved naturally origin radionuclides, but their concentration may vary in a wide range. In some cases elevated level of dose exposure can be expected. In spite of this fact most of the recommendations do not apply to natural mineral waters and to waters that are classified as of medicinal benefit. From dosimetric point of view, it has a great importance to screen the radioactivity of these waters as well. Within this work, the activity concentrations of uranium isotopes (^{234}U, $^{23\%}$U, and ^{238}U) were analyzed in some of the popular and regularly consumed Hungarian mineral-, spring-, therapeutically waters and tap waters. Samples were selected randomly and were taken from different regions of Hungary (Balaton Upland, Bükk Mountain, Somogy Hills, Mezőföld, Lake Hévíz). Uranium isotopes were separated from impurities by extraction chromatographic resin and the activity concentrations were determined by using α spectrometry. Concentration (mBq L^{-1}) of ^{234}U, ^{235}U, and ^{238}U in the waters is varied from 0.71 to 741.95, from <0.3 to 9.43, from 0.5 to 274.3 respectively. In general, the highest uranium concentrations were measured in spring waters, while the lowest were found in tap waters. It can be established, in most cases radioactive disequilibrium was observed between uranium isotopes. The activity ratio between ^{234}U and ^{238}U varies from 0.630 to 4.717. The doses for the analyzed samples of spring water are in the range 0.04 - 35.87 μSv y^{-1} with an average 4.86 μSv y^{-1}.This is well below the 0.1 mSv y^{-1} reference level of the committed effective dose recommended by WHO. The other naturally occurring α emitting radionuclides (^{226}Ra, ^{210}Po) will be analyzed later to complete the dose assessment. This study provides preliminary information for consumers and authorities about their internal radiological exposure risk due to annual intake of uranium isotopes via water consumption.

REG.P41 (Id: 248)

CHARACTERISTICS OF CONCENTRATION CORRELATIONS FOR THE PAIRS OF $^{238}U/HCO^{3-}$, $^{234}U/HCO_3$-, $^{226}Ra/^{238}U$, $^{228}Ra/^{226}Ra$, $^{226}Ra/HCO_3$-, $^{228}Ra/HCO_3$-, $^{210}Po/^{238}U$, $^{210}Po/^{226}Ra$' $^{228}Th/^{232}Th$, $^{228}Th/^{228}Ra$, $^{228}Th/HCO_3$- AND $^{210}Po/^{210}Pb$ IN DRINKING WATER

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Some important naturally occuring α and β radionuclides in seventeen brands of drinking water samples collected in Italy were determined. The mean activity concentrations (mBq L^{-1}) of the radionuclides in the water samples were in the order: 26±36 (^{234}U) > 21±30 (^{238}U) > 8.9±15 (^{226}Ra) > 4.8±6.3 (^{228}Ra) > 4.0±4.1 (^{210}Pb) > 3.2±3.7 (^{210}Po) > 2.7±1.2 (^{212}Pb) > 1.4±1.8 (^{224}Ra) > 1.1±1.3 (^{235}U) > 0.26±0.39 (^{228}Th) > 0.0023±0.0009 (^{230}Th) > 0.0013±0.0006 (^{232}Th). Based on the HCO_3^- concentrations and the radionuclide concentrations in the analysed waters, correlation analyses were made, and statistical positive correlations were found among the pairs of $^{228}Ra/^{226}Ra$, $^{226}Ra/HCO_3^-$, $^{228}Ra/HCO_3^-$, $^{210}Po/^{238}U$, $^{210}Po/^{226}Ra$, $^{228}Th/^{232}Th$, $^{228}Th/^{228}Ra$, $^{228}Th/HCO_3^-$ and $^{210}Po/^{210}Pb$ in drinking water.

REG.P42 (Id: 67)

LONG-TERM MONITORING OF RADIOACTIVITY IN ENVIRONMENTAL OF THE CITY OF MOSCOW

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Moscow is the historical center of the atom project of the former USSR. Radioecological assesment of the territory of Moscow has been provided in compliance with the conception and program of comprehensive radioecological monitoring of the City of Moscow approved by the Moscow Administration. The methodology of the envitonmental monitoring is based on long-term determination of concentration of the major dose-forming radionuclides in natural environment and some components of metropolitan megapolis accumulated by mobile and stationary systems. Such an approach ensures to monitor variations in temporary-and-spatial distribution of radiation parameters of the environmental objects as well as to identify the areas where radiation values are abnormal ones and to assess the radiation situation both at separate sites and in the city in general. Monitoring data are used in Radiation-Hygiene Passport of the City of Moscow.

REG.P43 (Id: 136)

THE ACCUMULATION OF TRANSURANIC ELEMENTS OF THE CHERNOBYL EMISSION BY MEADOW VEGETATION OF THE ZONE CHERNOBYL NPP

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The Chernobyl accident was accompanied by release into the atmosphere of a large number of radionuclides with long half-lives. With respect to the exposure of biological objects the most dangerous nuclides in the long term are α-emitters, particularly transuranic elements (TUE-239, ^{240}Pu, ^{241}Am) from the fuel. Radionuclides with different half-lives and environmental features found in the terrestrial ecosystems in various ways become widespread in the ecosystem. To build realistic models of behavior of transuranic elements it is necessary to carry out a focused analysis of the features of the long-lived radionuclides distribution in various types of ecosystems. The aim of this work was to study the behavior of long-lived radionuclides of the Chernobyl genesis in meadow phytocenosis. The evaluated accumulation rates, numerically equal to the ratio of specific activity of TUE in overground phytomass of plants and their concentration in soil (Bq/kg), have been used for characterizing the level and species specificity of the $^{239,240}Pu$ and ^{241}Am accumulation in plants. Fluctuation of values of specific activity phytomass of $^{239,240}Pu$ was observed in range from 0.11 to 0.77 Bq/kg and ^{241}Am - from 1.20 to 21.0 Bq/kg. TUE uptake in plants depends on soil conditions, time of selection and climatic conditions. The experiments were conducted for several years. The results exhibit very large scatter, but a number of peculiarities can be observed. Studies identified vegetation types that are most active in the TUE uptake. For ^{239,240}Pu the following plants were found to be the most active ones: Oenothera biennis L.> Helichrysum arenarium (L.) Moench> Vicia cracca L.; for ^{241}Am: Koeleria gracilis Pers.> Oenothera biennis L.> Elytrigia repens L. > Lysimachia vulgaris L. > Cytisus ruthenicus Fisch. Biological characteristics and life cycle of plants will undoubtedly affect the value of radionuclide accumulation, but the spread of values of accumulation factor on one site within the same family has definite boundaries, characteristic for the family. We have found that the contribution of ^{241}Am contamination of vegetation is predominant among TUE, which indicates its greater biological mobility in comparison with Pu isotopes. This should be taken into account in decisions concerning the use of forage resources in regions polluted in different degrees. The significant uptake of ^{241}Am activity should be taken into consideration when calculating the dose for the common public.

REG.P44 (Id: 339)
DISTRIBUTION PATTERN OF NORM IN RED SEA SHORE SEDIMENTS AND THEIR RELATION TO NON-NUCLEAR INDUSTRIES

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The Red Sea is a deep semi-enclosed and narrow basin that has an intensive non-industrial activities on and near its shore. Oil exploration, phosphate mining and trading, navigation activities and intensive touristic activities are considered as non-nuclear pollution sources. They could impose a serious radiological and ecological impacts on the Red Sea marine environment. Both oil and phosphate related activities could increase the concentration of Naturally Occurring Radioactive Materials - NORM such as ^{238}U series, ^{232}Th series and ^{40}K. Forty representative shore sediment samples were collected from the Egyptian Red Sea shore, from Shuqeir to Marsa Alam City region. Activity concentration of ^{238}U, ^{232}Th, ^{40}K were measured using ICP-MS analytical techniques. Previous study showed the possible impact of industrial activities on the activity concentration of NORM in shore sediment. This study will investigate such relationship and the distribution pattern of NORM in relation to the elemental composition of the shore sediment.

REG.P45 (Id: 367)
SPECIFIC ACTIVITY OF ^{40}K IN DRINKING WATER IN SLOVENIA

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The European legislation (Council Directive 98/83/ec) requests the member sates to control the radioactive contamination of drinking water. As a part of these measurements γ-ray spectrometric measurements are performed. Samples of 50 L of water are evaporated and the residue after evaporation is measured. Typically, 20 g - 40 g of residue is obtained. Since by γ-ray spectrometry activity concentrations of radioactive isotopes emitting γ-rays can be determined also the concentration of natural ^{40}K in the samples is measured. The measurements of activity concentration of ^{40}K are relatively easy since it radiates at an energy, where no interference with other γ-ray emitters occurs. Drinking water in Slovenia is gained mainly from the ground water. Therefore the concentration of potassium in drinking water reflects the properties of the rock of the aquifer as well as the geochemical conditions there. The activity concentration of ^{40}K in drinking water may bear information on the circumstances in the aquifer from where drinking water is extracted. In the contribution the correlation between the activity concentration of ^{40}K in drinking water and the potassium concentration in the soil will be presented. The correlation will be discussed in terms of the geological properties of the aquifers and the level of the water table.

REG.P46 (Id: 338)
RADIOECOLOGICAL IMPACTS OF NORM FRACTIONATION IN PHOSPHATE ROCK BENEFICIATION PROCESSES

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Phosphate rock beneficiation processes (crushing, screening, washing, magnetic separation and wet screening) aims to increase the phosphorus content of the run-of-mine (ROM) ore. It starts with the ROM ore and ends with the wet rock, and different rejects (by-products). These by-products have potential environmental hazards due to their content of Naturally Occurring Radioactive Materials – NORM especially uranium-238 series. They have also potential industrial and agricultural useful applications due to their physicochemical properties such as their relative high content of phosphorus, clay and iron. Representative samples of ROM ore, wet rock, and beneficiation processes by-products (wet screening, magnetic separation, slim, and clay and dolomite rocks) were collected. Natural radionuclides (^{238}U, ^{235}U, ^{232}Th, ^{40}K, ^{210}Pb, ^{210}Po) were measured using γ-ray spectrometer, α particle spectrometer and ICP-MS analytical techniques. Radiologically, internal hazards index, external hazard index, representative level index, γ absorbed dose rate and occupational dose equivalent due to inhalation were calculated. Potential hazards due to beneficiation processes and their by-products were discussed. Some of the by-products could have potential useful applications such as clay rock and slim for agricultural soil reclamation. The aspects of these useful applications were discussed.

6. PRODUCTION AND APPLICATION OF RADIONUCLIDES (PAR)

Lectures

PAR.L01 (Id: 263)
HIGH SPECIFIC ACTIVITY ^{177g}Lu FOR METABOLIC RADIOTHERAPY: DEUTERON CYCLOTRON VS. NUCLEAR REACTOR

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This work is focused on production of ^{177g}Lu for uses in metabolic radiotherapy of tumors, due to the appropriate average β-energy and γ rays suitable for detection by γ-camera and SPET: $t_{1/2} = 6.734$ d, β 100 %, Eβ,max = 489.3 keV, <E> = 163 keV, main $E_{\gamma} = 113$ and 208 keV. It is one of the most promising β-emitters for small cancers. The production methods are either direct neutron capture ^{176}Lu(n,γ)177(m+g)Lu on (60-70 %) enriched ^{176}Lu target (σ-th = 2 + 2100x10^{-28} m^{2}, plus a contribution of epithermal neutrons from resonance peaks), with a lower specific activity AS than the theoretical carrier-free one: AS(CF) = 4.05 GBq.μg^{-1}, or neutron capture on highly enriched ^{176}Yb, followed by β-decay ^{176}Yb(n,γ)^{177}Yb -> ^{177g}Lu (σ-th = 3.1x10^{-28} m^{2}). Due to the long half-life ^{177m}Lu is relevant for rad-waste and dose to the patient and medical personnel. The second method produces a high AS NCA ^{177g}Lu, whilst the first one a lower AS CA mixture of both, diluted in stable Lu isotopic carrier. Thus, in the latter case the ^{177g}Lu is contaminated by the long-lived radionuclidic impurity ^{177m}Lu. Several commercial samples of reactor produced ^{177}Lu were submitted to accurate measurement of both decay patterns and radionuclidic purity vs. time using HPGe and LSCS. The measurement of the isomeric ratio of Lu (direct neutron activation) and the absence of other Lu RNs (indirect neutron or deuteron activation) allows identifying the production method adopted. In case of direct ^{176}Lu(n,γ) route, it was evidenced that - at administration time - the typical amount of the long-lived ^{177m}Lu was of the order of 0.01 %. The experimental $t_{1/2}$ for ^{177g}Lu (6.724 ± 0.006 d) is in very good agreement with the reference value taken from the literature of 6.734 d. An alternative method is based on deuteron activation of ^{176}Yb, by (d,p) reactions followed by decay of the short-lived ^{177}Yb and direct (d,n) reactions as well. Deuteron activations have been carried out at JRC-Ispra Cyclotron (K = 38) of EC, with deuteron beams up to 19 MeV. To conclude ^{177g}Lu can be produced in no-carrier-added form, by either neutron activation on enriched ^{176}Yb or by deuteron irradiation on very highly enriched ^{176}Lu, both followed by decay of ^{177}Yb. In both cases a AS value could be very close to the CF one, after selective radiochemical separation of Lu from Yb target. It is remarkable that ^{177}Yb decays solely to the ground level of Lu, leading to a very high radionuclidic purity ^{177g}Lu, not contaminated by the long-lived metastable radioisotopic impurity ^{177m}Lu. A much higher value of radionuclidic purity is achievable (theoretically 100 %) in case of direct ^{176}Yb(d,n)^{177}Lu route, whose effective threshold is at 13 MeV. Several activation of thin Yb targets by deuterons led to the conclusion that a maximum thin-target yield for ^{177g}Lu of 246 MBq.C^{-1}.MeV^{-1} is achievable (σ-max = 250x10^{-25} m^{2}) at 12.5 MeV, on 100 % ^{176}Yb target. Our data will be compared with the data taken from the literature for the two neutron routes. Finally ET-AAS and NAA were used to determine chemical purity and AS of the products.

PAR.L02 (Id: 9)
OPTIMISATION OF THE LONG LIVED ^{121}Te CONTAMINANT IN PRODUCTION OF ^{123}I TROUGH THE ^{124}Xe(p,x) ROUTE

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At present bombardment of highly enriched ^{124}Xe with 35 MeV protons and taking advantage of the cascade decay ^{123}Cs-^{123}Xe-^{123}I is the preferred route for ^{123}I ($T_{1/2} = 13.2$ h) production. After irradiation the gas targets are allowed to cool for 7 h ensuring optimal in-growth of ^{123}I from its precursors. A separation of I from all other elements is performed resulting in a nca, pure solution. An unavoidable contaminant is ^{121}I ($T_{1/2} = 2.12$ h) produced by ^{124}Xe(p,α) reaction with a cross section maximum around 20 MeV. This rather short lived radioiodine will disappear quickly from the ^{123}I solution, but its long lived decay product ^{121g}Te (16.8 d) accumulates and impairs the late use (several half lives of ^{123}I after calibration date) of the solution. This situation could be improved by lowering the ^{121}I production through limitation of the target thickness and imposing a higher exit energy. As the only values for the ^{124}Xe(p,α) reaction were reported by Tarkanyi et al. [1] at higher energy, reliable data on the excitation function had to be measured for the first time. Highly enriched ^{124}Xe was bombarded with protons between 13 and 37 MeV with the double aim of determining cross sections for ^{121}I production and resolving discrepancies existing in the previously published values for production of ^{123}Cs and ^{123}Xe [2]. Here only results for ^{121}I are presented and compared with results from different theoretical codes. Under the experimental (and production) conditions formation of ^{121}Cs and ^{121}Xe is impossible or extremely low (practical threshold of 1 mb at 38 MeV [1]). Direct formation of ^{121g}Te (16.8d) or ^{121m}Te (154d) through the (p,3pn) reaction are not of importance because of the needed chemical separation at EOB+7 h . The cross sections for ^{121}I production show a practical threshold at 9.5 MeV rising to a maximum of 13 mb at 21 MeV, not in agreement with theoretical results. From a fit to our excitation curve, production yields for short irradiations of thick targets are calculated. The ^{121}I activity present at the optimal cooling time is then calculated for different energy degradation in the target and for different irradiation times (up to 13 h, 1 half life of ^{123}I, saturation of ^{121}I). By comparing to the cumulative produced ^{123}I under the same irradiation conditions, the evolution of the relative activity of both ^{121}I (decreasing in time) and ^{121}Te (increasing in time) is calculated. By defining maximal admissible contaminations levels for late use of the ^{123}I solution, limits on target thickness are defined.

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Tarkanyi F. et al.: Appl. Radiat. Isotopes *42*, 221 (1991).
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PAR.L03 (Id: 16)
DETERMINATION OF THE VALENCE STATE OF ^{237}U, OBTAINED IN THE PHOTONUCLEAR REACTION ^{238}U (γ,n) ^{237}U USING NANOSTRUCTURED MATERIAL.

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^{237}U was obtained in the ^{238}U (γ, n) ^{237}U reaction in the electron accelerator - microtron MT-25 of the Flerov Laboratory of Nuclear Reactions (FLNR JINR, Dubna). The method of recoil atoms capture with application to the nanostructure material - hydrous manganese dioxide (cryptomelane-type) in a solid-solid system was used for the separation of ^{237}U and ^{238}U. The change of the distribution coefficient of the isotope ^{237}U between solution and sorbent in dependence on the acidity of nitrate solutions was studied. These experiments show that most of the ^{237}U is in the +4 valence state.

PAR.L04 (Id: 17)
RADIOCHEMICAL MEASUREMENT OF CROSS SECTIONS FOR THE PRODUCTION OF THE THERAPEUTIC RADIONUCLIDE ^{193m}Pt

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The radionuclide ^{193m}Pt ($T_{1/2} = 4.33$ d), a high spin isomer, decaying by highly converted isomeric transition, emits about 33 Auger electrons per decay. It is of great potential interest in internal radiotherapy because of its suitable decay properties and platinum-complexes (like cis-platin and others) used in chemotherapy, i.e., potent antitumor agents. In order to avoid side effects of chemotherapy, it is advisable to use this radionuclide in the same chemical complex with high specific activity. So far the specific activity of ^{193m}Pt produced has been rather low due to the use of the ^{192}Pt(n,γ) process at a reactor. In a recent study, the ^{192}Os(α,3n) reaction was measured up to 28 MeV [1]. The aim of the present work was to establish reliable and reproducible chemical preparation of thin targets from highly enriched osmium and to extend the measurement of excitation function of the ^{192}Os(α,3n) reaction up to 35 MeV, where the optimum energy range was expected, and to increase the yield and the specific activity of the ^{193m}Pt at a cyclotron. We optimized the dissolution and conversion of osmium to osmium tetroxide vapor and trapping in KOH solution, the electrolytic preparation of thin target samples of highly enriched ^{192}Os on Ni, and the complete radiochemical separation of radioplatinum. The excitation function of the ^{192}Os(α,3n)^{193m}Pt reaction was measured from 26 MeV to 39 MeV using the conventional stacked-foil activation technique. Several stacks containing 99.65 % enriched ^{192}Os targets and thin Ti and Cu foils were irradiated by 40 MeV α-particle beam of about 100 nA for 2 h at the Brussels Cyclotron. The Ti-nat(α,X)^{51}Cr reaction induced in the Ti foil was used to monitor the incident beam intensity. After complete separation of radioplatinum, the radioactivity of ^{193m}Pt was measured by X-ray spectrometry using the Pt X-ray line of 66.8 keV. The measured count rates were converted to decay rates by correcting for the X-ray intensities, self absorption in the source, and the efficiency of the detector using the well known formula; thereafter the nuclear reaction cross section was calculated. The cross section of the ^{192}Os(α,3n)^{193m}Pt reaction reaches a value of about 1.5 b at the maximum at about 35 MeV. It was also calculated theoretically using the compound-precompound nuclear model codes TALYS and STAPRE. The calculated values are consistent with the measured data. The optimum production of the ^{193m}Pt radionuclide by this method was found in the energy range 30-38 MeV; the integral yield of ^{193m}Pt being about 40x higher than that in the energy region below 28 MeV [1]. The results provide basic information on the production of high-quality ^{193m}Pt for therapeutic application. Tests on impurity, the yield and specific activity are underway.

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PAR.L05 (Id: 44)

PC-CONTROLLED RADIOCHEMISTRY SYSTEM FOR PREPARATION OF NCA ^{64}Cu

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Due to the rapid increase of the use of nuclear medicine techniques in modern clinical diagnosis and in a selected series of therapies, researchers efforts are focusing for the standardization and optimization of different production routes for a series of emerging radioisotopes like ^{64}Cu, ^{67}Cu, ^{114m}In, ^{211}At. In particular the EC/β^+/β- decay of ^{64}Cu makes it a promising candidate for both PET imaging and internal targeted radio therapy. In the last decades several groups studied different production routes to obtain this radionuclide, i.e. ^{64}Ni(p,n), ^{64}Ni(d,2n),^{64}Zn(d,2p). Taking into account the wider availability of the medium energy proton beam machines, the (p,n) reaction on ^{64}Ni seems to be the most attractive one, although ^{64}Zn(d,2p) may be considered as an alternative where lower activity is necessary, as it may require less investment in enriched material. The production of large activities of ^{64}Cu on regular basis requires a fast and reliable chemistry system. Based on the experience gathered in the last decades in our laboratory we present here an efficient, remote controlled chemistry system for production of the non-carrier added ^{64}Cu via ^{64}Ni(p,n) reaction. To avoid excessive investment in a gold target carrier, a good practice is to coat the copper target carrier with a thin inert material, i.e. 5-6 μm of gold, followed by electrodeposition of the ^{64}Ni target layer. In that way, the cross contamination of the non-carrier added ^{64}Cu with the copper present in the target carrier is excluded. In general the irradiations are performed with protons having incident energy of about 15 MeV, and, depending on irradiation condition, may lead to curie amount of induced activity of ^{64}Cu. To reduce the thickness of the ^{64}Ni target layer, and, as consequence, to minimize the problems related with the plating and dissolution of the target layer, a low beam/target angle geometry (6 degrees) is desired. Nevertheless, the separation of target/activation product is required. Upon irradiation, our chemistry system proposes the dissolution of the ^{64}Ni layer in a heated flow trough stripper by means of diluted nitric acid. Next, the non-carrier added ^{64}Cu is selectively extracted into benzene (containing 0.1 M benzoylacetone) at pH 4.5, leaving the enriched ^{64}Ni and possible Co induced isotopes in the inorganic phase. The back extraction of ^{64}Cu is done in a small volume of diluted hydrochloric acid (6 N). The final purification step is achieved using an anion exchange column Dowex 1X8. Finally, the NCA ^{64}Cu is eluted with a small volume (10 mL) of diluted hydrochloric acid (1 N). The overall yield of the chemistry is estimated as being higher than 95% with a short total chemistry time, less than 3 hours, while the gold plated target carriers can be reused as long as the thin gold layer remains intact, meaning that scratches and cracking by careless handling are avoided.

PAR.L06 (Id: 30)

PRODUCTION OF ^{26}Al, ^{59}Ni, ^{44}Ti, ^{53}Mn AND ^{60}Fe FROM PROTON IRRADIATED COPPER BEAM DUMP

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The station for pions cancer therapy was operated at PSI from 1980 to 1992. For this period the copper beam dump of the facility received a total dose of approx. 0.16 Ah of 590 MeV protons. The sampling collected about 500 g of high active copper chips that can be used for separation of exotic radionuclides. The γ analyses showed main nuclides present to be ^{60}Co, ^{54}Mn, ^{22}Na, ^{65}Zn and long lived ^{44}Ti with a daughter nuclide ^{44}Sc. Further analyses by LSC and AMS demonstrated that significant amounts of ^{26}Al, ^{53}Mn, ^{59}Ni, ^{63}Ni, ^{55}Fe and ^{60}Fe are present in the copper beam dump. The analytical results estimate that about 100 MBq ^{44}Ti, 500 MBq ^{53}Mn, 7 kBq ^{26}Al, 8 MBq ^{59}Ni and 5 kBq ^{60}Fe are available in the collected copper chips [1]. Due to the high activity of ^{60}Co, approx. 5 GBq in total, the separation should be implemented using a hot cell. The purpose of this work is to develop a simple, selective, efficient and easy method for the separation of ^{26}Al, ^{59}Ni, ^{53}Mn, ^{44}Ti and ^{60}Fe from gram amounts of the copper beam dump. For the separation of the exotic radionuclides from the copper beam dump, a procedure combining selective precipitation, ion exchange and solvent extraction was developed. Copper as the main matrix element interferes with the separation of all elements of interest. For this reason, after the copper dissolution in 7 M HNO_3 the solution is conditioned to 1 M HNO_3 and Cu (II) is precipitated selectively by saturation with H_2S, while the exotic radionuclides remain in the solution. Further the solution is transferred in 12 M HCl and passed on Dowex 1x8 anion exchange column. Al and Ni are not retained and are washed out of the column with 12 M HCl. The remaining ions are eluted consequently in the following way, Mn-10 M HCl, Ti-8 M HCl, Co-5 M HCl, Cu-2.5 M HCl and finally Fe with 0.5 M HCl. Finally Al is separated from Ni on Dowex 50x4 cation exchange column and Ni is purified with Eichrom Ni resin based on the traditional dimethylglyoxime precipitation chemistry. The proposed separation procedure is easy for remote controlled implementation in a hot cell. The ion exchange separation of Ni, Al, Mg, Ti and Fe is complete and high decontamination factors for copper and cobalt were achieved. The full scale system is installed in a hot cell where high activity levels could be handled. During the test period 13.86 g in total of the proton irradiated copper beam dump were processed for separation of ^{26}Al, ^{59}Ni, ^{44}Ti, ^{53}Mn and ^{60}Fe. The results showed the system is operational and the radionuclides separation is selective with high chemical yield. The procedure manages as well the generated liquid wastes containing high level of ^{60}Co activity, reducing drastically their volume.

Reference:

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PAR.L07 (Id: 99)

FOSSIL FUEL CO_2 DETECTION BY ATMOSPHERIC ^{14}C AND CO_2 MIXING RATIO MEASUREMENTS IN THE CITY OF DEBRECEN, HUNGARY

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Fossil fuel CO_2 content in the air of a major Hungarian city (Debrecen) was determined using simultaneous measurements of CO_2 mixing ratio and radiocarbon (^{14}C) content in air. In this project we developed a high precision atmospheric CO_2 monitoring station in Debrecen. An integrating sampling system (developed by ATOMKI) was applied for radiocarbon measurements. One sampler was installed in Debrecen station and two independent $^{14}CO_2$ sampling lines were installed ~ 300 km far from Debrecen at Hegyhátsál station as independent background references, where high precision atmospheric CO_2 mixing ratio measurement is also running since 1997. During the winter of 2008/09 we measured the mixing ratio and radiocarbon content of atmospheric CO_2 at Debrecen and the reference station simultaneously. It was concluded that trends in CO_2 mixing ratio variations in time are very similar at the three different sampling points (2 m above ground in Debrecen, 10 m and 115 m above ground in Hegyhátsál). Air quality in Debrecen during September of 2008 seemed to be relatively clear from the point of view of its CO_2 content at least. When winter came closer in October, with lower outside temperature and less sunshine hours the CO_2 content of air increased in all the three sampling points, but this effect was more intensive closer to the ground level. According our radiocarbon observations it was clearly indicated that there was not significant amount of fossil fuel CO_2 in the air of Debrecen during September in 2008. But during the winter of 2008/09 the average ^{14}C value of atmospheric CO_2 of Debrecen decreased by more than 40 ‰ relative to September's results, and according our calculations it was caused by about 20 ppm fossil fuel CO_2 which appeared as a surplus amount in the air above the September level.

PAR.L08 (Id: 101)

INVESTIGATION OF THE DYNAMICS OF EROSION-ACCUMULATING PROCESSES USING ^{137}Cs AND EXCESS ^{210}Pb AS A MARKER OF SOIL MATERIAL DISPLACEMENT

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Global ploughing up of large areas, increased particularly in the last two centuries, lead to fundamental changes of the sediment load balance of flat territories, namely by the repeatedly increasing erosion-accumulating processes. Use of ^{137}Cs as a marker of erosion-accumulating processes allows to estimate the intensity of these processes during the period from the mid-fifties, and in a number of cases (especially, in zones of Chernobyl contamination) during much shorter periods. Atmospheric part of ^{210}Pb (excess ^{210}Pb) is recently used as a marker for estimating the rate of erosion-accumulating processes in 100-120-year intervals. At the same time certain methodical problems of the radiolead method exist, in particular the insufficient understanding of the excess ^{210}Pb fallout spatial variability. There are also other factors, bearing on the precision and reliability of the erosion-accumulating processes rates evaluation. Application of these methods to the central Russian plane has showed that the arrangement of erosion and accumulation zones naturally changed along cultivated slopes in dependence on their morphology; dynamics of the sediment load accumulation rate in dry valley bottoms determines the intensity of the repeated sedimentation process at the expense of secondary erosion development.

PAR.L09 (Id: 127)

SORPTION OF TUNGSTEN AND RHENIUM ON ALUMINA IN $^{188}W/^{188}Re$ GENERATORS

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Rhenium-188 is a recommended radioisotope for diagnostic and therapeutic techniques because of its attractive physical and chemical properties: half-life 16.98 hours, a strong particulate emission (β energy of 2.12 MeV) and an imageable γ emission (15 %, 155 keV) ideal for current γ camera imaging of tumors. Radiopharmaceuticals labeled with ^{188}Re are currently used in clinical trials, such as: ^{188}Re-MAG3 as renal excreting agents, ^{188}Re-HEDP for palliation of metastatic bone pain or ^{188}Re-HDD for treatment of liver cancer. ^{188}Re is the daughter product of $^{188}W/^{188}Re$ generator with a useful shelf-life of several months and can continually provide a supply of ^{188}Re to remote sites at a reasonable cost. In these generators ^{188}W is adsorbed on alumina, charged in a chromatography column, and ^{188}Re is eluted with saline solution. A systematic study of W and Re sorption on alumina as a function of the medium concentration, medium pH, alumina particle size and alumina weight (capacity) and an evaluation of the performances of the $^{188}W/^{188}Re$ generator systems constructed from the W and Re adsorption data are reported. ^{188}Re is not adsorbed in HCl and NaCl media whereas ^{188}W is strongly adsorbed in both media at pH < 5.5. In accordance with the W and Re adsorption results, alumina $^{188}W/^{188}Re$ generators can be prepared and eluted with HCl (< 0.1 mol/L) and 0.9 % NaCl solutions (pH <5.5), using alumina with a particle size of 0.075-0.048 mm and a ratio of 10.5 mg W/g alumina, at this ratio the W is sorbed about 99.9% on alumina and the Re around 3 %. Alumina based $^{188}W/^{188}Re$ generators eluted with HCl or 0.9 % NaCl solutions and with ratios of 10 or 20 mg W/ g alumina were prepared and evaluated for six months. Optimum performances were obtained in the generator eluted with 0.9 % NaCl solutions, a ration of 10 mg W/ g alumina and ^{188}W fixed with 0.01 mol/L HCl: ^{188}Re elution yields of 83 %, a ^{188}W breakthrough of 4.5x10^{-4} %, a radiochemical purity of 92 %, an eluate volume and pH of 8.3 mL and 5.5, respectively, and an aluminum content of less than 10 μg/cm^3.

PAR.L10 (Id: 141)
PROGRAMMING OF MATERIAL'S PROPERTIES OVER EXTENDED TIME INTERVALS VIA SELF-IRRADIATION PHENOMENA

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We consider theoretically the possibility of programming the ageing processes in materials via the introduction of small quantities of unstable nuclei in a material's structure. Ageing of materials is usually considered as a harmful and undesirable process which limits the lifetime of many consumer products. In certain instances, however, changing of physico-chemical properties with time constitutes a part of material's functionality. Structure transformations due to self-irradiation of materials by fission products of unstable isotopes constitute a special case of ageing processes. They are quasi-independent on environmental factors, such as heat, light, humidity, aggressive chemicals. A well-known example is metamictisation of zircon due to the decay of uranium and thorium atoms present in the mineral's crystal lattice. In our work we suggest to exploit self-irradiation phenomena to programming material's properties over extended time intervals. Using literature data on the action of ionizing radiation on different materials, we argue that incorporation of minuscule amounts of radioactive species, such as tritium, in the structure of certain polymers can lead to profound change of their physico-chemical characteristics over the times ranging from few months to several years. For instance, introducing tritium in the structure of polytetrafluoroethylene (PTFE) via substitution of a small fraction of the fluorine atoms can be used for programming radiolysis of the polymer and it's self-destruction to a certain date. The date can be preset by concentration of tritium in the material, which is typically a few mg of tritium per kilogram of the polymer. Radioactive isotopes can be incorporated in the structure of conductive polymers, such as a blend of polyaniline (PANI) with poly (vinyl chloride) (PVC). Self-irradiation can lead to significant change of electronic properties of the material due to self-doping of PANI by hydrochloric acid released by the partner polymer upon irradiation. It is known that refractive index of materials (polymers, glasses) also can vary upon the action of ionizing radiation. Therefore it might be possible to program the value of the refractive index by radioisotope engineering. Finally, self-irradiation can be applied to scheduled relaxation of internal mechanical stresses in materials. Materials with time-programmed characteristics can find numerous applications in different fields of technology. For instance, polymer or inorganic semiconductors with programmed electronic properties can be used for the design of electronic devices which can be disabled or activated at a well-defined date. Due to miniaturisation of electronic circuits, tiny quantities of radioactive substances might be sufficient for this purpose.

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Posters

PAR.P01 (Id: 115)
SEPARATION OF ^{131}Cs FROM BARIUM RADIONUCLIDES

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The implanted seeds of ^{131}Cs are successfully used in brachytherapy to treat prostate cancer and other malignant tumors. X-rays of ^{131}Cs (30 keV) make damage to and kill genetic materials of cancer cells, making it impossible for these cells to continue growing while saving or minimizing damage to the healthy tissue. Besides the radioisotope ^{131}Cs has a short half-life ($T_{1/2} = 9.7$ d), making it possible to deliver 90% of its dose energy in just 33 days compared to 204 days for ^{125}I ($T_{1/2} = 60$ d) and 58 days for ^{103}Pd ($T_{1/2} = 17$ d). In this work, the radiochemical methods of separation of ^{131}Cs from barium radioactive isotopes are presented. The targets of barium components ($BaCl_2$, $BaCO_3$, BaO) were irradiated for 3-7 days by thermal neutrons at fluxes of 5-10x10^{13} cm^{-2} s^{-1} in nuclear reactor. Irradiation formed about 10 undesirable radionuclides that represent a certain difficulty and radioactive safety problem in the separation of ^{131}Cs from radioactive solutions. Carrier-free ^{131}Cs is separated from the irradiated barium target by precipitation of Ba^{2+} cations with addition of SO_4^{2-} anions in excess and filtration of the formed precipitate. Chemical purity of ^{131}Cs was determined by γ spectrometer with HPGe detector.

PAR.P02 (Id: 146)
AN EXTERNAL CYCLOTRON TARGET SYSTEM FOR NANOPARTICLE SAMPLE ACTIVATION

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The need for toxicity assessment of industrially relevant nanoparticle materials implies that in vitro and in vivo biokinetics studies are required. Radiolabelled nanoparticles are an excellent candidate for such biodistribution measurements and are also of high interest for particle tracing and fate studies in other areas. We describe here an external target system for direct activation of dry nanoparticle samples, with He or water cooling, which is attached to an external beamline of the JRC Scanditronix MC-40 cyclotron at Ispra, Italy. A specially designed capsule is used to hold several tens of mg of nanoparticle powder material to be irradiated. The target system design and cooling system is described, and considerations regarding energy deposition, yield, heat transfer, radiation damage and radiotracer recoil are discussed.

This work was performed in support of several projects, including the European Commission's 7th framework programme "NeuroNano" project - contract number NMP4-SL-2008-214547

PAR.P03 (Id: 151)
PREPARATION OF ^{95m}Tc RADIOTRACER

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Several procedures of the preparation of the radionuclide ^{95m}Tc, a radiotracer useful for various purposes, were tested. A foil made of natural molybdenum was irradiated with protons or deuterons and after 5-6 weeks of decay the ^{95m}Tc radionuclide was separated. Three different separation procedures were tested. 1) Alkaline-oxidative fusion of the Mo target with sodium peroxide and sodium hydroxide followed by the dissolution of the melt in water and liquid-liquid extraction of Tc with methyl ethyl ketone. The organic phase with Tc was evaporated and the residue was dissolved in 2 M HNO_3. 2) The target was dissolved in the mixture of hot concentrated HNO_3 and H_2O_2. The solution was evaporated, the residue was dissolved in 0.1 M HCl and Tc was separated by extraction chromatography with a Tc selective chromatographic resin (PAN-Aliquat 336 with similar properties as Teva® Resin). 3) The molybdenum foil was dissolved in the mixture of concentrated H_2SO_4 and HNO_3 (25:1) using microwave assisted digestion. To remove sulphate ions, which would interfere in the subsequent separation, barium chloride was added to the solution, the precipitate of barium sulphate was separated by filtration, and Tc was separated from the aqueous phase by extraction chromatography as given above. The column was washed with 1 ml of 0.1M HCl and Tc was stripped with 5 ml of 8 M HNO_3. The overall separation yield was in the range of 70 to 95 %. All the three methods can be used for Tc separation. However, the following difficulties have been encountered in the first two methods. In the first method, it is necessary to repeat the alkaline fusion step since the amount of reagents in one fusion is to be limited due to a very reactive nature of the flux used. Consequently, the liquid-liquid extraction step is to be repeated several times. In the second method tested, a precipitate of undetermined composition appears during dissolution, which, unfortunately, contains about 25 % of ^{95m}Tc present in the target. The third method appeared superior over the other two methods tested, because it is fast, simple and provides a very good separation yield of technetium (> 90%).

PAR.P04 (Id: 159)
MEASUREMENT OF EXCITATION FUNCTIONS FOR (D,X) REACTIONS ON NATURAL MOLYBDENUM

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Cross sections for deuteron induced reactions on natural molybdenum leading to ^{93}Tc, ^{93m}Tc, ^{94}Tc, ^{94m}Tc, ^{95}Tc, ^{95m}Tc, ^{96}Tc, ^{96m}Tc, ^{99m}Tc, ^{99}Mo, ^{92m}Nb, ^{95}Nb and ^{89}Zr were measured in deuteron energy range 9.0-19.6 MeV on the cyclotron U-120M of the Nuclear Physics Institute AS CR. Special attention was paid to excitation function for formation of ^{95m}Tc, which is used as a tracer for determining ^{99}Tc in environmental samples, and to excitation functions and thick target yields for formation of ^{99m}Tc and ^{99}Mo, the most widespread radionuclide generator pair in nuclear medicine. If appropriate, obtained data are compared with the heretofore published cross sections.

PAR.P05 (Id: 200)
PREPARATION AND CHARACTERIZATION OF TARGET FOR ^{64}Cu PRODUCTION ON THE IBA 18/9 CYCLOTRON.

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Copper has many radionuclides (^{60}Cu, ^{61}Cu, ^{62}Cu, ^{64}Cu, and ^{67}Cu) that can be used in nuclear medicine and molecular imaging. Isotope ^{64}Cu (Iβ^{+} = 17.6 %, Eβ$^{+}_{max}$ = 653 keV, Iβ- = 38.5 %, Eβ-$_{max}$ = 579.4 keV) is a positron emitter with half life 12.7 hrs and is suitable for the preparation of radiopharmaceuticals for PET imaging. The ^{64}Ni(p,n)^{64}Cu reaction route is popular for its preparation because its entrance channel is accessible at low energies and yield of reaction is quite high. Disadvantage of the reaction used is a high price of the enriched ^{64}Ni. Thick gold or platinum target were used for target preparation by electrodeposition. Irradiation surface of target was optimized for COSTIS station. COSTIS target station was installed at the end of the external beam line of the IBA Cyclone 18/9 cyclotron. The target station has been equipped with aluminum or Nb window foil in front of the target to degrade energy to optimal energy for nuclear energy less then 12 MeV. Since the external beam line of the cyclotron has no beam diagnostic devices, several aluminum plates were irradiated in the COSTIS target station with a 5 µA proton beam for 15 min with different settings for the beam focusing quadrupole magnets. After one day decay time the plates were scanned by a TLC scanner along the horizontal and vertical central axes of the plates in order to visualize the beam shape. The settings providing most homogeneous beam spot on the target were selected and used further for the real target irradiations. Presentation will concerns of preparation, optimization of target for irradiation in COSTIS target station, separation procedure and characterization of ^{64}Cu radiochemical prepared.

PAR.P06 (Id: 213)
RECOVERY AND ISOLATION OF HAFNIUM ISOMERS FROM 176Yb TARGET IRRADIATED BY ^{4}He IONS IN OPTIMIZED CONDITIONS

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Interest to high-spin (16+) long-lived (31 y) ^{178m2}Hf isomer was established since 20 years ago because of a possibility to produce it in a micro-weight amount for consecutive studies of nuclear reactions with high-spin target and for potential application as an energy accumulator. Production of ^{178m2}Hf was described in [1] via spallation of Ta by protons at LAMP facility and via ^{176}Yb(^{4}He,2n) reaction at Dubna cyclotron [2]. Series of experiments with the ^{178m2}Hf target had been carried out due to cooperation of FLNR with European research centers and the results were summarized in [3] and refs. therein. On today, the innovative perspectives for creation of tiny energy sources support the continuing interest to ^{178m2}Hf. In the present report, we describe an optimized production of ^{178m2}Hf using the ^{176}Yb target of special construction irradiated at internal beam of the U-200 cyclotron in Dubna. The 96% enriched ^{176}Yb oxide was deposited onto water-cooled Al backing, and the target surface was inclined to the beam at angle near 10 degrees. The target thickness of 7 mg/cm^{2} has provided effective thickness in a beam direction of about 80 mg/cm^{2} in ligature weight. The ^{4}He^{+1} ions reach an energy of 35 MeV. With beam intensity limited to be lower 15 µA, the corresponding beam power does not exceed 500 W. Such conditions were safe, but the target could stand even at 3 times higher beam intensity. After irradiation the target material Yb_2O_3 was washed off from the backing with 16 M HNO_3 and anion exchange in conc. HCl solution was applied to separate Hf-fraction. With the same purpose precipitation of YbF_3 was also tested [4]. Deep purification was carried out using the anion exchange chromatography in HF solutions with an overall chemical yield upon the average 85%. Activity of the irradiation products was controlled by the γ-spectroscopy method using HPGe spectrometer. All γ-lines belonged to ^{178m2}Hf and ^{179m2}Hf were quantitatively measured and absolute yields of both high-spin isomers were determined with appropriate accuracy. Under described conditions several ^{176}Yb targets were irradiated, and, in total, 6x10^{14} atoms of ^{178m2}Hf were produced over beam time of 900 hours. The isomer cross-section and impurities content obtained in the present experiments are in a good agreement with the data published earlier [2]. Change to the internal beam at the cyclotron and more tangential beam-to-target geometry supply higher yield of the products and make the production method more economic. The position of a target inside the cyclotron camera leads to the decreased neutron flux outside and meets the requirements of modern radiation safety rules. Improving the biological shielding at the accelerator and increasing the beam current, one can expect a productivity of about $3x10^{15}$ of the $^{178m2}Hf$ isomeric material per 1000 hours of effective irradiations.

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PAR.P07 (Id: 239)
CHARACTERIZATION OF A CU SELECTIVE EXTRACTION CHROMATOGRAPHIC RESIN

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Cu isotopes (e.g. ^{64}Cu) increasingly find use in radiopharmaceutical applications, accordingly fast and reliable methods for the production of these isotopes are needed. Aim of the presented project is the characterization of a Cu selective extraction chromatographic resin for the fast and selective separation of Cu radionuclides, e.g. from irradiated Ni targets. The characterization of the resin includes the determination of k' values of Cu, Ni and other potentially interfering elements and impurities for varying acids and pH values, the influence of macro amounts of Ni on the extraction of Cu as well as the influence of other potential interferents. Based on the obtained results a method for the separation of Cu, and its purification, from irradiated Ni targets was developed and tested on simulated Ni targets.

PAR.P08 (Id: 354)
ISOTOPIC EFFECT OBSERVED BY DISTILLATION OF ^{123}I AND ^{124}I FROM IRRADIATED $^{124}TeO_2$ TARGET MATRIX

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Non-conventional longer-lived PET radionuclides with half-life higher than that of ^{18}F (109 min) are very promising PET tracers for imaging of biochemical processes or body structures requiring slower kinetics. Among them, ^{124}I (4.18 d) plays an important role. Radiochemistry of iodine radioisotopes (^{123}I, ^{124}I, ^{125}I and ^{131}I) is well known. Widely used production process of ^{124}I is based on the (p,n) reaction on highly enriched $^{124}TeO_2$ (99.4 %), and distillation of iodine from irradiated target matrix at temperatures slightly above the melting point of TeO_2 (733 C). Depending on the entrance beam energy, some amount of ^{123}I can be produced by the (p,2n) reaction (E_{thr} · 11.5 MeV). For production we have developed our own target system. Target layer is water and helium cooled. The target was irradiated at the beam angle 10 in order to increase effective thickness of the target layer and to get more favourable distribution of the beam over the target area. The entrance proton beam energy varied between 11.5 and 13.2 MeV, beam currents were 10-22 μA and irradiation times 0.5-2 h. The distillation was performed in pre-heated quartz tube (735-750 C). The radioiodine was carried out with stream of air. The flow rate was set to 25 mL/min. Evaporated iodine was trapped in 0.01M NaOH. Time of separation was 15-20 min. Radionuclidic purity and activity of produced isotopes was measured with γ-ray spectrometry using HPGe detector, in all steps of production (irradiated target, target after distillation and solution of separated product). Since the protons' entrance energy was higher then treshold of the (p,2n) reaction, we also measured activity of the co-produced ^{123}I. We have observed an interesting effect. The ratio of decay-corrected activities of ^{123}I and ^{124}I in the target matrix after the separation of radioiodine was 2-3 times higher than in the target matrix after irradiation. In accordance to this effect, we also observed that ratio of decay-corrected activities of ^{123}I and ^{124}I in the solution of separated product was proportionally lower than the ratio in target.

7. RADIATION CHEMISTRY (RCH)

Lectures

RCH.L01 (Id: 290)
EFFECTS OFM γ IRRADIATION ON POLYETYLENE ISOPHTHALATE

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Radiation methods are largely used for polymerisation and polymer modification [1,2], since irradiation induces changes in the structure of materials which can be exploited to obtain new features or to improve their performance. On the other hand, degradation of the polymer properties is observed under the combined action of ionising radiation and oxygen [3]; chain scissions and cross links are often induced [4,5] and the presence of new functional groups such as carbonyls, carboxyls, esters, hydroxyls and unsaturation is observed. Correspondingly, worsening of properties such as mechanical strength, electrical insulation power and water repellence may be registered. Therefore, the change of the chemical and physical properties of polymers under irradiation is an interesting field of research in view of future applications. In this work we discuss data on the changes of the physical properties of a polyester, polyetylene isophthalate (PEI), subjected to γ irradiation. It is a semicrystalline polymer with a structure similar to polyetylene terephthalate; in particular, the presence of an aromatic ring [6] makes this polymer a good candidate to withstand radiation effects. We studied the effects of γ irradiation on PEI for doses up to 1 MGy by means of viscosity and differential scanning calorimetry measurements, which allow to express changes in the structure in terms of variations in the molecular weight, as well as a percentage crystallinity dependent on the dose. Furthermore, positron annihilation lifetime spectroscopy supplied us information on the free volume present in the amorphous phase of the irradiated polymer.

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RCH.L02 (Id: 280)
COMPARISON OF RADIATION AND THERMAL AGEING SIMULATION PROCEDURES FOR NPP CABLES

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The accelerated radiation and thermal ageing of cables has a special importance in the type qualification test of NPP cables as it should provide for the cable ageing by an accelerated but real process of ageing, i.e., by the homogeneous oxidation of the cable polymeric materials. A comparison of three cable ageing techniques was performed on the representative NPP cables based on EVA and XLPE jacket and insulation materials. These techniques cover separated and combined radiation and thermal ageing under high dose rate (3 kGy/h) and low dose rate (5 Gy/h) at temperatures from 75 °C to 150 C. The results of mechanical and physico-chemical properties are discussed and a recommendation for qualification test procedures are proposed.

RCH.L03 (Id: 189)
PHOTO - AND RADIATION-INDUCED PREPARATION OF NANOCRYSTALLINE COPPER AND CUPROUS OXIDE CATALYSTS

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Radiation method of copper and cuprous oxide preparation produces material of high chemical purity and catalytic activity. Reduction of metal ions in aqueous solutions via UV, γ irradiation or accelerated electrons results in formation of nanosized particles of both Cu/Cu_2O with uniform spherical shapes at normal temperature. This method of preparation is very simple and relatively efficient; it requires mostly only one additive - ·OH scavenger - in order to promote process of reduction. Oxygen present in water readily oxidises formed nanoparticles of non-noble metals and causes their dissolution; therefore the solutions must be deaerated prior to irradiation. In this work, aqueous solutions containing copper sulphate or formate (0.001 to 0.01 mol.dm^{-3}) in the presence of ·OH scavenger propan-2-ol (1.3 mol.dm^{-3}) and surfactant polyvinyl alcohol PVA were irradiated.

Following radiation sources were used: Medium pressure mercury lamp with power input 400W for UV irradiation, LINAC 4-1200 for electron irradiation and ^{60}Co radionuclide source Gammacell 220 for γ irradiation. Effects of radiation were evaluated using UV-Vis absorption spectrophotometry, SEM and XRPD. Catalytic activity of prepared Cu_2O powder was studied by measuring the rate of catalytic decomposition of hydrogen peroxide H_2O_2 to oxygen at several temperatures. In separated black or brown solid phases, the presence of crystalline copper, cuprous oxide Cu_2O or their mixture was confirmed. Irradiation of 0.01 mol.dm^{-3} solutions with low doses yields pure Cu_2O, whereas at lower pH and lower copper concentration, copper particles are formed predominantly. SEM images showed that prepared particles are spherical in shape and have wide size range (50 - 500 nm in diameter, average size being cca 200 nm). The stability of colloidal copper was investigated with respect to changes of absorption spectra and pH in the presence of air. Whereas the value of pH steadily increased with time, the absorbance changed non-monotonously - in the first stage, a rapid increase of absorbance in the range from 600 to 900 nm occurred, followed by long lag-phase (absorbance remained constant) and finally in the third stage, copper absorption spectrum reappeared, but slowly decreased.

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RADIATION STABILITY AND EXTRACTION PROPERTIES OF THIACALIXARENES

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This study deals with the effects of irradiation on the stability and extraction properties of thiacalixarenes [1], the prospective liquid-liquid extractants for the extraction of actinides and lanthanides from aqueous solutions. Thiacalixarenes dissolved in various organic diluents were irradiated with ^{60}Co γ rays or accelerated electrons with energy of 4.5 MeV. The absorbed doses ranged from 0 to 100 kGy. Possible effect of the presence of aqueous phase during irradiation was also investigated. The extraction properties were studied in synergistic systems. The aqueous phase consisted of a solution of cosane (chloro-protected bis(1,2-dicarbollide) cobaltic acid) in 0.1 mol/L nitric acid; solutions of thiacalixarenes in nitrobenzen or 1,2-dichlorethane were used as the organic phase. Distribution ratios D for ^{152}Eu and ^{241}Am and their mutual separation factors were evaluated. The concentration of thiacalixarene in the irradiated samples was measured using HPLC and the activity of aqueous and organic phase was measured by γ spectrometer with HPGe detector. The results confirmed previously published data [2] which showed a strong synergistic effect in the systems containing thiacalixarenes and cosanes. This effect depends on substituents at the lower and/or upper rim. It was demonstrated that the synergic effect is independent of the used type of cosane (chloro-, resp. bromo-protected bis(1,2-dicarbollide) cobaltic acid or bis(1,2-dicarbollide) cobaltic acid) and it strongly depends on the diluent used in the organic phase. The studied systems proved not to be very stable towards the radiation. After irradiation with the dose of 20 kGy, the distribution ratios dropped to one half for thicalixarene solutions in nitrobenzene; they dropped even to one tenth for dichlorethane solutions at the same dose.

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RCH.L05 (Id: 243)
EFFECTS OF DOSE AND DOSE RATE OF γ RADIATION ON CATALYTIC ACTIVITY OF CATALASE

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Catalytic activity of catalase from bovine liver was studied using experimental device designed for kinetic measurements of hydrogen peroxide decomposition at constant temperature and pressure. The kinetics of the reaction was evaluated by measuring the volume of released oxygen. The measurement was performed at four temperatures in the range of 25 - 45 °C. Rate constants of the reaction were estimated for all four temperatures. The values of apparent activation energy were obtained from their temperature dependence in Arrhenius coordinates. Reproducibility of both sample preparation and kinetics measurement was experimentally evaluated. Solutions containing $1x10^{-3}$ - $5x10^{-6}$ g/mL of catalase in phosphate buffer were used for the study. γ irradiation was performed using ^{60}Co radionuclide source Gammacell 220 with two different dose rates 7 and 70 Gy/h and doses ranging from 350 to 1000 Gy. It was confirmed that the observed reaction of catalase with hydrogen peroxide is of the first order in the whole measured interval. Irradiation significantly decreases catalytic activity of catalase. However, apparent activation energy does not depend on the dose of radiation indicating that the mechanism of the hydrogen peroxide degradation does not significantly change with the dose, either.

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MEASUREMENT OF GROWTH CURVES OF MICROORGANISMS INFLUENCED BY RADIATION

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Relatively high attention is paid to various agents (e.g. chemical substances [1], hyperthermia and/or radiation [2,3]) influencing the growth of microorganisms. The influence of radiation on the microbial cells may be indicated not only by the survival curves, but also by the growth curves of irradiated and non-irradiated cultures. The aim of the study is to point out to some aspects of the measurements of such growth curves. The Saccharomyces cerevisiae culture from variously irradiated inoculum in liquid peptone-glucose medium was used in all experiments. Irradiation of yeast cells in solution physiological (salt) was performed with γ-rays of ^{60}Co in Gammacell 220 or with UV-irradiation using ultraviolet lamp UVH 1016-6 for various time periods. Various dose rates of both types of radiation were used. The absorbed doses reached some hundreds Gy for both γ and UV radiation. The growth curves were measured using the nephelometric data of yeast suspension in various time periods of growth. The results of many measurements showed that the slope of growth curves expressed relatively exactly the negative influence of irradiation when the standard and uniform method of evaluation was used. When the curves were fitted to the data points by the least-squares method, the average standard errors were found to be in the range from 0.6 to 0.8 % and from 0.2 to 0.5 % for γ- and UV- irradiation, respectively. The negative effect of irradiation depended monotonously and non-linearly on the applied dose of irradiation. Quantitatively different effects were found in the case of γ and UV irradiation.

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Posters

RCH.P01 (Id: 135)

RADIATION AND CHEMICAL DURABILITY OF ACTINIDE CRYSTALLINE MATRICES

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Isolation of long-lived radionuclides from the environment requires their immobilization in highly stable forms (matrices). Crystalline materials of the mineral-like type are considered to be the most promising form of the matrix incorporating high-level waste (HLW). Alpha particles and heavy recoil nuclei destroy the actinide-containing crystalline phase structure up to their complete amorphization that can effect isolation properties of the HLW matrices. The aim of the present work was to investigate radiation and chemical durability of crystalline matrices based on titanate and titanate-zirconate pyrochlore, ferritic garnet and murataite. The materials obtained contained short-lived α-emitting isotopes of ^{238}Pu ($T_{1/2} = 87.7$ years) and ^{244}Cm ($T_{1/2} = 18.2$ years). Synthesis was performed by cold pressing with subsequent sintering of the low calcined oxide mixture. The initial charge was saturated with high-activity ^{244}Cm or ^{238}Pu nitrite solutions. Compositions based on titanate $(Ca_{0.89}Gd_{0.1})(U_{0.44}Hf_{0.23}Pu_{0.22}Gd_{0.11})Ti_2O_7$ (mass fraction of ^{238}Pu is 8.7%), titanate zirconate $Gd_{1.935}Cm_{0.065}TiZrO_7$ (mass fraction of ^{244}Cm is 2.1 %) pyrochlores, ferritic garnet $Ca_{1.5}Gd_{0.908}Cm_{0.092}Th_{0.5}ZrFe_4O_{12}$ (mass fraction of ^{244}Cm is 2.0%) and murataite $Ca_{2.5}Mn_2Th_{0.41}Cm_{0.12}Ti_{7.5}Zr_{0.5}AlFeO_{24.5}$ (mass fraction of ^{244}Cm is 1.82%) were obtained. Optimum annealing temperatures for each composition varied from 1300 to 1400°C and were established in the preliminary experiments with samples which did not contain highly active actinides. The phase composition of the ceramics was made up by the basic target phases in the presence of minor phases. The change of the ceramic crystalline structure during irradiation was studied using the time-dependent changes of X-ray diffraction indicating swelling and amorphization of the lattice. Amorphization doses of the matrix phases were as follows: $2.5x10^{18}$ for titanate pyrochlore, $4.6x10^{18}$ for titanate-zirconate pyrochlore, $1.6x10^{18}$ for ferritic garnet, $2.6x10^{18}$ for sintered murataite and 2.46-$2.53x10^{18}$ α-decay/g for melted murataite. The doses were obtained during storage of materials in argon at room temperature (293 K). Chemical durability of crystalline and metamict samples was studied using the MCC-1 test (distilled water, 90°C, solution sampling for analysis on the 3rd, 7th and 14th day). Quantitative analysis of cations in solutions was made using the emission-spectral analysis and α-spectrometry. The calculated values of the leaching rates for the basic and potentially dangerous components (actinides) for all studied ceramics are within 10^{-6} to 10^{-7} g/cm^{2}day. The change of the element leaching rate in the metamict ceramics is different for different types of matrices.

RCH.P02 (Id: 356)

BEHAVIOR OF CADMIUM(II) IN IRRADIATED AQUEOUS SOLUTIONS

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The radiation removal of cadmium(II) from aqueous solutions in presence of different scavengers has been investigated. 100 mg/L of cadmium dissolved from $Cd(NO_3)_2$ requires dose of 15 kGy to be effectively removed from the system containing 1×10-2 mol/L of HCOOK as a scavenger of OH radicals. The positive effect of deaeration with N_2O or N_2 was observed in the range of lower doses. Similarly, the addition of solid promoters (bentonite, active carbon, zeolite, Cu_2O, NiO, TiO_2 and CuO) reduced the efficiency of radiation removal of cadmium. 25 % of dissolved cadmium is present in the solution before irradiation in the form of Cd(Formate)$^+$, majority in form of Cd^{2+}. Product of irradiation of the solutions containing Cd(II) is $CdCO_3$. In the system with cadmium dissolved from $CdCl_2$ radiation reduction takes place up to metallic cadmium. In the system with lower concentration of scavenger ($1×10^{-3}$ mol/L of HCOOK) no radiation removal of cadmium occurs which was validated up to an absorbed dose of 100 kGy. Systems contained organic complexants (ethylenediaminetetraacetic acid-EDTA and citric acid) were also studied. The solutions of $Cd(NO_3)_2$ containing initial concentration 27 mg/L of Cd(II) were mixed with $3×10^{-4}$ mol/L EDTA. The efficient degradation of cadmium complexed with EDTA proceeds up to 90 % at a dose of 45 kGy with addition of $5x10^{-3}$ mol/L carbonate as OH scavenger and simultaneously pH buffer (pH 10.5). Different OH radical scavengers ($1×10^{-2}$ mol/L and $1×10^{-3}$ mol/L HCOOK, $2×10^{-3}$ mol/L Na_2CO_3, $2×10^{-3}$ mol/L $NaHCO_3$, or 10 % methanol) were added to these solutions. In the presence of the carbonate, no effect of further addition of any other reagent was observed. The product of irradiation is $CdCO_3$. On the contrary, the presence of $1×10^{-2}$ mol/L of HCOOK in the solution is necessary for the radiation removal of cadmium complexed with citric acid ($1×10^{-3}$ mol/L) at pH 8. With increasing concentration of HCOOK (up to $5×10^{-2}$ mol/L) decreases the pH value necessary for the radiation induced precipitation of cadmium from the aqueous solution. The efficiency could be enhanced by adding zeolite as a solid promoter in the irradiated system.

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RCH.P03 (Id: 357)
RADIATION CHEMISTRY APPROACH TO THE STUDY OF SEDIMENTARY MICROENVIRONMENTS AS MODELS FOR THE PROTECTION OF BIO-ORGANIC MOLECULES ON THE EARLY EARTH

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Studies in radiation chemistry can provide a deeper insight into the chemical process that may have importance for chemical evolution studies that led to the origin of life. Chemical evolution encompasses the physical and chemical aspects that try to explain the origin/permanence of bio-organic compounds prior the appearance of life. Our aim is to stress the relevance of ionizing radiation as a tool for the study of formation/destruction of bio-organic compounds such as nucleic acid components. Also to remark the importance of chemical reactions that occurs at the hydrosphere/lithosphere interfaces, and study if solid surfaces can protect the organic compound adsorbed in the clay. On the other hand, it is conceivable that nucleic acids components or their precursors appeared at early stage in the evolution of this planet. Nucleic acid bases and their derivatives are important compounds in biological systems. Their synthesis and stability in environmental conditions is of paramount importance in chemical evolution. To ensure that organic compounds endured in the primitive Earth there are several possibilities: a) their synthesis was continuous and they reach steady state concentration; b) the compounds present a long half-life in the environmental conditions of the primitive Earth; c) solid surfaces protect the organic compound adsorbed in the clay. To this end, we determine the survival of bases and their corresponding nucleosides exposed to a high radiation field in an aqueous solution and adsorbed in a clay mineral. We have studied five nucleosides (cytidine, uridine, adenosine, guanosine and xanthosine) in simulated primitive Earth conditions. These samples were irradiated adsorbed in Na-montmorillonite (a clay mineral). Using X-ray diffraction, UV-Vis spectrophotometry and HPLC, the hypothesis of the protective role of solid surfaces like clay for organic compounds adsorbed on them, when exposed to external sources of ionizing radiation (γ-ray), was proved.

RCH.P04 (Id: 358)
BEHAVIOR UNDER γ IRRADIATION OF SINGLE CRYSTALS OF NACL DOPED WITH DIVALENT CATIONS

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In this work, is analyzed the interactions of the γ-radiation with two alkali halide single crystals: NaCl doped with Ca^{2+} (0.3 %) or Mn^{2+} (0.3 %), at two irradiation temperatures (298 K and 77 K), and different doses at a fix dose rate, for their possible use as dosimeters. The irradiated crystals were analyzed using their thermoluminescence properties. The application of the thermoluminescence (TL) phenomenon has been proved to be a very efficient method to evaluate energy deposited by γ radiation on irradiated samples. The productions of irradiation defects in the solid were correlated with the glow curve. F-centers were measured as function of the dose. The bleaching of the F-centers produced a decrease of the peak of the glow curve. The bleaching of the color centers associated with the reduction of the glow curve suggests that F centers are intimately involved in the production of the thermoluminescence phenomenon. Their exact role can not be centered and much discussion has still to be done. Nevertheless, these behaviors suggest that the glow peaks very probable are result from the thermal release of holes from different kinds of traps, recombining at F centers. The preliminary results show that the linear response obtained was fairly good for the system NaCl:Cd, at low irradiation doses.

RCH.P05 (Id: 120)
RADIOGRAPHIC EFFECTS DURING SELF-IRRADIATION OF CURIUM INTERMETALLIC COMPOUNDS

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Considered are changes in the elementary cell volumes versus time at room temperature and annealing behavior of some curium intermetallic compounds with platinum metals (Pt, Ir, Rh and Pd) eTc. caused by internal irradiation by α-decay products of radioactive component [1,2,3]. The intermetallide x-rays show that with increasing irradiation dose a decrease in the diffraction reflection intensity was observed up to their complete disappearance due to a crystal structure amorphization as well as disappearance of “superstructural” reflections of the Pd_3Cm cubic lattice followed by a reversion (decrease) of the lattice parameter value after several days, and recovery of the crystalline state and initial parameters of the intermetallide crystal lattice under annealing. Swelling of the lattice volume of most of the intermetallides makes up 1% achieving 3.5% in separate cases. A phenomenological model is proposed which considers peculiar features of trends of the experimental curves (with a maximum change in the physical property). The model is based on the assumption that the evolution rate in the direction of ordering of initial disordered centers is less than that of initial disordering (that is proportional to the irradiation intensity).

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8. NUCLEAR METHODS IN MEDICINE, RADIOPHARMACEUTICALS AND DIAGNOSTICS, LABELLED COMPOUNDS (RFD)

Lectures

RFD.L01 (Id: 319)
AFFIBODY MOLECULES, A NOVEL CLASS OF PEPTIDE-BASED RADIOPHARMACEUTICALS. OPTIMISING LABELLING CHEMISTRY FOR THE BEST TARGETING

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Radionuclide molecular imaging is a promising approach to detection of molecular targets for cancer therapy. Small imaging agents provide higher sensitivity and better specificity than antibodies or their fragments. Molecular display techniques (phage, ribosomal, yeast or bacterial display) allow for combinatorial selection of small peptides with high specificity to different proto-oncogene products. The use of scaffolds, robust frameworks, which hold variable amino acid in place, enables to improve both stability and affinity of targeting molecular-display selected peptides. Affibody molecules, small (7 kDa) robust scaffold peptide constitute a new promising class of high-affinity molecular probes for in vivo molecular imaging. Affibody molecules were labelled with ^{99m}Tc, ^{111}In for SPECT and ^{18}F, ^{68}Ga, ^{64}Cu, ^{76}Br and ^{124}I for PET, and demonstrated excellent imaging in pre-clinical studies. The first clinical results confirm capacity of anti-HER2 affibody molecules to visualize HER2-expressing breast cancer metastases. Robustness of the affibody scaffold enabled labelling in harsh labelling conditions without loosing specificity of target binding. It is known, that labelling has profound influence on biodistribution and targeting properties of short peptides, such as somatostatin or bombesin analogues. Biodistribution of monoclonal antibodies or their fragments is rather insensitive to labelling, though label can influence cellular retention of radionuclide in tumours or excretory organs. However, affibody molecules are four-fold larger than typical short peptide and four-fold smaller than the smallest antibody fragment, scFv. This put forward a requirement of systematic studies of influence of labelling chemistry on biodistribution of affibody molecules. This was possible due to the development of site-specific labelling of affibody molecules providing well-characterised uniform conjugates with defined biodistribution. The site-specific labelling was obtained either by an incorporation of chelators during peptides synthesis of affibody molecules or by an introduction of a single cysteine in the originally cysteine-free affibody scaffold and the use of a thiol-directed coupling. It was shown that labelling chemistry can influence biokinetics of affibody molecules. For example, the use of non-residualizing halogen labels reduced renal retention of radioactivity in comparison with radiometals. ^{68}Ga increased clearance rate in comparison with ^{111}In, when the same DOTA chelator was used. More hydrophilic chelators enabled to suppress undesirable liver uptake and hepatobiliary excretion, improving contrast in the case of abdominal metastases. It was shown, than not only hydrophilicity of a chelator, but also its position in the peptide influences imaging properties. Results of our studies show that selection of optimal labelling chemistry is essential for the development of scaffold-based imaging agents.

RFD.L02 (Id: 24)
NEW AMINO ACID SYNTHONS FOR PREPARATION OF [^{18}F] FDOPA AND α- [^{11}C] METHYL AMINO ACIDS FOR POSITRON EMISSION TOMOGRAPHY

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PET diagnostics using radiolabelled amino acids is an emerging branch of nuclear medicine. This includes visualisation and grading of brain, neuroendocrine and prostata tumours, measurement of protein synthesis rate in tumour cells, quantitative in vivo measurement of dopamine and serotonin metabolism in brain. Development of clinical applications is limited by complexicity of robotic devices necessary for multi-step preparation of the enantiomerically pure amino acids. Robust and reliable approaches not requiring sophisticated separation of radiolabelled intermediates have to be created for everyday clinical routine. Nickel(II) complexes of Schiff bases of (S)-N-(2-benzoylphenyl)-1-benzylpyrrolidine-2-carboxamide (BPB) and α-amino acids were developed as artificial analogues of pyridoxal 5'-phosphate (PLP)-dependent enzymes for asymmetric synthesis of amino acids. Current approach using BPB in nickel(II) complex of its Schiff base with glycine allows easy preparation of O-(2'-[^{18}F]fluoroethyl)-L-tyrosine ([^{18}F]FET) with 94-97% e. e. with no need for separation of diastereomers of alkylated complexes. Preparation of 6-[^{18}F]FDOPA using the same starting nickel complex gives only 77±5% e. e. without separation of diastereomers of alkylated complexes while the same preparation of α-[^{11}C]DOPA leads to 92-99% e. e. Preparation of α-methyl DOPA or α-methyltyrosine labelled with carbon-11 or fluorine-18 requires formation of quaternary chiral centre, stereochemistry of which is controlled kinetically. Kinetic control is much less efficient then thermodynamic one, thus diastereomeric excess of complexes of α-methyl amino acids is inferior. Enhancement of stereodivergent power in both thermodynamically and kinetically controlled alkylation reaction is the biggest priority in development of new metallocomplex tools for the preparation of PET amino acids. In this presentation the evaluation of new amino acids synthons bearing C_2-symmetric benzyl groups with electron-donating and electron-withdrawing substituents will be described. Compatibility of amino acid side chains protective groups will be assessed in relation to the reaction conditions used for preparation of the complexes.

RFD.L03 (Id: 140)

OPTIMISATION OF THE RADIOSYNTHESIS OF 4-[^{18}F] FLUOROMETHYL-L-PHENYLALANINE AND [^{18}F] FET VIA A SAME SCINTOMICS AUTOMATED SYNTHESIS HOTBOXTHREE MODULE.

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It was recently reported that the success of O-(2'-[^{18}F]fluoroethyl)-L-tyrosine ([^{18}F]FET) as PET tracer is limited to brain, head and neck tumours and that there is still a need for more potent amino acid analogues for peripheral tumour imaging. We hereby report on the radiosynthesis of a new tracer 2-amino-3-(4-[^{18}F]fluoromethyl-phenyl)-propionic acid (4-[^{18}F]FMP), taken up in tumours by the LAT1 amino acid transport system, performed with a customised Scintomics automatic synthesis hotboxthree module. The labelling reaction is based on Br for [^{18}F] aliphatic nucleophilic exchange on 3-(4-bromomethyl-phenyl)-2-L-tert-butoxycarbonyl-amino-propionic acid tert-butyl ester. The critical point in the synthesis pathways was the deprotection of the NCA 3-(4-[^{18}F]fluoromethyl-phenyl)-2-L-tert-butoxycarbonyl-amino-propionic acid tert-butyl ester obtained after labeling due to the highly activated ^{18}F-benzyl group and the presence of a strong acid (trifluoroacetic acid). HPLC purification and mini-column trapping followed by appropriate recovery allowed to obtain NCA 4-[^{18}F]FMP in a overall yield of 30% and with a radiochemical purity of >99 %. This tracer was found to be stable in its radiopharmaceutical formulation for at least a working day, even at high radioactivity concentrations. Additionally, for a comparative clinical study, [^{18}F]FET was synthesized applying the in literature described Tosyl [^{18}F] aliphatic nucleophilic exchange followed by deprotection and HPLC separation using the same module set-up. Here a 40% overall yield is obtained with a radiochemical purity >99%. The described automated radiosynthesis module allows the high yield production of these two amino acid analogues with minor alternations to the parameter settings of the automated system, although two different labeling reactions are performed, rendering this unit versatile for both research and clinical practice.

RFD.L04 (Id: 186)

HETEROCYCLIC LABELLED DERIVATIVES OF BETULININES

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The lupane type compounds have played a singular role among pentacyclic cytotoxic triterpenoids from the beginning of 21st century, mainly for their various biological activities[1], which include antineoplastic, antiviral, anti-inflammatory, antimicrobial activities and also hepato- and cardioprotective effects. The derivatives of betulin and betulinic acid have been tested on various tumor cells lines for their activity[2]. The heterogeneous family of triterpenoids, including highly oxidized lupane, des-E-lupane, oleanane and others derivatives, posses a significant cytotoxic and antiviral activity and was named Betulinines[3-6]. Betulinines have proved multispectral cytotoxic activity on the panel of 10 cell tumor lines of different histogenetical origin, including multidrug resistance. Our research group has three main pathways of interest. The first is a chemical path, which is focused on synthesis of new derivatives based on SARS[6]. The second one is a radiochemical path, which is specialized in synthesis of selectively labelled compounds of the most cytotoxic active compounds[7]. Finally, it is a biological path, which tests biological activities of synthesized compounds, investigates of mechanism of action and formulates of SARS conclusions[6]. During the past years have been synthesized several labelled derivatives, that have been used for investigation of mechanism, labelled by deuterium, tritium, carbon-13, carbon-14 and nitrogen-15. Recent interest of our research is synthesis of heterocyclic labelled derivatives of heterobetulinic, morolic and betulinic acid. Mentioned compound will be studied for their biological activities, especially for antitumor and anti-HIV in cooperation with LEM in Olomouc.

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RFD.L05 (Id: 10)

PREPARATION OF [^{48}V]TiO$_2$ LABELLED NANOPARTICLES FOR BIOKINETICS STUDIES.

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Engineered nanoparticles are of current interest in many applications (e.g. industry, medicine, energy). As they are produced in ever higher quantities, the need to assess their biokinetics and toxicity becomes more and more important. The aim of this study was to prepare stable, radiolabelled bioavailable TiO$_2$ nanoparticles to assess their in vivo biokinetics. The preparation of [^{48}V]TiO$_2$ nanoparticles by direct proton irradiation is described. Irradiations were performed on the Scanditronix MC-40 cyclotron at JRC Ispra, Italy with a 5 µA proton beam of 23.5 MeV energy. After 16 hours irradiation, a commercial sample of TiO$_2$ (ST-01, Ishihara Corp. Japan) yielded approx. 40 MBq of ^{48}V activity at EOB. Half of the irradiated material was processed two weeks after EOB to size-select and purify the labelled nanoparticles from free ^{48}V by centrifugation and filtration. This yielded 855 kBq of final suspension of fine [^{48}V]TiO$_2$ labeled nanoparticles (Z average = 105 nm) without free ^{48}V. In vivo results in a rat model indicated good ^{48}V label stability with less than 1% of the administered dose eliminated by renal excretion, compared to ionic ^{48}V with approximately 50% renal clearance.

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RFD.L06 (Id: 216)

IN VIVO GENERATORS - RECOIL AND CONVERSION ELECTRONS CONSIDERATIONS

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The use of radionuclides as potential therapeutic radiopharmaceuticals is increasingly investigated. An important aspect is the delivery of the radionuclide to the target, i.e. the radionuclide is not lost from the chelating agent. For in vivo generators it is not only the log K of complexation between the metal ion and the chelator that is important but whether the daughter radionuclide stays inside the chelator after decay of the parent radionuclide. In our previous work [1] we have showed that the classical recoil effect is applicable only for decays with a Q value higher than 0.6 MeV (in the atomic mass range around 100). However, Rosch et al. [2] published a result for ^{140}Nd/^{140}Pr ($Q = 0.222$ MeV) which indicated that >95% of the daughter (^{140}Pr) was lost by a DOTA chelator upon decay of ^{140}Nd. This was explained to be due to a "post-effect" by the authors. Their experiment was repeated with the ^{166}Dy/^{166}Ho potential in vivo generator to confirm that our calculations were correct. It was found that indeed some of the daughter (^{166}Ho) was liberated from the DOTA chelator, therefore indicating that the "post effect" does exist in contrast to our recoil calculations. Upon further investigation we determined that one should not only consider recoil energy levels but also the mode of decay (decay of the parent nuclide and the height of excitation states of the daughter nucleus). If decay takes place via electron capture (EC), Auger electrons are emitted resulting in a daughter radionuclide with a very high oxidation state due to the loss of these electrons from the electron shells. From the well known described biological effects of ^{125}I [3] which decays to (^{125}Te)$^{21+}$ one knows that chemical bonds in the vicinity of the decay are broken. It is therefore postulated that the EC decay of ^{140}Nd/^{140}Pr results in a ^{140}Nd daughter that destroys the DOTA chelate and is therefore released from this chelator. In the case of ^{166}Dy/^{166}Ho ($Q = 0.486$) the decay mode is via β- decay (which according to our recoil calculations is not strong enough to break the bond with the chelator) followed by dissipation of excitation energy of the excited state of the formed daughter nuclide via a branched decay - γ radiation and conversion electron process. The percentage of decay via the conversion electron process was found to maTch the percentage of the amount of ^{166}Ho that was measured to be released from the DOTA chelator in our experiment. It is therefore concluded that the recoil effect below 0.6 MeV is not sufficient to rule out release of the daughter nuclide from chelators but one needs to consider the mode of decay as well. This knowledge has important implications for the design of future radiopharmaceuticals.

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RFD.L07 (Id: 245)
IN VIVO ^{212}Pb/^{212}Bi GENERATOR USING LIPOSOMES

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^{212}Bi is a potentially interesting α radionuclide for targeted α therapy. The principle is based on the stable binding of α emitting radionuclides to cancer selective carrier molecules, such as antibodies or peptides. The challenge is to deliver the radioactive atoms to the target with the objective to find the right balance between toxicity and anti-tumour effect. Considering its short period ($t_{1/2}$ = 60.6 min), ^{212}Bi is limited to situations where the labelled carrier molecule rapidly accumulates in the target tumor. To expand the range of applications, an interesting method is to use its parent, ^{212}Pb ($t_{1/2}$ = 10.6 h), which will generate in vivo ^{212}Bi. Data in the literature show that the classical chelation approach does not work. Although the chelating agent used (DOTA) is known to form strong complexes with both Bi and Pb, a significant part of Bi escapes from the carrier molecule as a result of the radioactive transformation ^{212}Pb/^{212}Bi [1]. An interesting alternative is to use liposomes [2]. Once ^{212}Pb is encapsulated in its internal compartment, the phospholipidic membrane prevents Bi release provided that the liposome size is large enough (~ 100 nm). In this work, liposomes functionalised with DTPA at the surface are used. Once complexed with In, the In-DTPA species corresponds to the recognition site for the targeting. An active encapsulation was tested, i.e. the liposomes are preformed before the encapsulation, using a methodology implying the Chelex-100 resin. DTPA is present in the internal compartment (0.025 M) to keep ^{212}Pb in the core as an anionic complex after its uptake. The passage of the membrane was obtained using different weak complexing agents (1,10-phenanthroline-2,9-dicarboxylic acid (DCP), 2,3-dimercapto-1-propanol (BAL), sodium acetate), the one leading to the best results being acetate. Under the optimised conditions ($[Pb]_{tot} = 10^{-9}$ M (^{212}Pb activity of 700 Bq), [liposome] = 2.5 mM, 65 C), 64% of Pb was shown to be encapsulated after 1 hour leading to 1 liposome labelled per 100. The stability of the labelled liposomes was monitored in biological media using chromatographic techniques (HPLC, AF4: Asymmetrical Flow Field-Flow Fractionation) coupled with γ detection.

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RFD.L08 (Id: 326)
RADIOMETRIC DETERMINATION OF ENZYME ACTIVITIES, EXEMPLIFIED BY SEVERAL KEY ENZYMES OF THYROID HORMONE METABOLISM

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Radiometric enzyme assays are based on the conversion of radio-labeled substrates to labeled products, and on the measurement of radioactivity of either products or residual substrate after their quantitative separation. In the present studies, we aimed to establish valid assay conditions for radiometric determination of the activities of several enzymes, which are involved in the metabolism of thyroid hormones (TH): Thyroid peroxidase (TPO), the key enzyme in the biosynthesis of TH in the thyroid gland, and uridinediphospho-glucuronyl-transferase (UDPGT) and iodothyronine sulfotransferases (ST), the enzymes responsible for biotransformations of TH in peripheral tissues. Further, with the aid of such developed assays, we attempted to follow the effects of some exogenous substances - bromide and perchlorate ions as supposed goitrogenic agents, and an antidepressant drug fluoxetine (Prozac) - on the metabolism of TH in the rat. The procedure of the radiometric assay for TPO in vitro was based on the ability of TPO to oxidize [^{131}I]-iodide in the presence of H_2O_2, generated in situ by glucose oxidase, and to catalyze subsequent iodination of specific tyrosyl residues in the added thyroglobulin. The measure of TPO activity in microsomal fractions of the thyroids was the amount of radio-iodine incorporated into thyroglobulin. This was determined either after TLC separation of the incubated samples, or simply after precipitation of radio-labeled protein and measurement of ^{131}I radioactivity in separated fractions. In the radiometric assay for UDPGT in rat liver microsomes, the rate of conjugation of phenolic group of L-3,3',5'-[^{125}I]-triiodothyronine ([^{125}I]-rT3) with glucuronic acid, which was catalyzed by UDPGT was measured. Reaction mixtures, containing uridinediphospho-glucuronic acid, the substrate [^{125}I]-rT3 and samples of microsomes, were analyzed after the incubation by chromatography on micro-columns of lipophilic Sephadex LH-20. The basis for radiometric determination of ST activity in liver cytosolic fractions was the transfer of sulfonate moiety from the "active sulfate", 3'-phosphoadenosine 5'-phosphosulfate, to the substrate [^{125}I]-rT3. The extent of conversion was determined after the separation of the sulfated product from the unmodified [^{125}I]-rT3, again by Sephadex LH-20 chromatography. With the use of the described radiometric assay for TPO, we found that the influence of exogenous bromide on the TPO activity in the rat thyroids was biphasic, with regard to the extent of bromide intake in the animals. Administration of fluoxetine alone caused a significant (about 2-fold) increase in UDPGT activity. In contrast, the radiometric determination of ST did not demonstrate any significant effects of the application to the rats of fluoxetine alone, or together with T3, on the induction of these enzyme activities.

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Posters

RFD.P01 (Id: 12)
RADIOSYNTHESIS OF 6-[^{18}F]FLUORO-3,4-DIHYDROXYPHENYL-L-ALANINE USING ACETONITRILE AS A SOLVENT

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6-[^{18}F]Fluoro-3,4-dihydroxyphenyl-L-alanine (6-[^{18}F]fluoro-L-DOPA) is an analog of L-DOPA and it is used for studying metabolism of dopamine function in humans with Positron Emission Tomography. Therefore, the need for reliable and simple production of this tracer had led to the improvement of the routine production. Various electrophilic and nucleophilic methods for the synthesis of this radiopharmaceutical have been reported in past two decades. Nowadays, the electrophilic fluorodemetalation has been preferred as a reliable and simple method especially via fluorodestannylation that is stereoselective and gives a good radiochemical yield [1]. Freon 11 is widely used as a solvent to dissolve a precursor in the radiofluorination step. Because the solvent depletes ozone, it is important to use deuterated chloroform instead of Freon 11 [2]. However, this solvent is a carcinogen, therefore, it would be advantageous for occupational health to use acetonitrile instead of chloroform, because acetonitrile is a less hazardous solvent. [^{18}F]F_2 was produced by the nuclear reaction ^{20}Ne(d,α)^{18}F with 8.4 MeV deuterons from GE PETtrace cyclotron. The delivery line of [^{18}F]F_2 was flushed with 1% F_2 in neon before irradiation. The [^{18}F]F_2 was released from target gas after the bombardment (1 hr, 40 μA) to TRACERlab FX-FDOPA module that was used for the synthesis of 6-[^{18}F]fluoro-L-DOPA. N-(Formyl)-3,4-di(tert-butoxycarbonyloxy)-6-(trimethylstannyl)-L-phenylalanine ethyl ester (45 mg) was dissolved in 5 mL of acetonitrile at 5 C. The solution was bubbled with [^{18}F]F_2 for 6 min. The solvent was evaporated at 80 °C in vacuum system. 2 mL of 5 M HCl was added to the residue at 130 °C for 10 min. 2 mL of 112 mM acetate buffer was added to the residue after the reaction vessel was cooled down to 40 C. The solution was purified by HPLC on two reversed phase C18 column using the acetate buffer as an eluent. The fractions eluted from the column were monitored by UV absorption at 280 nm and γ radioactivity. The fraction of 6-[^{18}F]fluoro-L-DOPA was sterilized by filtration through a 0.22 μm membrane filter. The final product was analyzed by HPLC, GC, pH meter, dose calibrator and limulus amebocyte lysate test prior to human use. An overall radiochemical yield obtained from this synthesis was 10.52% (decay-corrected) and radiochemical purity was >99%. The total synthesis time was about 45 min. The pH was around 4. The radiochemical purity and the specific activity were greater than 99% and 800 mCi/mmole, respectively. Although the radioactivity decreased during the evaporation step of the solvent, the sufficient amounts of 6-[^{18}F]fluoro-L-DOPA can be produced for clinical use. In conclusion, the reproducibility and simplicity of the production of 6-[^{18}F]fluoro-L-DOPA were improved for a routine production by using acetonitrile as a solvent.

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RFD.P02 (Id: 53)
APPLICATION OF TRITIUM LABELED DROTAVERINE AND FUROSEMIDE FOR ESTIMATION OF DRUG/BINDING CAPACITY OF BLOOD TRANSPORT SYSTEM

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For labeling pharmacological preparations of furosemide and drotaverine we used the method of labeling by thermally activated tritium. Furosemide and drotaverine were labeled by thermally activated tritium in apparatus for tritium labeling. The optimum regime of labeling was selected. The systems of purification of tritium-labeled furosemide and drotaverine by thin layer chromatography (TLC) has been developed. Tritium-labeled furosemide was purified by TLC on silicagel in system hexane:acetone (1:1). Tritium-labeled drotaverine was purified by TLC in system isopropanol:ammonia:water (8:1:1). Application of TLC for purification of tritium-labeled furosemide and drotaverine allows to purify completely furosemide and drotaverine from by-products. The output of purified tritium-labeled furosemide was 12 %, with specific radioactivity 202 MBq/mmol. The output of purified tritium-labeled drotaverine was 36%, with specific radioactivity 1.39 GBq/mmol. We have investigated the drug-binding capacity of serum proteins of patients using the tritium-labeled pharmacological drugs of furosemide and drotaverine (no-spa). Series of groups of pediatric patients in the age of 3 to 14 years with acute and chronic hepatitis A and B have been investigated. Control group includes the conditionally healthy children of same age. The binding capacity of serum proteins was determined by binding tritium-labeled drotaverine and furosemide with serum proteins in vitro. The micromethod consists in incubation in vitro samples of 20 microliter of serum with tritium-labeled drotaverine and with tritium-labeled furosemide. After incubation serum proteins were fractionated by chromatography and tritium radioactivity bound with fraction of serum proteins was measured. It was found that at the severe form of virus hepatitis B the binding capacity of serum proteins actually of all investigated patients in a stage of peak of disease was reduced in comparison with control group. At the moderate form of acute virus hepatitis B the decrease of binding capacity of serum proteins was observed at 69 % of patients. We have also investigated dynamics of changes of binding capacity of serum proteins during standard therapy of hepatitis A and B. We found, that during convalescence at application of standard therapy the binding capacity of serum proteins comes nearer to values of control group. Thus dynamics of changes of binding capacity of serum proteins at patients with hepatitis B differed from dynamics at patients with hepatitis A. It was found that children with an acute virus hepatitis B after basic treatment have an increased level of binding of tritium-labeled drotaverine by serum proteins, and at children with acute virus hepatitis A it does not occur. Obtained results in the whole indicate the reducing of binding capacity of serum proteins at virus hepatitis that allows to determine the optimal strategy of a pharmacological load on patient organism and thus to optimize the treatment of patients.

RFD.P03 (Id: 66)

IN VITRO STUDY OF PROTEINS SURFACE ACTIVITY BY TRITIUM PROBE

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The behavior of biologically active compounds in aqueous/oil systems is usually used as a model of their interaction with the lipid membrane of the cells. Thus, study of the processes proceeding in the system of two immiscible liquids is a central question of the research. In this work we propose to use tritium-labeled bioactive compounds and scintillation phase method [1] for studying adsorption and partition of the substances in aqueous/oil systems. Two water-soluble proteins, lysozyme and human serum albumin, were under the test. Tritium label was introduced into the protein molecules by tritium thermal activation technique. The labeling was carried out according to the improved procedure [2] that allowed increasing the specific radioactivity and the yield of labeled compound. Keeping target temperature during the reaction with atomic tritium at 295 K was an innovation. Variation of the W-wire temperature, the gas pressure and the exposition time leads to optimum conditions for tritium introduction into the proteins. Tritium-labeled proteins were purified by dialysis and size exclusion chromatography. Scintillation phase experiment was conducted for three organic liquids that were used as scintillators, namely p-xylene, octane and octanol. To provide octane and octanol with scintillation properties naphthalene and 2,5-diphenyloxazole were added. The experiment was carried out according the procedure described in Ref. [1]. Partition coefficients and adsorption isotherms of the proteins in aqueous/oil systems were obtained. The profound effect of the organic phase nature on the protein behavior was observed. The presence of ionic surfactants in the system can influence on protein behavior. Tritium probe is a unique instrument that allowed experimental studying interference of surfactants and proteins in aqueous/oil system. We observed that e.g. lysozyme can be substituted from water/p-xylene interface by both cationic and anionic surfactants and vice versa protein substituted surfactant. But the mechanism of protein-surfactant interaction depends on the charge of protein and surfactant. The reasons for the different results of substitution will be discussed.

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RFD.P04 (Id: 70)

APPLICATION OF THE METHOD OF THERMALLY ACTIVATED TRITIUM FOR LABELING ISONIAZIDE

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For labeling the pharmacological preparation of isoniazide we have used the method of labeling by thermally activated tritium. Isoniazide was labeled by thermally activated tritium in apparatus for tritium labeling. The effect of the labeling procedure on isoniazide has been investigated. During labeling the change of color of isoniazide was observed that indicates partial destruction of isoniazide. The optimum regime of labeling was selected. The system of purification of tritium-labeled isoniazide by thin layer chromatography (TLC) has been developed. Tritium-labeled preparation of isoniazide was purified by TLC on silicagel in the system ethyl acetate:isopropanol:liquid ammonia (concentrated):acetone (15:20:5:2). Application of TLC for purification of tritium labeled preparation allows to purify completely isoniazide from by-products. The output of purified tritium labeled preparation of isoniazide was 41.4%. The obtained preparation had specific radioactivity of 468.5 MBq/mmol, radiochemical purity of the preparation was 95 %. TLC purification seems inexpensive and fast method, suitable for purification of tritium-labeled isoniazide. Thus, the used method allows to obtain tritium labeled preparation of isoniazide suitable for medical and biologic research. Received tritium-labeled preparation of isoniazide is supposed to be used in studies of binding capacity of blood transport proteins of patients with various pathological conditions.

RFD.P05 (Id: 73)
ASSESSMENT OF LABELLED PRODUCTS WITH DIFFERENT RADIOANALYTICAL METHODS: STUDY ON 18F-FLUORINATION REACTION OF P-[^{18}F]MPPF

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The crude reaction mixture consisted of p-[^{18}F]MPPF and 2-4 labelled impurities eluting after the product fraction, and the reverse-phase HPLC method reported in the literature [1] failed sometimes to separate p-[^{18}F]MPPF from its radioactive by-product with close retention time. Analytical HPLC was based on similar chromatographic conditions. By comparing it to preparative results it could be used in the estimation of radiochemical incorporation yield of p-[^{18}F]MPPF. However, radio-TLC was not a reliable method for the radiochemical analysis of crude p-[^{18}F]MPPF: the sample in DMSO was not well distinguished on silica plates and there was labelled impurity with same R_f-value. Heating mechanism had no significant effect on the composition of labelled compounds. Based on the molecular structures of the substituted benzamides, p-MPPF and its precursor p-MPPNO$_2$, the reaction of basic nitrogen on piperazinyl moiety and/or amide hydrolysis could be reasonable explanation for the formation of side-products. Corresponding fractionated masses, molecule peaks with 192 and 120 smaller m/z ratios, were also measured in the ESI-MS studies of the inactive reference compounds. In the preliminary LC-(ESI)-MS tests of the radiolabelled product these peaks were not detected, suggesting different kind of decomposition of labelled product and/or precursor. However, p-[^{18}F]MPPF was identified with m/z ratio of 435. More than one radioanalytical method is needed to evaluate reliably the radiochemical incorporation of p-[^{18}F]MPPF. Identification of various labelled impurities by radiochromatographic methods without broad reference material is difficult and characterisation by more complex method, MS, is needed. The ion trap MS seemed to be sufficient for the qualitative analysis of p-[^{18}F]MPPF. Reliable identification of the labelled side-products, however, needs optimisation of the radioLC-method.

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RFD.P06 (Id: 182)
MIBG - STRUCTURE REVISITED

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The synthesis, labeling and careful spectroscopic investigation of meta-iodo-benzyl guanidine has been performed. As result of ESI MS and single crystal X-ray diffraction the correct structure was found to be meta-iodo-benzyl guanidine hemisulfate hemihydrate (2MIBG.H_2SO_4.H_2O).

RFD.P07 (Id: 187)
SELECTIVELY LABELLED TARAXASTANE AND LUPANE DERIVATIVES

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Triterpenes are a group of natural compounds. They are studied for their wide spectrum of biological activities: antitumor, antimutagenic, antiinflammatory, antiallergy, endocrine, immunomodulatory, antiviral, hepatoprotective, cardioprotective, antithrombotic activities or activities on central nervous system. [1] Nowadays the investigation of antitumor drugs is really important because over one million people get cancer each year and this quantity is increasing every year. Some of the most important triterpenes are betulin and betulinic acid and their derivatives [2], which have demonstrated selective cytotoxicity against a number of specific tumor lines, HIV disseas, a variety of infectious agents such as malaria and bacteria, and the inflammatory process in general. The derivatives and potencial drugs from betulinic acid can be used to kill and/or inhibit multiplication of tumor cells. This type of drugs need to have high cytotoxic activity, low toxicity and have to be easily aviable. We are focusing on the synthesis of semisynthetic derivatives of betulin and betulinic, heterobetulinic and morolic acids (lupane and taraxastane type) selectively labelled by deuterium ^{2}H and tritium ^{3}H, ^{13}C, and ^{14}C. These labelled compounds are essential for supporting the pharmacokinetic and toxicological evaluations which help to better understand the metabolic pathways which are proceeding in investigated organism. We trust that labelled derivatives are helpful in the investigation of mechanism of action and indentification of primary target. All the synthesized compounds are tested on the board of tumors lines for their cytotoxic activity in vitro and in vivo in LEM, with the aim to obtain new anticancer drugs.

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RFD.P08 (Id: 274)
RADIOIODINATION AND BIODISTRIBUTION OF THE MONOCLONAL ANTIBODY TU-20 AND ITS SCFV FRAGMENT

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The ability of the monoclonal antibody TU-20 and its scFv fragment to bind specifically to the C-end of the class III β-tubulin makes these substances useful as potential diagnostics for neurodegenerative diseases - especially peripheral neuropathies. TU-20 and its scFv were labeled with ^{125}I and ^{123}I by chloramine-T (with average yield 0.75 and 0.50, respectively). Radiochemical purity and stability was revealed by gel filtration (decrease to 80 % and 50 % in two months, respectively). Immunoreactivity of the labeled TU-20 was determined by ELISA - the range of the preserved immunoreactivity varies from 60 % to 95 % in accordance to the used radiolabeling process. RIA and affinity coupling analytic methods were specifically designed with focusing on specifics of the antibody and its fragment. The results of RIA differ in depandance on the type of the reaction vessel (glass or polystyrene) and the affinity coupling results depend on the experimental arrangement - in the baTch or at the column. Fragmentation of the labeled antibody and its fragment was estimated by bis-tris gel electrophoresis followed by silver staining and autoradiography (over 95 % of radioactivity bound in the substances). The antibody binding in tissue slices was studied in vitro by immunohistochemistry. The Purkinje cells were observed conjugated with the radiolabeled substances - either TU-20 or its scFv fragment in the area of the cerebellum. In vivo biodistribution of ^{125}I-TU-20, ^{125}I-scFv TU-20, ^{123}I-scFv TU-20 and Na^{125}I was proceeded in normal mice (wild type C57B/6/J). Both biomolecules labeled by ^{123}I were also proved in an imaging biodistribution study with use of the SPECT camera. Finally, a transgene population G93A1 Gur was used for comparative study to show the different behaviour of the substances in a normal mouse and in the modified organism with amyotrophic lateral sclerosis (ALS). The most part of differences is observed in the area of the muscles, rostal and caudal spinal cord. In summary, the monoclonal antibody TU-20 and its scFv were successfully radioiodinated and afterwards analysed by several quality control methods and biodistribution studies which confirmed their preserved or expected immunoanalytical characteristics in normal and genetically modified organism.

RFD.P09 (Id: 320)
TECHNOLOGY OF DTPA CONJUGATATION IMMUNOGLOBULINS AND THEIR LABELING WITH ^{90}Y AND ^{177}Lu RADIONUCLIDES

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Model study of the γ-immunoglobulin G (Human or Bovine IgG, polyclonal antibodies) and bifunctional chelating agent diethylenetriaminepentaacetic acid dianhydride (cDTPAA) conjugation was carried out. Various values of the cDTPAA/antibody conjugation ratio (15/1, 40/1, 105/1, 125/1 and 250/1) and the weight concentration of IgG 10, 5 and 1 mg mL^{-1} in 0.1 mol L^{-1} phosphate buffer (pH 7.3) and 0.1 mol L^{-1} carbonate buffer (pH 8.5) were used. Further, the labeling conditions of the DTPA-IgG conjugate by radionuclides ^{90}Y and ^{177}Lu were optimized and the labeling yield, the conjugate degrees of prepared radionuclide-DTPA-IgG conjugates were determined. The DTPA-MEM-97 (monoclonal antibody) was another tested conjugation system. This system was investigated at above-mentioned conjugation ratio of the cDTPAA/antibody and 2.5 mg mL^{-1} MEM-97 (the weight concentration) in 0.1 mol L^{-1} phosphate buffer and 0.1 mol L^{-1} carbonate buffer. Incubation time of the immunoglobulin conjugation, analogous to the monoclonal antibody MEM-97, was obtained after 30 minutes from mixing of individual components. The labeling yield of radionuclide-DTPA-antibody conjugate higher than 80 % was achieved. Higher values of conjugation degree of radionuclide-DTPA-antibody conjugate were achieved in 0.1 mol L^{-1} carbonate buffer, pH 8.5. It follows that the 0.1 mol L^{-1} carbonate buffer is suitable for the studied conjugation systems. This study has shown that the labeling yield as well as the conjugation degree of tested systems depend on the amount of antibody substance, on the bifunctional chelating agent/antibody conjugation ratio and on pH value of buffer in which the conjugation was carried out.

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RFD.P10 (Id: 324)
COMPARISON OF BIFUNCTIONAL CHELATES FOR Ga RADIOISOTOPES

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Both ^{67}Ga ($t_{1/2}$ 3.26 d) and ^{68}Ga ($t_{1/2}$ 68 min) are radioisotopes with appropriate emissions for nuclear imaging. ^{68}Ga is of particular interest due to its potential availability from a ^{68}Ge/^{68}Ga generator. The short half-life of ^{68}Ga requires the use of targeting vectors with fast localization such as small peptides. In addition, bifunctional chelates (BFCs) that facilitate fast radiolabeling under mild conditions and produce a high specific activity (SA) product in high radiochemical yield (RCY) that does not require time-consuming post-radiolabeling purification are needed. Two promising chelating moieties for gallium radioisotopes are Oxo (1-oxa-4,7,10-triazacyclododecane-

4,7,10-triacetic acid) and PCTA (3,6,9,15-tetraazabicyclo [9.3.1]pentadeca-1(15),11,13-triene-3,6,9-triacetic acid). The radiolabeling efficiency, SA and stability of the BFCs of Oxo and PCTA were compared directly to analogous BFCs of DOTA (1,4,7,10-tetraazacyclododecane-1,4,7,10-tetraacetic acid) and NOTA (1,4,7-triazacyclononane-1,4,7-triacetic acid). Small animal PET imaging and biodistributions were also studied to compare the in vivo clearance of the ^{68}Ga radiolabeled BFCs. Advantages with respect to RCY, SA, and stability were noted for the BFCs of PCTA and NOTA. An RGD-containing peptide was conjugated to three different BFCs, two commonly used BFCs of DOTA and NOTA and the novel BFC of PCTA. The resulting constructs were evaluated to ascertain the potential of the BFCs for ^{68}Ga peptide imaging. RCYs of >95% were achieved within 20 minutes using the PCTA and NOTA conjugates when the reaction was done at either room temperature or with mild heating (37 C). Further purification was not needed under these conditions. SAs of ≥0.5 mCi/nmol were achievable. The radiolabeling reaction required heating at higher temperatures (80 C) or extended reactions times at 37 °C in order to obtain equivalent RCY and SA with the DOTA conjugate. A challenge with apo-transferrin for 1 h showed that both the PCTA and NOTA Ga-radiolabeled conjugates were stable whereas the DOTA conjugate lost ~15% of Ga. In conclusion, BFCs of PCTA and NOTA were found to have superior Ga radiolabeling and stability properties compared to the BFC of DOTA. Cell and small animal imaging studies are planned to examine the effect of the BFCs on the receptor binding and biodistribution of Ga-radiolabeled peptide conjugates.

RFD.P11 (Id: 327)

RADIOMETRIC ENZYME ASSAYS: DEVELOPMENT OF METHODS FOR EXTREMELY SENSITIVE DETERMINATION OF TYPES 1, 2 AND 3 IODOTHYRONINE DEIODINASE ENZYME ACTIVITIES

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We elaborated novel, reliable methods for extremely sensitive radiometric determination of enzyme activities of iodothyronine deiodinases (IDs) of types 1, 2 and 3 in microsomal fractions of different rat and human tissues, as well as in homogenates of cultured mammalian cells. These enzymes catalyze selective 5'- (outer ring) and 5- (inner ring) monodeiodinations of iodothyronines and play crucial roles in the biotransformations of thyroid hormones (TH). The newly developed radiometric assays for IDs were based on the use of appropriate high-specific-radioactivity ^{125}I-labeled iodothyronines as substrates; high-performance TLC separation of radioactive products from the unconsumed substrates; film-less autoradiography of radiochromatograms using storage phosphor screens; and quantification of the separated compounds with a BAS-5000 (Fujifilm Life Science Co.) laser scanner. This methodology enabled us to determine IDs enzyme activities in the range as low as 10-18 katals. For the proper measurement of the individual IDs enzyme activities, we found out first the optimum assay conditions, including the concentrations of the respective radioactively labeled substrates, appropriate concentrations of thiol cofactor, the amount of total protein and enzyme concentration in the incubation mixtures, and suitable incubation times. Further, we demonstrated the applicability of our sophisticated methods by following the alterations of IDs activities induced in cultured rat astroglial cells by a series of purinergic agonists, retinoic acid, and their combination. In the case of ATP as a representative of purinergic agonists, we determined also time-course and dose-response curves to characterize in more details the induction of each type of deiodinase by purines. The introduced radiometric assays proved to be very sensitive and rapid and, at the same time, reliable and robust. Acknowledgments:

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RFD.P12 (Id: 328)

^{125}I-LABELLED IODOTHYRONINES: USEFUL TOOLS FOR STUDIES OF EFFECTS OF AN ANTIDEPRESSANT DRUG FLUOXETINE IN THE RAT

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With the use of adapted radiometric enzyme assays for iodothyronine sulfotransferases (ST) and uridine 5'-diphosphoglucuronyltransferase (UDPGT), as well as of our newly developed radiometric assays for iodothyronine deiodinases (IDs) of types 1, 2 and 3 (D1, D2 and D3), we studied the interaction of an antidepressant drug fluoxetine with the metabolism of thyroid hormones (TH) in the rat. TH are supposed to control the activity of some neurotransmitters (e.g., serotonin), which are hypothetically involved in the pathogenesis of depressive illness. One of the pathogenic factors of depression might be inadequate activities of brain IDs that could lead to local insufficient concentration of 3,3',5-triiodo-L-thyronine (T3). This hypothesis led to the development and production of a new group of non-tricyclic antidepressant drugs known as selective serotonine re-uptake inhibitors; fluoxetine is the most frequently used representative of this group. Effects of subchronic administration (for 25 days) to Wistar rats of fluoxetine by itself, or in combination with T3 were followed. The measurements of ST activities in liver and kidney cytosolic fractions did not demonstrate any significant effects of the administration of fluoxetine, alone or together with T3, on the induction of these enzymes. In contrast, in samples of liver microsomes of rats treated with fluoxetine, we found about two-fold higher UDPGT activities in comparison with control rats. Even more profound changes in enzyme activities were found in case of

IDs, especially in the pituitary and cerebellum. The treatment of rats with fluoxetine alone caused a moderate increase in D2 and, in turn, a slight decrease in D3 activities in cerebellum and some other regions of the CNS. No significant changes in D1 activity were detected. On the other hand, the administration of T3 alone caused, in accordance with our expectation, a substantial decrease in pituitary D2 activity and a simultaneous increase in D1 and D3 activities practically in all tissues studied.

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RFD.P13 (Id: 336)
PRODUCTION AND QUALITY CONTROL OF ^{177}Lu FOR LABELLING

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Lutetium-177 is a radionuclide used in nuclear medicine for therapeutic application. Both β and γ radiations emitted from ^{177}Lu allow imaging of the therapeutic radionuclide without need of adding any other radionuclide. It can be prepared in the nuclear reactor via direct (n, γ) activation from enriched lutetium target or via indirect neutron activation from ytterbium oxide. In this case, carrier free ^{177}Lu is prepared, however, separation from irradiated target is necessary. ^{177}Lu decays with a half life of 6.7 days. The research was focused on development of the semi-automatic module for processing the neutron irradiated targets and on optimizing the conditions of production and quality control of the ^{177}Lu radiopharmaceutical precursor. Enriched lutetium oxide in 5 M nitric acid was put into the titanium vial and evaporated to dryness. The screwed and tightened vial was irradiated for 96 hours in LVR-15 nuclear reactor (reactor power 7-9 MW) at the approximate neutron flux of $5x10^{13}$ n.cm^{-2}s^{-1}. The irradiated penal was allowed to cool for two days and then processed using semi-automatic module. Titanium ampoules were opened and filled subsequently by four aliquots 0.5 - 1.5 mL of nitric acid 1mol/L in order to dissolve irradiated target. All aliquots were collected, evaporated to dryness and re-dissolved with hydrochloric acid 0.01 mol/L. Final solution was passed through 0.22 mm filter and after dispensing, final solution was autoclaved for 20 minutes at 125 C. All production steps were operated using touch screen set-up on the front wall of the hot cell. The radioactivity was measured by ionization chamber Atomlab 100. The radionuclidic purity was determined by γ spectrometry. The radiochemical purity was determined by radio-TLC with ITLC-SG strips as a solid phase. The method consisted of two steps: Test for insoluble and colloid forms - liquid phase 0.15 M NaCl in 0.02 M HCl and the test for lutetium organic forms - liquid phase mixture of $NH_2OH : CH_3OH : H_2O$ (0.2 : 2 : 4) . The gas-flow proportional counting tube (Minigita, Raytest) was used for detection. The pH was measured by microelectrode Hamilton Biotrode. Bacterial endotoxins test was carried out using kinetic turbidimetric methods. The chemical impurities of Ti and Fe were checked by ICP for couple of baTches (external collaboration with Nuclear Research Institute Řež, plc.). The specific activities of ^{177}Lu at the end of irradiation ranged from 100 to 264 GBq/mg (2.9 to 7.1 Ci/mg), Specific acrivity of ^{177}Lu at the time of production ranged from 61 to 86 GBq/mg (1.6-2.3 Ci/mg). The γ-spectrometric measurements showed the content of ^{177m}Lu at the level 0.005% at the time of production. Content of Ti ranged from 0.004 to 0.385 μg/GBq of ^{177}Lu and content of Fe ranged from 0.023 to 0.708 μg/GBq of ^{177}Lu at the time of production. The semi-automatic elution device for production of ^{177}Lu was developed, tested and the operation qualification was carried out. Manufacture process was validated on three baTches of ^{177}Lu product. Methods for quality control were verified and GMP compliance documentation is currently under preparation.

9. SEPARATION METHODS, SPECIATION (SEP)

Lectures

SEP.L01 (Id: 241)
NATURAL ORGANIC MATTER (NOM) CHARACTERIZATION IN URBAN - AND NUCLEAR-WASTE IMPACTED WATERS USING MULTI-RESPONSE FLUORESCENCE

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It is well established that natural organic matter (NOM) plays an important role in metal speciation, in the transport of radioactive contaminants, in the acid-base control of natural waters, eTc. NOM originates from various natural and human-impacted processes, and its constituents feature various molecular sizes, functional groups, and other properties which could be important for metal or radionuclide speciation. In this work, NOM impacted by two contrasting human impacts was analyzed using by multiresponse fluorescence, decoupled with the multi-way resolution routine PARAFAC. In the first site at Chalk River, Ontario, Canada, water downstream from a former low-level waste infiltration pit and deep groundwaters were sampled. The second site is surface water from the Grand River in Cambridge, south-central Ontario, Canada, which is impacted by urban activities and agriculture. Our analysis was done on selections of raw water, fractions isolated by ultrafiltration, and solid phase aqueous extracts (SPE). The fluorescence spectra of the NOM, resolved with PARAFAC, showed three common features: humic-like components, at excitation/emission wavelengths 325-350/450-475 nm, fulvic-like components at 325/380-420 nm and protein-like components, at 275/300 nm. Most of the NOM from the urban-impacted sites and the clean Chalk River site was submitted to ultrafiltration, with >4% of the total in the large fraction (colloidal range, larger than 5000 Da). The proportion of colloidal-sized material in the NOM was substantially higher in the Chalk River contaminated water, with ~18-26% of the total. The protein-like components in the contaminated Chalk River water were largely removed by ultrafiltration, but less so in the clean Chalk River sample and the urban-impacted waters. SPE preferentially removed the protein-like component in the contaminated Chalk River water (typically 89-95% signal decrease), but had a limited effect on humic-like and fulvic-like components. The fulvic-like and humic-like components of the urban-impacted samples and from Chalk River behaved fairly similarly, exhibiting a slight enrichment of humic-like material in the large (colloidal) fraction. Finally, addition of metals (Cu and Co) to Chalk River sample colloidal fractions changed the fluorescence signals for the humic-like and fulvic-like components, but not the protein-like components. In contrast, there was no significant change in signal with addition of Cs. In conclusion, multiresponse fluorescence is a promising technique to determine the origins and dynamics of NOM in field samples, with potential application to metal (Cu) and radionuclide binding (with Co, but not so for Cs). Other new applications will be discussed.

SEP.L02 (Id: 46)
EFFECT OF SHORT CHAIN CARBOXYLIC ACIDS ON U(VI) SORPTION ON SILICA AND RUTILE STUDIED BY THE USE OF CAPILLARY ZONE ELECTROPHORESIS

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Capillary zone electrophoresis (CZE) was used to study of uranyl sorption on silica and rutile in the presence of short chain carboxylic acids. These acids are chosen as they may be released into the environment by decay of plant, animal and microbial tissues [1]. They can be also used as simple models of more complicated natural organic matter. First, separation and simultaneous determination of a number of short chain carboxylic acids (oxalic, formic, acetic and propionic) and U(VI) with direct UV detection is developed for analysis of solutions after the U(VI) sorption by CZE. From literature data it is known that complex formation of U(VI) with carbonate, used as a carrier electrolyte, allows U(VI) to be determined, as negatively charged species, in a single run with organic acid anions [2]. Matrix effect of Si(IV) (possible silica dissolution products) and perchlorate (added for constant ionic strength in sorption experiments) on the separation of U(VI) and organic acids is investigated. The influence of methanol in carrier electrolyte on separation selectivity of given mixture is also studied. Under the chosen conditions calibration plots are linear in two ranges of concentration from $\sim 1\times10^{-5}$ to 1×10^{-3} M for oxalate, acetate, propionate, U(VI) and $\sim 1\times10^{-4}$ to 1×10^{-3} for formate. Accuracy of the procedure is checked by the “added-found” method in standard mixture solutions. Relative standard deviation is within the range of 2-10% and the recovery is in the range of 90-110%. This method is applied for the analysis of real solutions after U(VI) sorption on silica and rutile in the presence of short chain carboxylic acids. The sorption experiments indicate that given organic acids decrease uranium sorption either on silica or on rutile. These experiments demonstrate that short chain carboxylic acids can influence the mobility and chemistry of uranium in the environment.

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SEP.L03 (Id: 313)
HETERONUCLEAR HYDROLYSIS COMPLEX OF THORIUM(IV) AND IRON(III)

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The solubility of the Th(IV)/Fe(III) system has been studied as function of pH in the range 2.00-3.50. In the individual systems of thorium(IV) and iron(III) precipitation takes place due to formation of hydrolysis products. However, in the mixed Th(IV)/Fe(III) system precipitation of ferrihydrite takes place at low pH value, pH = 2.00, whereas at higher pH no precipitation was observed after 20 months. The mixed heteronuclear complex of composition $[Th_2Fe_2(\mu_2\text{-}OH)_8(H_2O)_{12}]^{10+}$ was formed in solution, with Th-Th, Th-Fe and Fe-Fe distances of 3.95 and 3.949; 3.42 and 3.4; 3.05 and 2.921 Å as determined by EXAFS and LAXS, respectively. Two and four line ferrihydrite was formed in solutions at low pH values, 2.00-2.30, as identified by X-ray diffraction (XRD). SEM analysis of these precipitates showed that some of them contained only iron, whereas the others contained both iron and thorium. Mixed Th/Fe system showed increased solubility what may affect the design of tanks for spent nuclear waste.

SEP.L04 (Id: 229)
NEW POLAR FLUORINATED DILUENTS FOR DIAMIDE LIGANDS

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Nuclear waste management is nowadays based on the usage of light hydrocarbon liquids as diluents in extraction technology. PUREX process deals with tributylphosphate solution in n paraffins; various extraction processes developed for high level waste treatment also use light hydrocarbon diluents (e.g. dodecane) to be PUREX-compatible. Heavy polar diluents that are widely studied for purposes of extraction are an alternative to light diluents. The most attractive among them are fluorinated diluents. Polar fluorinated diluents have some advantages over hydrocarbons, e.g. high chemical and radiation stability, high density, low fire risk. Fluorinated compounds of various classes (ethers, esters, nitroaromatic compounds, fluorinated sulfones) have been tested as possible diluents for neutral extractants. The using of polar fluorinated diluents in solvent extraction allow to significantly increase the extraction capacity and solubility of neutral ligands. In the present work new polar fluorinated aromatic-free compounds were tested as diluents for various diamide extractants. New diluents - 2,2,3,3,4,4,5,5,6,6,7,7-dodecafluoroheptylmethyl ether (DDFHME), 2,2,3,3,4,4,5,5,6,6,7,7-dodecafluoroheptyl acetate (DDFHA) 1,1,2,2,3,4,4,10,10,11,11,12,12,13,13-hexadecafluoro-6,8-dioxatridecane (Formal-2) were compared with well known meta-nitrobenzotrifluoride (F-3) and trifluoromethylphenyl sulfone (FS-13). The extraction ability of diamides of dipicolinic acid, diamides of 2,2'-dipyridyl-6,6'-dicarboxylic acid, diamide of diglycolic acid in polar diluents towards some radionuclides and post-transition metals were studied. It was found that the extraction ability of diamide ligands could be highly increased while using DDFHME and Formal-2 as diluents.

SEP.L05 (Id: 18)
SOLVENT EXTRACTION STUDIES FOR THE SEPARATION OF RADIOARSENIC, RADIOGERMANIUM AND RADIOGALLIUM

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Solvent extraction techniques were developed to separate ^{72}As from bulk amounts of proton-irradiated germanium dioxide targets, ^{68}Ge from proton-irradiated gallium(III) oxide targets and ^{68}Ga from parent ^{68}Ge. Several methods have been reported to isolate arsenic radionuclides from irradiated Ge and germanium dioxide targets [1,2]. In this work the germanium dioxide target, irradiated with 17 MeV protons, was dissolved in either HCl or KOH. Then the ratio of As(III) to As(V) was determined via thin layer chromatography using ^{60}Si phase plate as a stationary phase and a mixture of sodium bitartrate/methanol in the ratio of 3:1 as the mobile phase. Our separation method utilizes the observation that arsenic (III) can be extracted selectively and quantitatively by cyclohexane from hydrochloric acid solution containing alkali iodide [1]. The influence of concentrations of different acids and of KI as well as the effect of various organic solvents was investigated. Optimally, the extraction of As(III) into cyclohexane occurred with 4.75M HCl and 0.5M KI, and its back-extraction into the aqueous phase with water containing 0.1% hydrogen peroxide. The overall radiochemical yield of the radioarsenic separation was about 95±2%. The method was used in the separation of high purity ^{72}As, a potentially useful positron emitter, formed via the ^{72}Ge(p,n)-reaction. An optimised method of separation of the radionuclide ^{69}Ge, used as a tracer in gallium/germanium separation studies, was worked out. The gallium (III) oxide target, irradiated with 17 MeV protons, was dissolved in 8M sulphuric acid and used as a stock solution. To an aliquot HCl was added and its concentration adjusted to 0.4M in order to transfer germanium to the chloride form. With this the extraction of radiogermanium using toluene was then studied at varying concentrations of H_2SO_4. From a solution of 8M sulphuric acid and 0.4M HCl the extraction was maximum. Using the optimised separation, the radionuclide ^{68}Ge, formed via the ^{69}Ga(p,2n)-reaction, was separated from a gallium (III) oxide target irradiated with 45 MeV protons. Further on, the separation of ^{68}Ga from the parent ^{68}Ge was investigated. The method is based on anion exchange solvent extraction using Aliquat 336 in xylene from hydrochloric acid [3]. Aliquat 336 in xylene has been used for the first time for separations involving radiogallium and radiogermanium. Optimisation studies were related to the influence of HCl concentration and different solvents. The solvent extraction using Aliquat 336 in xylene led to a high separation yield of ^{68}Ga from its parent ^{68}Ge. The time of separation was short, about 20 min, the contamination from ^{68}Ge was less than 0.1 % and the final product was obtained in 0.5M KOH.

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SEP.L06 (Id: 322)
NOVEL SOLID-PHASE EXTRACTANTS FOR RADIONUCLIDE PRECONCENTRATION

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Preconcentration of radionuclides is a necessary stage for their accurate determination in aqueous natural and technological solutions. Sorption methods are the most perspective for selective recovery and separation of radionuclides from other elements. Various sorption materials containing complexing groups have been widely applied due to their selectivity and high degree of recovery. It is especially important for radionuclide preconcentration from high salinity nitric acid solutions generated under the reprocessing of spent nuclear materials. The solid-phase extractants have gained considerable attention for preconcentration of trace elements. Special interest is given to the solid-phase extractants prepared by non-covalent immobilization of ligands on the surface of solid supports. For actinide and lanthanide preconcentration some organophosphorus and other ligands (tri-n-butylphosphate, carbamoylmethylphosphine oxide, malonamides, amines, macrocyclic compounds, eTc.) have been widely used. These reagents are usually immobilized on silica-based matrices, polystyrene and polyacrylate polymers and other supports. In the present work we used ionic liquids and various ligands as reagents for immobilization and the most perspective solid supports to prepare the novel solid-phase extractants designed for radionuclide preconcentration. The ability of ionic liquids to be kept on solid surfaces and to reveal ion-exchange and complexing properties makes possible the synthesis of novel type solid-phase extractants. We prepared the solid-phase extractants by the non-covalent immobilization of tetraalkyl phosphonium and 1-alkyl-3-methyl imidazolium ionic liquids with different anions and studied their sorption properties. The matrices with large surface and high sorption ability were chosen as supports: multi-walled carbon nanotubes, highly cross-linked polystyrene, Amberlite XAD-7 and polyacrylonitrile fiber. The conditions of ionic liquid immobilization and radionuclide recovery were determined. The application of prepared solid-phase extractants for preconcentration of actinides, rare earth elements and radiopalladium from 1-3M nitric acid solutions was demonstrated. Special attention was being paid to the use of carbon nanotubes as a support for solid-phase extractant preparation. Carbon nanotubes have come under intense multidisciplinary study due to their unique physical and chemical properties, which make them attractive for myriad uses including radiochemistry. The features of nanostructure and large surface ensure high sorption ability of carbon nanotubes and the possibility for their modification. We prepared the solid-phase extractants on the base of multi-walled carbon nanotubes "Taunit"® (Russia) modified with ionic liquids and various ligands. Experimental data have shown high efficiency of the novel solid-phase extractants for radionuclide preconcentration from nitric acid solutions.

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SEP.L07 (Id: 137)
SPECIATION AND THERMODYNAMICS OF LANTHANIDE AND ACTINIDE IONIC SOLUTIONS DESCRIBED USING THE MEAN SPHERICAL APPROXIMATION (MSA)

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A theoretical description of speciation and of the thermodynamic properties of lanthanide and actinide ionic solutions is proposed in the framework of the mean spherical approximation (MSA) [1-4]. In this model the ions are regarded as charged hard spheres immersed in a dielectric continuum representing the solvent (water). An interesting feature of the MSA is that it yields analytic expressions. To account for complex formation, the multivalent cation (a lanthanide or an actinide ion) is assumed to possess a finite number of sites on its surface, on which anions can bind [5]. This property leads to the formation of 1:1, 1:2, 1:3,... complexes according to a stepwise complexation-equilibrium process. Explicit formulas are obtained for the speciation and the thermodynamic properties (osmotic and activity coefficients) within this model. They include the effects of complexation and electrostatic interactions. The model is applied to the case of binary and ternary aqueous solutions of divalent, trivalent and tetravalent lanthanides and actinides.

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SEP.L08 (Id: 302)

BENTONITE STABILITY AND MIGRATION BEHAVIOR OF SOME CRITICAL RADIONUCLIDES

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The contribution is focused on geotechnical, chemical, geochemical and mineralogical research of bentonite stability with the aim to determine the effect of saturation medium composition and loading by heat on bentonite stability. The main part the research is directed to the experimental results of bentonite and bentonite leachate samples obtained for the bentonite interaction under laboratory or in situ experiments. The markedly different values of Na^+, K^+, Ca^{2+}, Mg^{2+} and SO_4^{2-} ion concentrations are caused by the mineralogical transformation and interaction of the original bentonite material with synthetic granitic water. Bentonite is included in buffer and backfill materials in most high-level waste repository concepts. Its low hydraulic conductivity, micro-porous structure, good sorption properties and plasticity make this material an effective barrier - protecting the canister and restricting the movement of radionuclides released from the waste packages after canister failure. The effect of the variable aqueous phase composition on bentonite stability using Ca-Mg and Na-Ca bentonites for the experiments was studied. The synthetic granitic waters with the higher concentration of the K^+ and Mg^{2+} cations were applied for the study of bentonite stability. These investigations were carried out for the 12 months under the laboratory and temperature increase conditions (95 - 100 C). The mineralogical transformation and characterization of the solid phase by RTG diffraction, infrared spectroscopy and cation exchange capacity measurement were evaluated after the termination of these experiments. These mixtures were simultaneously applied for the sorption study of the selected radionuclides (^{134}Cs, ^{125}I and $^{99}TcO_4^-$) with the aim to evaluate the changes of sorption behaviour as the result of bentonite with synthetic granitic water interaction. The geochemical software tool Geochemist's Workbench calculated the simple thermodynamic model of bentonite transformation during bentonite with synthetic waters interaction. It was confirmed that smectites are transformed to more stable silicates phases, such as illite/smectite mixed layers, illite. The kinetics of the smectite to illite reactions strongly depends on temperature, time and K^+ porewater concentrations. Radionuclides of ^{99}Tc and ^{129}I may exist in the various oxidation states in the dependence of the redox conditions of the aqueous environment. Technetium exists in oxidation states from +7 to -1. In natural environments, the most stable oxidation states of technetium are +7 and +4 under oxidizing and reducing conditions, respectively. The -1, +5, and molecular I_2 [I_2^0(aq)] oxidation states are those most relevant for iodine in environmental systems. Iodine usually exists in fresh water in the -1 oxidation state as iodide (I^-), in alkaline and marine environments, iodine usually exists in the +5 oxidation state as iodate (IO_3^-).

SEP.L09 (Id: 20)

SPECIATION OF CURIUM(III) AND EUROPIUM(III) IN HUMAN URINE SAMPLES

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Due to their radiation and heavy metal properties, radionuclides represent a serious health risk to humans in case of incorporation. To understand their toxicity, transport, deposition and elimination in the human organism, it is therefore crucial to elucidate their chemical behavior and properties on a molecular level. For trivalent actinides originating from nuclear power plants knowledge about their metabolism is very limited. In case of incorporation, they tend to accumulate in liver and skeleton and are excreted to maximum 10 - 20 % within the first week. Nevertheless this excretion occurs mainly through the kidneys, which are known to be particularly radiation sensitive. To address the lack of knowledge, we studied the speciation of curium (as a representative of trivalent actinides) and europium (as the lanthanide analogue) in human urine and their complexation with single constituents. Since both of these heavy metals exhibit unique fluorescence properties, time-resolved laser-induced fluorescence spectroscopy (TRLFS) is an adequate tool for this purpose. Fresh 24-hours-urine samples were collected from healthy volunteers and analyzed within few days. The inorganic composition of all samples was determined using mass spectrometry with inductive coupled plasma (ICP-MS) and ion chromatography (IC). Then all samples were spiked in vitro with curium or europium and single as well as time-resolved laser-induced fluorescence spectra were measured. We analyzed at least 10 different urine samples and were able to divide all samples into two different groups according to their fluorescence spectra. We found that all samples with pH below 5.6 and all samples with pH above 6.0 each exhibit strikingly similar spectra. Compared to each other the spectra of both groups are very different and therefore easy to distinguish.

Furthermore the lifetime of the metals in samples with higher pH is substantially longer than in samples with lower pH. ICP-MS and IC analysis revealed that the inorganic composition of samples which exhibit the same fluorescence spectra can vary broadly within magnitudes. Therefore the most important factor influencing the speciation of metals in human urine does not seem to be the composition but rather the pH value. To identify the dominating species we also recorded the fluorescence spectra of both metals in electrolyte solution containing all inorganic but no organic components of urine. Furthermore we studied the complexation of both metals with urea and citric acid as main organic urine constituents. Comparing the measured spectra and lifetimes with this reference data, we found that at lower pH a complex citric acid species dominates the speciation of both metals while at higher pH the spectra were identical to those in electrolyte solution ruling out any involvement of organic ligands. The exact nature of this inorganic metal species has yet to be clarified.

SEP.L10 (Id: 171)
COMPLEXATION OF CURIUM(III) WITH PYOVERDIN-MODEL COMPOUNDS

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The aerobic groundwater bacterium Pseudomonas fluorescens (CCUG 32456) isolated from the aquifers at the Äspö Hard Rock Laboratory, Sweden, secretes siderophores of the pyoverdin-type. Besides iron(III), these unique bioligands are also able to form strong complexes with actinides (e.g., U(VI), Np(V), and Cm(III)) [1-3]. For U(VI) and Np(V) we could show that mainly the catecholate and to less extent the hydroxamate functionalities of the pyoverdin molecule are involved in the actinide coordination [1,2]. For Cm(III) it is still not clear which functional group of the pyoverdin molecule causes the great stability constants. In general, Cm(III) interactions with pyoverdin-model compounds and especially with chromophore-models are poorly understood. To address this lack, we present findings regarding the speciation of Cm(III) with 2,3-dihydroxynaphthalene in aqueous solution by time-resolved laser-induced fluorescence spectroscopy (TRLFS) at trace Cm(III) concentrations (0.3 μM) over a wide pH range. TRLFS is a well established direct speciation technique for investigating the complexes formed by actinides in both geochemical and biochemical environments. Four Cm(III)- dihydroxynaphthalene species of the type MxLyHz could be identified from the spectroscopic measurements. The stability constants of these strong Cm(III)-bioligand complexes and their individual spectroscopic properties (excitation and emission spectra, lifetimes) are reported. TRLFS shows that Cm(III)-dihydroxynaphthalene species cause a strong red shift of the characteristic Cm(III) (aq) emission band at 593.8 to 614.4 nm. The findings of our Cm(III) speciation study in comparison with the literature indicate a stronger affinity of Cm(III) to the catechol functionality of the pyoverdin molecules as found for U(VI) and Np(V).

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SEP.L11 (Id: 204)
THE POURBAIX DIAGRAM OF ASTATINE IN AQUEOUS MEDIUM

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Astatine (At), element 85, is below iodine in the periodic table of elements. One of its isotopes, ^{211}At, is a promising candidate as a therapeutic agent in nuclear medicine [1]. Although it is clear that much of the chemistry described for halogens is applicable to astatine, a more metallic character is expected as compared to its nearest halogen neighbor, iodine. However, At chemistry in aqueous solution remains poorly understood. There are no stable isotope of astatine, its longest-lived isotope having a half-life of 8.3 h. All investigations are thus derived from radiochemical studies at ultra-trace concentration, typically between 10^{-12} and 10^{-15} mol/L and no spectroscopic tools can be used to identify unambiguously the formed species. The chemical forms of astatine are usually deduced from its behavior in given conditions with respect to the behavior of expected model compounds. In this work, a combined experimental and theoretical approach is used to define the potential/pH diagram of astatine (Pourbaix diagram) in non-complexing medium with the aim of answering the two main questions raised in the literature: does At(0) exist in aqueous solution and what is the chemical form of At(III), if it exists? The experimental methodology considers that a given species is characterized by its distribution coefficient (D) experimentally determined in a biphasic system. The change in speciation arising from a change in experimental conditions is observed by a change in D value [2]. Unlike most of previous studies, we present a quantitative analysis of the experimental data based on equilibrium reactions, to identify the species formed and derive the thermodynamic parameters.

The theoretical methodology is based on quasi-relativistic quantum chemistry computations and solvation free energy calculations using polarizable continuum models. The results show that At at the oxidation state 0 cannot exist in aqueous solution. The three oxidation states present in the range of water stability are At(-I), At(I) and At(III) and exist as At^-, At^+ and AtO^+, respectively, in the 1 to 2 pH range [3]. When the pH increases, AtO^+ reacts with water to form two hydrolysis species, AtO(OH) and $AtO(OH)_2^-$.

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SEP.L12 (Id: 300)

THE INFLUENCE OF THE SIDE GROUPS OF THE BTP AND BTBP TYPE LIGANDS ON LIQUID-LIQUID EXTRACTION OF ACTINIDES AND LANTHANIDES

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Fossil fuels are no longer a choice for energy production, both because of their diminishing availability and the by-products of their combustion processes. A possibility to replace the fossil fuels would be utilization of nuclear power. No green-house gases are produced, but as any industry, it generates wastes. A proper plan for reusing valuable nuclides, both by preparing another kind of fuel or by transmutation, which, besides transforming the long-lived nuclides into short-lived ones, generates energy, seem to be P&T (partitioning and transmutation). The partitioning is the process of separation nuclides from the rest of the waste and involves a separation process which utilize an organic solvent containing one or several molecules and a diluent for extraction purposes. Among the last extractant families developed in Europe, the 2,6-di(5,6-diethyl-1,2,4 triazin-3 yl)pyridine (BTP) and 6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo[1,2,4]triazin-3-yl)-[2,2']bipyridine (BTBP) were/are the most promising molecules for an eventual industrial application. The present work focuses on several aspects insufficiently studied before and concerning the influence of the side groups attached to the "core" of the molecule, for both BTPs and BTBPs. The importance of the diluents regarding both the extraction capabilities of the formed system and the role of the diluents in protecting the molecule against radiolysis without adding a scavenging molecule will be very briefly discussed. However this part will not be further developed here. The importance of the side groups added to the molecule first to facilitate dissolution, easing the phase transfer and thus enhancing the extraction capabilities and the role the side groups, in protecting the molecule against the radiolysis will be discussed.

Posters

SEP.P01 (Id: 306)
STUDY OF HDEHP-PAN SOLID EXTRACTANTS FOR ^{90}Sr DETERMINATION

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Application of solid extractants containing di-(2-ethylhexyl)phosphoric acid (HDEHP) in the support based on modified polycrylonitrile (PAN) has been proposed at the CTU in Prague, Department of Nuclear Chemistry, for the determination of ^{90}Sr by means of measuring the activity of its ^{90}Y daughter utilising a procedure similar to that developed by Burnett et al. in 1975 for the determination of ^{228}Ra [1]. For the introductory study, ^{152}Eu and ^{133}Ba were used as chemical homologues of ^{90}Y and ^{90}Sr. For these radionuclides, dependences of mass distribution coefficients (D_g) on the nitric acid concentration were measured for several types of HDEHP-PAN solid extractants; the results obtained were compared with the data presented by Horwitz et al. in his 1975 paper [2]. For one of the solid extractants, similar dependence was also determined for the hydrochloric acid. The mechanism of the Eu^{3+} and Ba^{2+} ions was confirmed to follow the theoretical two-phase equation for the chelating extractants. The shifts of the curves measured for various solid extractants could be ascribed to the different amounts of HDEHP in the extractants. Further, influence of the presence of nitrates (total salinity), calcium (Sr homologue) and iron (Y major interferent) ions on the D_g values were determined. For both Eu and Ba, a decrease of D_g values with increasing nitrates concentration was observed. As expected, the D_g(Eu) values were suppressed already by relatively low iron concentrations. The influence of iron ions could be masked by the addition of ascorbic acid if working in hydrochloric acid. The D_g(Eu) values were not influenced by the presence of calcium while, as expected, uptake of barium at low acidities was strongly suppressed already by relatively low calcium concentrations. The results obtained make application of solid extractants containing HDEHP in PAN support prospective for ^{90}Sr determination.

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SEP.P02 (Id: 312)
INVESTIGATION OF MOBILITY OF PLUTONIUM IN ENVIRONMENTAL AND NUCLEAR WASTE SAMPLES USING SEQUENTIAL EXTRACTION

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Fractionation of plutonium isotopes (^{238}Pu, ^{239,240}Pu) was conducted for environmental samples including soil and sediment, and bio-shielding concrete from decommissioning of nuclear reactor in this work. The fractionation were carried out by dynamically sequential extraction system using an on-line sequential injection (SI) combined with a specially designed extraction column. Plutonium in the fractions from the sequential extraction was separated by ion exchange chromatography and measured using α spectrometry. Different distributions of plutonium in environmental samples and bio-shielding concrete were observed. The analytical results show a higher mobility of plutonium in concrete sample than in environmental samples analyzed in this work, which means attention should be paid to the treatment and disposal of low level concrete waste from decommissioning of nuclear facilities.

SEP.P03 (Id: 149)
SOLVENT EXTRACTION OF AMERICIUM BY IMIDOPHOSPHATES

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Solvent extraction was described for the analytical determinition of americium in the liquid samples. Arylesters of imidodiphosphoric, imidothiodiphosphoric, imidodithiodiphosphoric acids and tetraphenylimidodithiodiphosphine were used as representatives of bidentate organophosphoric chelating agents. Pentaphenyldiimidotriphosphate, from the group of tridentate agents, was used. The extraction properties of tetraphenyl imidodiphosphates and their sulphur analogues for trivalent americium in 0.1-1 mol/L HNO_3 into toluene and also in linear combination with TOPO were investigated. The dependences of equilibrium ratios of americium on analytical or equilibrium concentration of chelating agents, pH, initial concentration of nitric acid and initial concentration of TOPO were studied. The structures of the complexes in the organic phase were determined and the value of extraction constants were calculated for all systems. The optimal extracted species were AmA_3, $AmA_3(HA)$, the addition of TOPO induced synergistic extraction of AmA_3.TOPO. The utilization of sulphur analogues was insignificant.

SEP.P04 (Id: 308)

THE LIQUID-LIQUID EXTRACTION OF MINOR ACTINIDES WITH CyMe$_4$-BTBP IN SELECTED DILUENTS

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The $CyMe_4$-BTBP (6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-1,2,4-benzotriazine-3-yl)-2,2'-bipyridine) has been demonstrated to be a prospective extractant for the liquid-liquid extraction of minor actinides. For industrial applications, its solubility in 1-octanol is low and the kinetics of extraction in this diluent is slow. Better performance has been demonstrated in cyclohexanone, however, the partial miscibility of this solvent with water makes its industrial use problematic. Therefore, the main purpose of this study was to find the most suitable diluent for the $CyMe_4$-BTBP. The separation of americium(III) from europium(III) was studied. This is in particular reference to the separation of minor actinides from the high-level liquid waste issuing from the reprocessing of spent nuclear fuel. The results obtained using the diluents tested were compared with the data for cyclohexanone or 1-octanol as reference diluents. The principal diluents tested were alkylcyclohexanones. The parameters studied included the dependence of distribution ratios of americium(III) on the position of the alkyl group on the cyclohexanone ring, on the length of the alkyl substituent and on the number of alkyl groups in the ring. Extraction by the neat diluents (without $CyMe_4$-BTBP extractant) was also studied as well as the dependence of the distribution ratio on the concentration of nitric acid. It has been shown that the alkylated cyclohexanones are promising diluents for the partitioning of minor actinides from lanthanides by $CyMe_4$-BTBP. In particular, 3-methylcyclohexanone was found to have the optimum properties compared with the other cyclohexanones.

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A TRLFS STUDY OF EUROPIUM SPECIATION WITH GLYCOLIC ACID

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Europium is often used as a model of the fission products from the lanthanides and minor actinides group. Glycolic acid is used in modern decontamination solutions and has been also proposed as a complexing agent in the reprocessing of spent nuclear fuel. The aim of this study was to carry out a more detailed study of europium complexation with glycolic acid using Time-Resolved Laser-Induced Fluorescence Spectroscopy (TRLFS), including determination of the stability constants of europium complexes with glycolic acid, and to compare the results with published data [1]. Furthermore, this work should also clarify the impact of sample preparation in air and under N_2 atmosphere, i.e. the role of hydroxo- and carbonato- complexes at pH > 7. Because Eu complexes with glycolic acid show good fluorescent properties, the TRLFS method was chosen for their study. The studied parameters included the influence of the concentration of CO_3^{2-} on the formation of europium carbonato- complexes, effect of pH on the formation of hydroxo- complexes of europium (solutions prepared under N_2 atmosphere), and europium complexation in solutions with glycolic acid (comparison of the samples prepared on air and under N_2 atmosphere). The fluorescence spectra of individual samples and the time of fluorescence of prepared complexes were recorded. The results obtained were compared with those published by Stumpf et al. [1]. Based on the comparison of the results obtained for samples prepared on air and under N_2 atmosphere, it can be concluded that the transfer of atmospheric CO_2 is negligible even at relatively high pH value and that the influence of carbonate complexes in europium-glycolic acid systems is negligible. The TRLFS method was further supplemented with the extraction method, which can be also used for determining the stability constants of complexes of europium with glycolic acid. In the following step, stability constants of prepared complexes will be obtained from measured data using factor analysis.

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SEP.P06 (Id: 206)

COMPLEXATION OF At^+ AND AtO^+ WITH INORGANIC LIGANDS; A COMBINED EXPERIMENTAL AND THEORETICAL APPROACH TO CHARACTERIZE THE FORMED SPECIES

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Astatine (At, Z = 85: [Xe]4f145d106s26p5) is below iodine and belongs to the halogen group. One of its isotopes, ^{211}At, is a promising candidate as a therapeutic agent in nuclear medicine [1]. It has been reported that astatine presents a metal-like behavior when existing under the oxidation states +I and +III as At^+ and AtO^+ species [2]. However, the number of studies dealing with the complexation properties of these cationic forms is limited. Low availability of astatine makes experimental work difficult (usually investigations derived from radiochemical studies at ultra-trace concentrations, typically between 10^{-12} and 10^{-15} mol L^{-1}). In this work, we propose a combined experimental and theoretical approach to characterize the interaction between these metallic forms and simple inorganic anions (Cl^-, Br^-, SCN^-). A competition method based on solid/liquid separation or liquid/liquid extraction is proposed to determine the equilibrium constants [3]. To assess the reliability of the experimental results, they are compared to theoretical computations. The spin-orbit density functional theory (SO-DFT) approach, which uses two-components relativistic effective core potentials (RECPs), has been used to investigate the spectroscopic properties of At, At_2 and HAt species. In conjunction with our own built up basis sets (double and triple ζ augmented with diffuse and polarization functions), SO-B3LYP and SO-M06 functionals offer results in good agreement with that in the literature. In order to compare measured and computed equilibrium constants of the astatine reactions, aqueous solvation effects have been introduced using polarizable continuum models (PCM with UAHF and UAKS cavities). Obtained results show a good agreement between theoretical and experimental values. They also indicate that astatine species have a soft character according to the HSAB theory.

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SEP.P07 (Id: 57)

THE STUDY OF THE SPECIATION OF URANYL-SULPHATE COMPLEXES BY UV-VIS ABSORPTION SPECTRA DECOMPOSITION

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Uranyl-sulphate complexes are the most significant U(VI) species in acid solutions containing sulphate ions. The study of their properties is of practical significance for characterisation of solutions used in underground uranium ore leaching and following remediation of leaching sites. At low pH values, the speciation of U(VI) mainly depends on its total concentration $[U^{VI}]$ and sulphate ions concentration $[SO_4^{2-}]$. As absorption UV-VIS spectra of $UO_2(NO_3)_2$ + Na_2SO_4 solutions were found significantly dependent upon the ratio $\Phi = [SO_4^{2-}]/[U^{VI}]$, the spectrophotometric method for closer study of U(VI) - SO_4^{2-} complexes speciation was used. The solutions under study were of constant uranium concentration $[U^{VI}] = 5x10^{-2}$mol L^{-1}. Required values of the Φ ratio were adjusted by proper additions of Na_2SO_4 solution and varied from $\Phi = 0$ up to $\Phi = 20$. Constant pH value pH = 2 was maintained by the additions of either HNO_3 or NaOH solution of negligible volume. Absorption spectra were recorded using UV/VIS spectrophotometer CARRY 100 within the wave-length range 190 - 900 nm. The following U(VI) species were considered under the given conditions: UO_2^{2+}, UO_2SO_4 (aq), $UO_2(SO_4)_2^{2-}$ and $UO_2(SO_4)_3^{4-}$. As the most pronounced effects of the Φ value changes on the spectra appeared at wave-lengths between 380 and 500 nm, these parts of spectra were used as a database for the identification of individual U(VI) species and for determination of their individual spectra by deconvolution of the experimental spectrum. Proposed simple deconvolution method requires a preliminary estimation of relative amounts of individual species. Stability constants from NEA database and SIT method of activity coefficients calculation at high ionic strength were used for this purpose. The spectrum of $UO_2(NO_3)_2$ solution without Na_2SO_4 ($\Phi = 0$, pH = 2) was considered to be the individual spectrum of pure UO_2^{2+} species. Absorbance data following from it and calculated relative amounts of all four species for each Φ value were taken as the basic set of input data for deconvolution. The absorbance of each of the three remaining species (UO_2SO_4 (aq), $UO_2(SO_4)_2^{2-}$, $UO_2(SO_4)_3^{4-}$) at given wave length can be find by solving the system of three linear equations involving experimental data from three spectra of different Φ ratio. The disagreement of spectra calculated from different triplets of experimental spectra can be caused either by experimental errors or by incorrect estimation of relative amounts of individual species. The presented method can be helpful even for estimation of accuracy of stability constants and SIT activity coefficients used in speciation calculations.

SEP.P08 (Id: 26)
THE EFFECT OF INDUCED α-RADIOLYSIS ON THE U(IV)/U(VI) RATIO IN URANIUM ORES

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The variation of the concentration of U(IV) and U(VI) species from uranium ores due to the α-radiolysis induced by an ^{241}Am source in several deposit conditions is studied. The X-rays patterns show some differences for the studied ores before and after external α-radiolysis. The concentrations of U(IV) and U(VI) species was determined spectrophotometrically.

SEP.P09 (Id: 275)
USE OF HYDROLYSIS FOR SEPARATION AND REMOVAL OF RADIONUCLIDES FROM SOLUTION

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Behaviour of radionuclides in such processes as sorption, ion-exchange, ultrafiltration, eTc., that are used for removal of radionuclides from solution, is largely determined by their speciation in solution. The hydrolysis of metal ions in solution is particularly interesting in that respect, since most of the liquid radioactive wastes are aqueous solutions. The following forms of metal ions (Me^{z+}) occur in aqueous solution as the pH increases from acidic to basic conditions: - hydrated cations ($Me(H_2O)^{z+}$) - mononuclear hydroxocomplexes ($Me(OH)_q^{(z-q)+}$) - polynuclear hydroxocomplexes ($Me_p(OH)_q^{(pz-q)+}$) - pseudocolloids. Each of the above forms possesses specific physicochemical properties that can be effectively used for separation of radionuclides from solution. It has been shown that using the differences in ability of various metals to form hydroxocomplexes and the differences in behaviour of the above species in sorption and ultrafiltration processes it is possible to develop highly selective methods for extraction of radionuclides from solution, including anionic species.

SEP.P010 (Id: 51)
PHOTOCATALYTIC DEGRADATION/SORPTION OF RADIOCOBALT FROM EDTA-Co COMPLEXES USING CRYPTOMELANE-TYPE MnO_2

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Manganese dioxide was synthesised from hydrometallurgical wastewater simulant and the material was identified as cryptomelane-type MnO_2 (potassium form of α-MnO_2). The material showed good trace level heavy metal uptake properties, uptake preference Co>Cd>Ni, from 0.1M $NaNO_3$ background solution. The material showed also very fast copper uptake kinetics but its powdery physical form prevented its use in typical column experiment. However, using the material in baTch mode in application, such as fluidized bed-type UV-photoreactor was found to be ideal for the material. The synthesised cryptomelane-type MnO_2 was tested in UV-photoreactor for its Co-EDTA degradation properties (10 μM Co and 10 μM EDTA concentrations in 0.1 M $NaNO_3$ background). In these experiments cobalt uptake of 99% was observed after 1 hour of UV photon irradiation. As for comparison, a well known TiO_2 was tested as reference material that showed about 90% cobalt uptake after 6 hours of irradiation under identical experiment conditions. It was also noted that the cobalt uptake on cryptomelane without UV irradiation was very modest, only about 10%. It was concluded that synthesis of cryptomelane-type MnO_2 was successful from rather complex precursor solution (hydrometallurgical wastewater simulant) and that the material has interesting ion exchange/catalytic properties but the powdery physical form of the material prohibits its use in traditional column applications.

SEP.P11 (Id: 58)
SEQUENTIAL SEPARATION OF ^{90}Sr FROM α EMITTERS WITH MIXED SOLVENT ANION EXCHANGE AND THEIR DETERMINATION BY LSC AND A SPECTROMETRY

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Strontium, plutonium and americium are isotopes the determination of which is most often encountered in practice (from all α and pure β emitters) because of their radiotoxicity, long period of half life and potential hazardous effects on human life. It is well known that their determination, because of their radiochemical properties, requires isolation from sample and separation from other elements (active or inactive) before detection. In past many methods were developed for their separation from different kind of samples. Main characteristics of almost all this methods is that separation and determination of strontium isotopes are separated from α emitters. Therefore the main aim of this paper is the development of a method for strontium and α emitters separation in one step by mixed solvent anion exchange. It will be shown that Sr, Y and the mentioned α emitters can be isolated from complex samples and separated from great amounts of Ca and many other elements and, subsequently, separated mutually on the chromatographic column filled with strong base anion exchangers (Amberlite CG-400 or Dowex 1x8) in nitrate form combined with alcoholic solutions of nitric acid as eluent. It will be also shown how combination of the used alcohols can improve separation efficiency between mentioned isotopes. Separation of Sr and Y from

Am and Pu can be achieved on the column filled with the mentioned exchanger and alcohol (ethanol, methanol)-water mixture as eluent. Pu and Am can be easily stripped from column by water. In addition, it will be shown how this type of isolation can accelerate ^{89,90}Sr determination on LSC or proportional counter and determination of Pu and Am by α spectrometry.

SEP.P12 (Id: 59)

ANALYSIS OF ^{55}Fe BY COMBINATION OF CHROMATOGRAPHIC SEPARATION AND LIQUID SCINTILLATION DETECTION

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^{55}Fe is an activation product, low energetic β emitter with approximately 2.74 year half life. Therefore, its presence in natural systems is limited in time and mainly dependent on nuclear reactor operation. As a rule determination of ^{55}Fe requires chemical separation prior detection. For this purposes separation on anion exchange column in two steps is usually used which makes it time-consuming. In addition, in the last decade improvements of methodology were described in few publications but mainly for its determination in liquid radioactive waste samples. Therefore, the main aim of this paper is to show the way for iron isolation from liquid samples with a high concentration of ions that enable simple determination of ^{55}Fe. Iron binding on extraction chromatographic resins developed by Horwitz and co-workers, so called TRU and Ln resin, will be discussed. The method for concentration and separation from all interfering isotopes on extraction chromatographic column which enables rapid and simple determination will be proposed. It will be shown how Fe can be easily separated from great amounts of macro elements, α and β emitters on column filled with TRU or Ln resin by using HCl and/or HNO_3 in one step. The advantages and disadvantages of using of Ln and TRU resins will be discussed. After the isolation, activity of ^{55}Fe is determined by counting on the liquid scintillation counter by using quench correction curve. It will be described which type of quenching mainly affects the accuracy of determination. Type of the polynomial approximation for efficiency determination (and quench correction) in sense of minimization of determination error will be proposed. Budget of uncertainty of determination will be discussed.

SEP.P13 (Id: 148)

ELECTRODEPOSITION OF SELECTED α-EMITTING RADIONUCLIDES FROM OXALATE-AMMONIUM SULFATE ELECTROLYTE AND MEASURED BY MEANS OF SOLID-STATE ALPHA SPECTROMETRY

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This method describes electrodeposition of selected group of α-emitting radionuclides such as method suitable for measuring by means of solid-state α spectrometry. The effect of volume and pH of electrolyte, distance of electrodes, current and plating time versus different α-emitting radionuclides was observed in order to optimize conditions to obtain maximum yield.

SEP.P14 (Id: 184)

SORBENTS OF HEAVY METALS BASED ON COMBINATION OF LOW RANK COALS AND CHITOSAN

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The study has been aimed at development of cheap sorbents of toxic heavy metals and radionuclides based on low rank, low energetic coals in combination with chitosan for use in treatment of both waste and drinking water and construction of active geochemical barriers. Sorbents based on lignite and naturally oxidized lignite (oxihumolite, leonardite) were prepared, both raw and impregnated with chitosan from its solution in oxalic acid. The sorbents were characterized by chemical and structural analyses. Their sorption performance for Co, Cu, and Cd was tested in equilibrium sorption experiments with respect to pH. The sorption efficiencies were calculated from the amount of metal retained by the sorbent, which was determined by instrumental neutron activation analysis in the short time irradiation mode with epithermal neutrons (in Cd shielding). Although chitosan provides only surface coating of the coal sorbents, its presence has affected significantly their sorption performance. At identical pH values, the sorption efficiencies of the chitosan coated coals were higher than those of the raw coals for all three metals, and this difference increased with pH. The chitosan coating obviously stabilizes coal in basic solution and inhibits leaching of humic substances and their complexes with the metals from coal into solution. Besides fixing metal humates within the sorbents, chitosan may positively affect the sorption efficiency also by its buffering action against the hydroxide addition.

SEP.P15 (Id: 289)
DETERMINATION OF ^{59}Ni IN RADIOACTIVE WASTES

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The aim of this study was suggestion and examination of method for radioanalytical determination of ^{59}Ni in radioactive wastes using low energy photon spectrometry (LEPS). New composite material PAN-DMG, containing chelating agent dimethylglyoxime (DMG) immobilized in porous matrix of binding polymer polyacrylonitrile, was used for nickel separation and concentration. Method for preparation of ^{59}Ni sample for LEPS was developed using homogeneous precipitation of nickel with DMG. Proposed radioanalytical method was tested with two types of real radioactive wastes (boric acid concentrate from NPP evaporator and primary circuit coolant ionex resin).

SEP.P16 (Id: 309)
PREPARATION OF SAMPLES FOR α-SPECTROMETRY BY DIRECT EVAPORATION OF EXTRACTED SPECIES

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Because of the energy loss of α particles by self-absorption, α spectrometry requires thin, uniform, and nearly weightless samples. Several methods exist for sample preparation e.g., electrodeposition, or co-precipitation. Unfortunately, the methods yielding the best energy resolution are not always quantitative and are usually relatively demanding and time-consuming. This fact makes application of α-spectrometry for screening tests with radiotracers complicated. For its simplicity and fastness, the possibility to prepare samples for α spectrometry by direct evaporation was investigated in order to evaluate the efficiency of Am/Cm separation. The simple aliqouts of aqueous or organic phase, aliquots in the presence of an excess of volatile organic solvent (acetone) or mixed with tensioactive (tetraethylene glycol) were deposited on stainless steel planchets, evaporated under infrared lamp and heated in flame until glowed with dull red colour. The influence of sample preparation technique on counting efficiency and energy resolution has been investigated. The results show there is not one versatile technique, but the preparation should consider the particular composition of samples to be measured.

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SEP.P17 (Id: 254)
SOLUBILITY AND SORPTION BEHAVIOR OF MONAZITE CHEMICAL COMPONENTS IN HUMIC ACID SOLUTION

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Using the method of solubility it is shown that long (15 days) exposure of monazite powder in the aqueous solutions of natural or synthetic humic acids (HA) give rise to 10 to 100 times higher solubility of heavy p-, d- and f-elements of Mendeleev periodic system in comparison with solutions without humic acids. The level and rate of the solubility is shown to depend on the initial concentration of HA in solution and pH. It has been concluded that the resistance of inorganic phosphates toward HA should be taken into consideration in the construction of phosphate matrixes for immobilization of poisonous and radioactive chemical elements. Another aspect of monazite solubility in HA natural water solutions concerns the transfer of the solubility products, the humic complexes of p-, d- and f-elements through heterogeneous porous media with ion-exchange properties. This problem is extremely complicated and manifold [1,2]. As the first approximation the transfer (or sorption, S) of the microelements (ME) in the “gross-system”, which assumed to include the water soluble and colloid HA, HA- and hydroxo-complexes of ME, the ion-exchanger, depends on the heterogeneous interaction in the sub-systems “ME-ion-exchanger”, “ME-HA”, “HA-ion-exchanger”. Within the frame of this concept [2] the presentation summarizes experimental data on sorption behavior of La(III), Th(IV), U(VI), Sr(II) and other microelements, the soluble chemical components of monazite in HA water solutions, toward strong cation-exchanger in Na-form. Example of such a data shows that sorption of a ME is strongly dependent on the HA solubility as a function of pH. Another coexisting factor is the “ME - HA(colloid)” - “ME- ion-exchanger” competitive sorption at critical micelle concentration point (pH<4). Physicochemical aspects of the gross-system evolution in the field of such parameters as pH, HA content, monazite solubility are discussed in the presentation.

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SEP.P18 (Id: 218)

UPTAKE OF ARSENIC BY MANGANESE DIOXIDE FROM WATER

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Arsenic contamination of drinking water has been reported from many parts of the world. The maximum permissible level of total arsenic in drinking water is 10 µg L^{-1} as recommended by WHO. Natural geochemical contamination through soil leaching is the primary contributor of dissolved arsenic in ground water around the world. The most common species present in water are the inorganic species: As(V), predominates in well oxygenated waters, and As(III), predominates in ground water. The reduced state, As(III), is more toxic, more soluble, and more labile than the oxidized As(V). Methods for removal of arsenic from water have been reviewed [1]. Arsenic remediation from water using manganese dioxide has been reported in the literature [2]. In the present work, uptake of As(III) and As(V) by manganese dioxide was studied. Manganese dioxide was chemically synthesized in laboratory by various methods. Uptake was studied by baTch equilibration method using ^{76}As as radioactive tracer. The influence of various parameters such as method of preparation and storage and aging, pH, initial concentration of arsenic and contact time has been studied. Uptake of arsenic is dependent on method of preparation of MnO_2. Also, uptake efficiency for As(III) and As(V) is different.

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10. EDUCATION / COORDINATION (EDU)

Lectures

EDU.L01 (Id: 303)
A COMBINED NUCLEAR TECHNOLOGY AND NUCLEAR CHEMISTRY MASTER - A UNIQUE INITIATIVE AT CHALMERS UNIVERSITY OF TECHNOLOGY, SWEDEN

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The need for engineers and scientists who can ensure safe and secure use of nuclear energy is large in Sweden and internationally. Chalmers University of Technology has therefore launched a new 2-year master´s program in Nuclear Engineering, with start from the autumn of 2009. Since the program is open to both Swedish and foreign students and the teaching language is English, the first year enrolment showed an unexpected interest. The program starts with compulsory courses dealing with the basics of nuclear chemistry and physics, radiation protection, nuclear power and reactors, nuclear fuel supply, nuclear waste management and nuclear safety and security. There are also compulsory courses in nuclear industry applications and sustainable energy futures. The subsequent elective courses can be chosen freely but there is also a possibility to choose informal tracks that concentrate on nuclear chemistry or reactor technology and physics. The nuclear chemistry track comprises courses in e.g. chemistry of lanthanides, actinides and transactinides, solvent extraction, radioecology and radioanalytical chemistry and radiopharmaceuticals. The program is finished with a one semester thesis project. The projects can be performed both in the University's headquarters (Nuclear Chemistry Department or Nuclear Technology) on own research themes or in cooperation with different nuclear facilities on themes of interest. The advanced combined courses in both nuclear chemistry and nuclear technology make this master program quite unique on international level so far.

EDU.L02 (Id: 363)
EDUCATION IN NUCLEAR CHEMISTRY AT THE CZECH TECHNICAL UNIVERSITY IN PRAGUE

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Department of Nuclear Chemistry (DNRC) exists at the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague since the establishment of this faculty in 1955. From the beginning, many recognized scientists were involved in the department activities, among others František Běhounek (student of Marie Curie), Jiří Starý and Petr Beneš. Since 2003, part of the research activities has been carried on in the newly established Centre for Radiochemistry and Radiation Chemistry (CRRC). The Department, together with the Centre, provide education in nuclear chemistry at three different levels - each commonly taking three years of study; we put emphasis on versatility and adaptability of graduates of all levels. Education of students at bachelor level is firmly rooted in physical and nuclear chemistry, with good basics in analytical, organic, inorganic and biochemistry, including practical exercises. Candidates of bachelor degree must pass state exams from general chemistry and nuclear chemistry. They must also successfully defend bachelor theses. Master level courses encompass all disciplines of nuclear and radiochemistry, including separation chemistry, trace radiochemistry, radiation chemistry, chemistry of radioactive elements, radioanalytical methods, chemistry of nuclear fuel cycle, application of radionuclides, radiation methods in biology and medicine eTc. Students may choose from three specializations - applied nuclear chemistry, environmental nuclear chemistry and nuclear chemistry in biology and medicine, practical abilities are trained in extended laboratory exercises from nuclear chemistry. Candidates defend diploma theses and pass state exams from nuclear chemistry, physical chemistry and one elective subject. The Ph.D. level course is oriented on independent research of the candidates. Moreover, all of them must also attend highly specialized lectures relevant to their specific field of study; the main fields of research are oriented on the chemistry of nuclear fuel cycle, radiation methods and radionuclide behaviour in the environment. Students have to pass Ph.D. state exam and defend their theses.

EDU.L03 (Id: 364)
CINCH - COOPERATION IN EDUCATION IN NUCLEAR CHEMISTRY

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The renaissance of nuclear power is already requiring a significant increase in the number of the respective specialists, amongst others are nuclear chemists. Because the current situation in nuclear chemistry education and training in Europe is quite diverse, a project for cooperation in education in nuclear chemistry (CINCH) seemed to be needed. The project aims to coordinate the education in Nuclear Chemistry, both at Ph.D. and undergraduate levels, within the EU, in collaboration with Russia. The system developed should enable formation of a long-term Euratom Fission Training Scheme (EFTS) providing a common basis to the fragmented activities in this field and thus move the education and training in nuclear chemistry to a qualitatively new level. The main cornerstones of the project are: coordination of the education, training and e-learning/dissemination in Nuclear Chemistry. A careful supervision and evaluation will be carried-out by an Advisory Board of end-users, academia and NGO (such as e.g. ENEN association). Previous experience gained by ENEN association during the coordination of nuclear engineering education together with data collected by the Division of Nuclear and Radiochemistry of EuChemMS (DNRC) and IAEA will strengthen the base of CINCH. Also, the project will serve as support and supplement to the training modules of EUROATOM “chemical” IPs and NOEs, namely that of ACTINET, ACSEPT, eTc; thus providing synergy rather than competition in the field.

LIST OF CONTRIBUTIONS

Topics abbreviations:

Plenary Session – **PLN**
Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste Management - **NFC**
Nuclear Analytical Methods – **NAM**
Chemistry of Actinide and Trans–actinide Elements – **TAN**
Radionuclides in the Environment, Radioecology – **REG**
Production and Application of Radionuclides – **PAR**
Radiation Chemistry – **RCH**
Nuclear Methods in Medicine, Radiopharmaceuticals and Diagnostics, Labelled Compounds – **RFD**
Separation Methods, Speciation – **SEP**
Education / Coordination – **EDU**

Plenary Session – PLN s91

PLN.L01	(Id: 122)	*X. Hou*	Radiochemical analysis for the determination of radionuclides difficult to measure for characteristics and decommissioning of nuclear facilities
PLN.L02	(Id: 370)	*B. Salbu*	Speciation analysis of radionuclides in the environment
PLN.L03	(Id: 365)	*W. Kutschera*	Accelerator Mass Spectrometry - from archaeology to astrophysics and human DNA
PLN.L04	(Id: 288)	*J. V. Kratz*	Chemistry of the transactinides
PLN.L05	(Id: 366)	*C. Nourry, P. Souček, R. Malmbeck, J. P. Glatz*	Electrochemical separation of actinides from molten LiCl-KCl on solid Al cathodes
PLN.L06	(Id: 297)	*K. Katti*	Radioactive and gastrin releasing peptide receptor specific gold nanoparticles in molecular imaging and therapy of cancer

Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste Management - NFC s94

Lectures s94

NFC.L01	(Id: 92)	*M. Precek, A. Paulenova*	Kinetics of reduction of hexavalent neptunium by nitrous acid in solutions of nitric acid
NFC.L02	(Id: 164)	*K. R. Kim, S. Y. Choi, Y. J. Jung, D. H. Ahn, S. Paek, S. W. Kwon, J. B. Shim, S. H. Kim, H. S. Lee, B. G. Park, K. W. Yi, I. S. Hwang*	Electro-fluid analysis of a molten-salt electrorefiner for pyrochemical nuclear waste treatment
NFC.L03	(Id: 295)	*P. Soucek, R. Malmbeck, E. Mendes, C. Nourry, J. P. Glatz*	Electrochemical separation of actinides from molten LiCl-KCl on solid Al cathodes
NFC.L04	(Id: 279)	*S. Koyama, K. Tatenuma, K. Okada, M. Ozawa*	Advanced ORIENT Cycle - progress on research, with focusing on safety and engineering
NFC.L05	(Id: 191)	*M. Lučaníková, P. Selucký, B. Grüner, M. Kvíčalová*	Cobalt bis(dicarbollide) ions with covalently bonded TODGA-like substituents for actinide and lanthanide extractions
NFC.L06	(Id: 35)	*A. Shadrin, D. Shafikov, A. Murzin, V. Kamachev*	Extraction of radionuclides with solutions of complexing agents in supercritical fluids and compressed gases
NFC.L07	(Id: 81)	*R. Harjula, A. Paajanen, R. Koivula*	Removal of antimony-124 from PWR coolant water
NFC.L08	(Id: 69)	*A. Shimada, T. Haraga, A. Hoshi, Y. Kameo, M. Nakashima, K. Takahashi*	Analysis of Th, U, Pu, and Am in radioactive metal waste using extraction chromatography
NFC.L09	(Id: 144)	*E. Wieland, N. Macé, R. Daehn, X. Gaona, D. Popov, J. Tits*	Micro-scale investigations of U(VI) immobilization by cementitious materials

NFC.L10	(Id: 80)	*A. Orlova, T. Salomadina, E. Loginova, D. Mikhailov, V. Kurazshkovskaja*	New f-element phosphates with langbeinite-type structure
NFC.L11	(Id: 23)	*S. Osvath, N. Vajda, Z. Molnar, E. Szeles*	Determination of ^{237}Np, ^{93}Zr and other long-lived radionuclides in medium- and low-level radioactive waste samples
NFC.L12	(Id: 60)	*F. S. Ebong, E. N.D.M*	Sorption of Ni and Eu in a multi-element system
NFC.L13	(Id: 63)	*R. Hallam, N. D. M. Evans*	Sorption of Tc(IV) to geologic materials associated with a geological disposal facility
NFC.L14	(Id: 71)	*C. Joseph, K. Schmeide, S. Sachs, G. Bernhard*	Effect of Ca^{2+} on the sorption of uranium(VI) and humic acid on natural clay
NFC.L15	(Id: 103)	*V. E. Badillo-almaraz, J. M. Rodriguez Soriano, C. Alliot*	Sorption of Pd, one long lived fission product, onto synthetic hydroxyapatite
NFC.L16	(Id: 100)	*S. Jain, N. Evans, M. Charalambous*	The adsorptive behaviour of cadmium on clays
NFC.L17	(Id: 221)	*T. Reich, S. Amayri, S. Wendt, B. Baeyens, R. Dähn, M. H. Bradbury, A. C. Scheinost*	Speciation and surface complexation modelling of Np(V) sorption on montmorillonite
NFC.L18	(Id: 41)	*A. Vokál, D. Vopálka*	An approach for acquiring data for description of diffusion in performance assessment of radioactive waste repositories
NFC.L19	(Id: 54)	*N. Evans, R. Hallam, P. Warwick*	The complexation of Tc(IV) with organic ligands in radioactive waste
NFC.L20	(Id: 95)	*E. Andrade, M. J. Madruga, I. Bobos, I. Paiva, F. Maia, A. Mateus, R. Trindade, M. C. Freitas, M. A. Gonçalves*	Characterization of Portuguese geomaterials, the clay component of rañas, as potential liners for low and intermediate radioactive disposal sites
NFC.L21	(Id: 114)	*V. Havlová, J. Landa, A. Martin, J. Eikenberg*	Long term diffusion experiment (LTD) in Grimsel URL: Comparison of modeling and in-situ results
NFC.L22	(Id: 124)	*N. Evans, P. Warwick, S. Anton-Gason*	The effect of competition from other metals on nickel complexation by α-isosaccharinic, gluconic and picolinic acids
NFC.L23	(Id: 249)	*V. Havlová, P. Večerník, M. Lofgren*	Migration characteristics of rock samples studied by electromigration method: methodology: procedure modification
NFC.L24	(Id: 61)	*Á. Bihari, Z. Szűcs, M. Mogyorósi, T. Pintér*	Joint determination of ^{99}Tc and ^{108m}Ag in L/ILW liquid wastes
NFC.L25	(Id: 113)	*V. Havlová, D. Vopálka*	Tritium (HTO) as a conservative tracer used for characterization of contaminant migration in porous rock environment
NFC.L26	(Id: 234)	*D. Eriksen*	Extraction of thorium from the Fen deposit in Norway
NFC.L27	(Id: 262)	*F. Poineau, P. Forster, P. Weck, K. Czerwinski, G. Jarvinen, E. Mausolf, E. Johnstone, A. Sattelberger*	Technetium chemistry at the University of Nevada Las Vegas
NFC.L28	(Id: 299)	*Z. Varga, M. Wallenius, K. Mayer*	Recent developments of nuclear forensic signatures of yellow cakes

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NFC.P01	(Id: 22)	*P. N. Pathak, D. R. Prabhu, A. S. Kanekar, V. K. Manchanda*	Evaluation of dihexyloctanamide as extractant under plutonium rich feed conditions
NFC.P02	(Id: 283)	*A. Szabó Nagy, K. Varga, B. Baja, Z. Németh, D. Oravetz, Z. Homonnay, E. Kuzmann, J. Schunk, G. Patek*	Comprehensive investigation of the corrosion state and surface properties of the stainless steel tubes of steam generators
NFC.P03	(Id: 142)	*D. E. Mohamed Mohamed*	Pyrochemical and electrochemical separations studies on plutonium (Part 2)
NFC.P04	(Id: 145)	*P. Selucký, M. Lučaníková, J. Rais, B. Grüner, M. Kvíčalová*	Extraction of Ln/An from highly acidic solutions using cobalt bis(dicarbollide) functionalized with complexing CMPO group.
NFC.P05	(Id: 167)	*K. V. Mareš*	Study of properties of extraction-chromatographic material TBP-PAN
NFC.P06	(Id: 222)	*M. Straka, M. Korenko*	Electrochemical behaviour of selected actinides and lanthanides in molten fluoride salts FLiNaK and FLiBe

NFC.P07	(Id: 291)	*M. Mariani, M. Mazzuccato, A. Buttafava, D. Dondi*	EPR measurements on N-bearing ligands used in spent nuclear fuel reprocessing for An(III)/Ln(III) partitioning
NFC.P08	(Id: 342)	*G. Lujaniene, J. Šapolaitė, V. Strelko, O. Oleksienko, T. Sčiglo, S. Meleshevych, V. Kanibolotskyy*	Removal of Cs, Sr, Pu and Am from contaminated solutions by inorganic sorbents
NFC.P09	(Id: 34)	*O. Shmidt, E. Pavlova, Y. Korneyko, A. Murzin, A. Shadrin, N. Sapozhnikova*	Improvement of radionuclide fixation in cement matrixes during immobilization of liquid radioactive waste
NFC.P10	(Id: 85)	*A. Young, M. Felipe-Sotelo, P. Warwick*	The solubility of Ni(II) and Eu(III) in the presence of cement superplasticisers
NFC.P11	(Id: 134)	*V. Pet'kov, I. Korchemkin, E. Asabina, V. Kurazhkovskaya, A. Zaripov, S. Rovny*	Immobilisation of cesium and divalent metals into single-phase stuffed tridymite-based ceramics
NFC.P12	(Id: 138)	*V. Radchenko, K. Rotmanov, V. Peretrukhin*	Production of artificial metal ruthenium from irradiated technetium
NFC.P13	(Id: 185)	*A. Aloy, A. Strelnikov, I. Shasmolova*	Study of new filter properties ^{137}Cs vapour capture at high temperature
NFC.P14	(Id: 230)	*P. Kovařík, J. John, J. D. Navratil*	Radioactive waste destruction using molten salt oxidation
NFC.P15	(Id: 255)	*V. Avramenko, S. Bratskaya, A. Egorin, D. Marinin, E. Modin, I. Sheveleva, O. Voitenko, V. Zheleznov*	Functionalized latex particles for preparation of colloid-stable nanosized selective sorbents and composite materials for decontamination of radioactive wastes
NFC.P16	(Id: 284)	*Z. Németh, K. Varga, A. Szabó Nagy, J. Schunk, G. Patek, B. Baja, K. Radó*	Comparative study of the corrosion and surface analytical effects of the decontamination technologies
NFC.P17	(Id: 285)	*K. Berkesi, A. Szabó Nagy, E. Deák Horváth, B. Baja, K. Varga, K. Radó, Z. Németh, D. Oravetz, P. Halmos, J. Schunk*	Observation of efficiency of the decontamination technologies in nuclear power plants
NFC.P18	(Id: 49)	*Y. Korneyko, V. Garbuzov, O. Schmidt, B. Burakov*	Study of crystalline ceramics for immobilization of ^{99}Tc
NFC.P19	(Id: 6)	*W. Wu, Q. Fan*	The sorption of cesium on Beishan soil under different physicochemical conditions studied by batch and EDS techniques
NFC.P20	(Id: 52)	*N. Jordan, H. Foerstendorf, V. Brendler*	Selenium(IV) retention onto illite
NFC.P21	(Id: 76)	*M. Felipe-Sotelo, P. Warwick, N. Evans*	Sorption of radionuclides to the cementitious material NRVB under near-field conditions
NFC.P22	(Id: 111)	*N. Evans, K. Farrow, S. Jain, K. Alsop, E. Young, R. Hall, B. Steve*	Sorption properties and behaviour of ^{137}Cs and ^{90}Sr on bentonite clays and magnox sludges
NFC.P23	(Id: 121)	*J. Vejsada, D. Hradil*	Study of cesium and strontium sorption on Rokle bentonite in different electrolytes
NFC.P24	(Id: 224)	*S. Amayri, D. Fröhlich, J. Drebert, T. Reich*	Sorption behaviour and speciation of neptunium(V) on Opalinus clay
NFC.P25	(Id: 301)	*K. Videnská, V. Jedináková-Křížová, H. Vinšová*	Sorption and diffusion changes of ^{134}Cs, ^{99}Tc and ^{129}I radionuclides on bentonites at various conditions
NFC.P26	(Id: 33)	*D. Dobrev, A. Vokal, P. Bruha*	Evolution of the redox potential in the corrosion system
NFC.P27	(Id: 40)	*M. Molnár, L. Palcsu, I. Futó, É. Svingor, Z. Major, M. Veres, P. Ormai, I. Barnabás*	Study of gas generation in real L/ILW containers
NFC.P28	(Id: 86)	*T. Nagy, I. Barnabás, P. Ormai, S. Kapitány, M. Veres, G. Vodila, Á. Bihari, M. Molnár*	Qualification of low and intermediate level radioactive wastes (L/ILW) within the framework of the "Demo" project by a Triathler type portable liquid scintillation spectrometer in Püspökszilágy, Hungary
NFC.P29	(Id: 87)	*T. Nagy, I. Barnabás, P. Ormai, S. Kapitány, M. Veres, G. Vodila, Á. Bihari, M. Molnár*	„Demo" program in Püspökszilágy, Hungary: Qualification of low and intermediate level radioactive wastes by a field γ spectroscopy system
NFC.P30	(Id: 294)	*P. polivka, A. vokal*	Impact of hydrogen generated by iron corrosion on compacted bentonite in deep geological repository

NFC.P31 (Id: 39)	*M. Molnár, L. Palcsu, Z. Major, É. Svingor, M. Veres, T. Pintér*	Dissolved gas measurements of the cooling ponds of Paks Nuclear Power Plant, Hungary
NFC.P32 (Id: 74)	*A. Leskinen, P. Warwick, D. Read*	Using natural organic matter as a remediation material in environmental applications

Nuclear Analytical Methods – NAM s122

Lectures s122

NAM.L01 (Id: 201)	*A. Chatt, Y. Shi*	Simultaneous speciation analysis of arsenic, antimony and selenium in natural waters by neutron activation coupled to HPLC, solvent extraction and solid-phase extraction
NAM.L02 (Id: 211)	*M. Frontasyeva*	NAA for life sciences at FLNP JINR: present and future
NAM.L03 (Id: 190)	*R. Zeisler, R. O. Spatz, R. Paul, J. Fagan*	Use of neutron activation analysis for the characterization of single-wall carbon nanotube materials
NAM.L04 (Id: 318)	*J. Kučera, I. Krausová, P. Dostálek, V. Potěšil*	Determination of silicon in beer and beer processing materials by fast neutron activation analysis. Is beer a possible protective factor in preventing Alzheimer´s disease?
NAM.L05 (Id: 170)	*M. Fukushima, H. Nakano, A. Chatt*	Total and bioaccessible fractions of trace elements in cultivated oyster tissues by INAA, PIXE and ICP-MS
NAM.L06 (Id: 181)	*J. Mizera, Z. Řanda, V. Havránek, I. Tomandl, J. Kučera*	Neutron and photon activation and ion beam techniques in geochemical characterization of moldavites and other impact glasses
NAM.L07 (Id: 193)	*R. Paul*	Development of a radiochemical neutron activation analysis procedure for determination of arsenic in biological materials
NAM.L08 (Id: 225)	*N. Tsibakhashvili, L. Mosulishvili, E. Kirkesali, I. Murusidze, M. V. Frontasyeva, I. Zinicovscaia, P. Bode, T. Th.G.van Meerten*	NAA for studying detoxification of Cr and Hg by Arthrobacter globiformis
NAM.L09 (Id: 368)	*E. A. De Nadai Fernandes, M. Arruda Bacchi, G. Gonçalves Bortoleto, S. R. Vicino Sarriés, G. A. Sarriés, A. E. Lai Reyes, C. L. Gonzaga, A. de Angelis Fogaça*	Neutron activation analysis for assessment of beef origin
NAM.L10 (Id: 210)	*A. A. Pantelica, O. Culicov, M. Frontasyeva, C. R. Badita, I. C. Calinescu*	Elemental concentrations in vegetable species from industrial zones in Romania determined by INAA
NAM.L11 (Id: 131)	*J. F. Facetti-Masulli, V. Romero de Gonzalez, Z. Villanueva de Díaz, P. Kump*	Selected trace elements in pottery of Guarani ethnic group
NAM.L12 (Id: 123)	*F. De Corte*	Comments on the correct interpretation of k_0-factors and on the proper use of the SMELS materials
NAM.L13 (Id: 215)	*R. Acharya, A. V. R. Reddy*	The k_0-based NAA at BARC: Developments, QA/QC and applications
NAM.L14 (Id: 157)	*M. C. Freitas, A. Pacheco*	Native and transplanted lichen and bark as air pollution biomonitors at three different meteorological conditions
NAM.L15 (Id: 178)	*R. Acharya, K. K. Swain, A. Kumar, N. Ajith, R. Verma, A. V. R. Reddy*	Standardization of k_0-NAA for short-lived nuclides using pneumatic carrier facility (PCF) at BARC
NAM.L16 (Id: 253)	*F. Farina Arboccò, K. Strijckmans, L. Sneyers, P. Vermaercke*	The impact of polyethylene vials on reactor channel characterization in k_0-NAA
NAM.L17 (Id: 84)	*R. Jacimovic, M. Taseska, P. Makreski, V. Stibilj, T. Stafilov*	Is extraction of Fe from iron based minerals an appropriate method for determining trace elements?
NAM.L18 (Id: 83)	*S. Szidat, S. Fahrni, M. Ruff, N. Perron, L. Wacker, H. A. Synal*	Online ^{14}C analysis of ultra-small samples with accelerator mass spectrometry (AMS)
NAM.L19 (Id: 264)	*F. Groppi, M. L. Bonardi, S. Manenti*	Radon measurements: a way to disseminate scientific culture among young students in Italy

NAM.L20 (Id: 31)	*C. Landstetter, W. Ringer, A. Achatz, C. Katzlberger*	Determination of ^{90}Sr in soil, grass and cereals
NAM.L21 (Id: 150)	*G. Bilancia, G. Cornara, L. Fornara, D. Rossi, A. Fausti, S. Travaglini, E. Dell'omo, A. Ravazzani, M. Roveri, F. Gentile*	Determination of strontium-90 in raw milk samples
NAM.L22 (Id: 236)	*R. Schupfner*	Radionuclide profiles of ^{14}C, ^{90}Sr, ^{228}Th and ^{232}Th for the purpose of dating human bone tissues or ivory
NAM.L23 (Id: 160)	*Z. Révay, T. Belgya, L. Szentmiklósi*	Prompt γ activation analysis as performed in Budapest
NAM.L24 (Id: 173)	*L. Szentmiklósi, Z. Révay, Z. Kis, T. Belgya*	Upgrade of the prompt-γ activation analysis (PGAA) and neutron induced prompt-γ spectroscopy (NIPS) facilities of the Budapest research reactor
NAM.L25 (Id: 261)	*L. Canella, P. Kudejova, S. Söllradl, A. Türler*	Prompt γ-ray activation analysis for H, B, Bi, Pb and Cd
NAM.L26 (Id: 199)	*V. Kolotov, D. Grozdov, N. Dogadkin*	Development of γ activation imaging method for studying of elements distribution in some ores
NAM.L27 (Id: 166)	*D. Grozdov, V. Kolotov, N. Dogadkin, V. Korobkov*	γ-activation digital autoradiography of large samples. Equalization of activating dose over the microtron-irradiated sample surface
NAM.L28 (Id: 197)	*I. Krausová, J. Mizera, Z. Řanda, D. Chvátil*	Determination of fluorine in selected reference materials by instrumental photon activation analysis
NAM.L29 (Id: 231)	*S. Happel, A. Zulauf, B. M. Mokili, H. Jungclas, A. Bombard*	Characterization of an extraction chromatographic resin for the separation and determination of ^{36}Cl
NAM.L30 (Id: 240)	*M. Korun, D. Glavič Cindro, K. Kovačič, B. Vodenik*	Probability for type I errors in γ-ray spectrometric measurements of drinking water samples
NAM.L31 (Id: 43)	*R. Jakopic, S. Richter, R. Wellum, Y. Aregbe, A. Verbruggen, R. Eykens, F. Kehoe, H. Kühn, Y. Kushigeta, U. Jacobsson, J. Bauwens*	An inter-calibration campaign using various selected Pu spike isotopic reference materials
NAM.L32 (Id: 246)	*J. Mantero, M. Lehritane, S. Hurtado, R. Garcia-Tenorio*	Radioanalytical determination of actinides in refractory matrices by linking alkali fusion with solvent extraction and chromatography extraction.
NAM.L33 (Id: 109)	*L. Palcsu, M. Molnar, Z. Major, E. Svingor, M. Veres, I. Barnabas, S. Kapitany*	Detection of tritium and α-decaying isotopes in L/ILW by helium measurements
NAM.L34 (Id: 162)	*V. Chistyakov, A. Bychkov, E. Erin, M. Kormilitsyn*	Development experience and concept of analytical support complex for industrial-scale fast reactor fuel production

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NAM.P01 (Id: 5)	*D. Vandenberghe, F. De Corte, G. Velghe*	The use of sediment-wax pucks in γ-ray spectrometry for luminescence dating: optimized fabrication and properties
NAM.P02 (Id: 19)	*G. Wallova, G. Wallner, N. Kandler*	Determination of ^{90}Sr and ^{210}Pb in deer bone samples by liquid scintillation counting after ionic exchange procedures
NAM.P03 (Id: 27)	*A. Cecal, F. Ionica, L. Airinei, K. Popa*	Comparative kinetic studies on the corrosion process using two methods based on the β-rays retention and atomic absorption spectroscopy
NAM.P04 (Id: 42)	*J. F. Facetti-Masulli, P. Kump*	Geochemical studies in water bodies of Western Paraguay
NAM.P05 (Id: 48)	*Z. Ilic, A. Vidic, D. Deljkic, D. Sirko*	Determination of radiostrontium in food and water samples using fuming nitric acid
NAM.P06 (Id: 56)	*M. Saito, F. Nishiyama, K. Takahiro*	Effect of Al coating on loss of light elements in polyethylene naphthalate foils during MeV proton beam irradiation
NAM.P07 (Id: 72)	*P. Grabowski, M. Dlugosz, H. Bem*	Determination of ^{210}Po and uranium in high salinity water samples
NAM.P08 (Id: 75)	*D. M. Ho, M. C. Freitas, N. Canha, D. Beasley*	Applicability of k_0-based neutron activation analysis using a Compton suppression γ-ray spectrometer

NAM.P09 (Id: 82)	*E. Constantinou, I. Pashalidis*	Alpha radiometry of uranium by liquid scintillation counting after pre-concentration by cloud point extraction
NAM.P10 (Id: 88)	*L. H. Duong, D. M. Ho, V. D. Cao, T. N. Le, S. T. Nguyen*	Mercury in Bach Ho crude oil of Vietnam as determined by k_0-based instrumental neutron activation analysis
NAM.P11 (Id: 94)	*T. Kiliari*	Alpha radiometric determination of plutonium and uranium isotopes after separation of the radionuclides by cation exchange and liquid extraction
NAM.P12 (Id: 104)	*R. González de Orduña, M. Hult, D. Budjáš, M. Misiaszek, E. Andreotti*	Pulse shape analysis to reduce the background of BEGe detectors
NAM.P13 (Id: 153)	*M. C. Freitas, N. Canha*	NORM at indoor environments using aerosols passively collected at classrooms of Lisbon basic schools
NAM.P14 (Id: 154)	*B. Štrbac*	The use of coincidence summing effect in γ spectrometry for the determination of full energy photopeak efficiency and activity of the ^{60}Co-60 point source
NAM.P15 (Id: 165)	*A. El-Sayed, H. H.F.Aly, N. N.S.Awwad, S. M.El-Afifi*	Separation, preconcentration of lanthanum from monazite and its determination by ^{138}La using non-destructive γ ray
NAM.P16 (Id: 183)	*J. Kameník, J. Mizera, Z. Řanda*	Chemical composition of silica phytoliths. Comparison of different isolation methods
NAM.P17 (Id: 192)	*G. Faria, M. A. Menezes, L. Ribeiro, C. Jacome, R. Jaćimović*	Characterization of Tupiguarani Tradition archaeological ceramics from Espirito Santo, Brazil, applying neutron activation and multivariate statistical analyses
NAM.P18 (Id: 196)	*D. Sas, P. Sladek, J. Janda*	Measuring of gross α and β activity by means of LSC
NAM.P19 (Id: 198)	*V. Chistyakov, V. Momotov, E. Erin*	Modification of precise technique for determining Pu mass fraction by automatic coulometric titration method
NAM.P20 (Id: 203)	*A. Chatt, W. Zhang*	Determination of magnesium in biological materials by neutron activation and anti-coincidence γ-ray spectrometry
NAM.P21 (Id: 205)	*I. Zinicovscaia, N. Tsibakhashvili, L. Mosulishvili, E. Kirkesali, T. Kalabegishvili, S. Kerkenjia, M. Frontasyeva*	Elemental content of indigenous bacteria under different chromium loadings
NAM.P22 (Id: 208)	*R. Acharya, Z. Revay, K. Sudarshan, R. V. Kulkarni, A. V. R. Reddy*	Composition analysis of zirconium alloys by k_0-based PGAA using Budapest cold neutron beam facility
NAM.P23 (Id: 214)	*K. B. Dasari, N. Lakshmana Das, A. V. R. Reddy, R. Acharya*	Analysis of large and non-standard geometry samples of ancient potteries and bricks by internal monostandard NAA using insitu detection efficiency
NAM.P24 (Id: 217)	*A. A. Dalvi, R. Devi P S, N. Ajith, K. K. Swain, R. Verma, A. V. R. Reddy*	Determination of silver and gold in copper concentrate
NAM.P25 (Id: 223)	*D. Arginelli, F. Badolato, O. Avataneo, S. Ridone, M. Montalto, A. Miranti*	Radioanalytical determination of ^{210}Pb and ^{210}Po in sea water samples
NAM.P26 (Id: 228)	*V. Babain, N. Gall, A. N. Bazhenov, L. Gall*	Metal detection in solution by new mass-spectrometrical method
NAM.P27 (Id: 232)	*P. Dararutana, P. Wathanakul, N. Sirikulrat, Y. Thongkam, K. Won-in, S. Pongkrapan*	Applications of nuclear analytical methods in the historical glass in Thailand
NAM.P28 (Id: 233)	*I. Světlík, A. Belanova, M. Vrskova, E. Hanslik, D. Ivanovova, T. Novakova, J. Meresova, L. Tomaskova*	Gross α activity determination in water and ^{210}Po
NAM.P29 (Id: 235)	*T. Novakova, I. Svetlik, G. Wallova, L. Tomaskova, G. Wallner*	Routines of ^{210}Po determination in fluvial sediments for dating purpose
NAM.P30 (Id: 237)	*R. Schupfner*	New kits of radiation sources for the purpose of energy dependent calibration of collimated in-situ-γ-spectrometry
NAM.P31 (Id: 238)	*R. Schupfner*	Radiochemical trace analytical purification procedures during decommissioning of nuclear facilities: experiences and results

NAM.P32 (Id: 244)	*D. Beasley, H. M. Dzung, M. C. Freitas, N. Canha*	Development of k$_0$-cyclic neutron activation analysis at the Portuguese research reactor
NAM.P33 (Id: 247)	*M. Hossain*	Assessment of contamination levels and dispersion in a mining impacted area by INAA and μ-PIXE analysis
NAM.P34 (Id: 257)	*M. A. Menezes, R. Jaćimović*	k$_0$-INAA using comparator and neutron flux monitor at CDTN/CNEN, Brazil: advantages and disadvantages
NAM.P35 (Id: 265)	*P. Sládek, M. Ohera, J. Janda*	Airborne and terrain γ spectrometry monitoring of natural and artificial radioactivity
NAM.P36 (Id: 268)	*M. Mariani, L. Cozzella, N. Cogliati, R. Sibello Hernandez, S. Spezia, E. Trivellone*	Isotopic characterization by Q-ICP-MS of some α emitters in Cuban soil
NAM.P37 (Id: 269)	*E. Tomarchio, S. Rizzo, P. Buffa*	Application of Monte Carlo simulation to design a modular ^{241}Am-Be neutron irradiator
NAM.P38 (Id: 278)	*M. Zoriy, M. Burow, R. Flucht, P. Hill, R. Lennartz, P. Zoriy*	Development of the method for determination of technetium in environmental and biological samples
NAM.P39 (Id: 298)	*J. Lee, B. J. Kim, H. M. Park, K. W. Lee, J. Y. Yun, D. M. Lee*	Development of radioactive xenon analysis system by diagnostic assessment using mass spectrometry
NAM.P40 (Id: 311)	*J. Qiao, P. Roos, M. Miró, X. Hou*	Rapid and simultaneous determination of neptunium and plutonium in environmental samples by extraction chromatography using sequential injection and ICP-MS
NAM.P41 (Id: 317)	*C. Viana, M. A. Menezes, E. Pereira-Maia*	A preliminary assessment of air quality in Mangabeiras' Park, Belo Horizonte, Brazil, using epiphytic lichens as biomonitor
NAM.P42 (Id: 323)	*N. Canha, M. C. Freitas, I. Dionisio, H. M. Anawar, H. M. Dung, C. Pinto-Gomes*	Characterization of abandoned contaminated Valongo mining area at Portugal, by INAA
NAM.P43 (Id: 329)	*M. C. Freitas, A. Pacheco, C. Galinha*	Transfer coefficients from soil to roots of wheat plants by INAA
NAM.P44 (Id: 334)	*C. Galinha, M. C. Freitas, A. Pacheco*	Determination of selenium in soft and durum wheat (plant and grain) using the short-lived nuclide
NAM.P45 (Id: 341)	*G. Lujaniene, G. Duškesas*	Determination of ^{238,239,240,241}Pu, ^{241}Am, ^{242,243,244}Cm, ^{90}Sr, ^{55}Fe and ^{63}Ni in low and intermediate level operational radwaste
NAM.P46 (Id: 344)	*A. Puzas, V. Remeikis, Ž. Ežerinskis, P. Serapinas, A. Plukis, G. Duškesas*	Determination of the impurities of reactor core graphite with neutron activation analysis, X-ray fluorescence and mass spectrometry techniques for graphite waste modeling
NAM.P47 (Id: 346)	*Y. Y. Yoon*	Tritium content distribution in Jeju island groundwater using Ni-Ni electrolytic enrichment method
NAM.P48 (Id: 351)	*C. Oprea, A. Oprea, M. Gustova, M. Niculescu*	Quantitative evaluation of essential and trace elements in nine medicinal plants by IPAA and XRF techniques
NAM.P49 (Id: 352)	*C. Oprea, P. Szalanski, M. Gustova, A. Oprea*	Trace element distribution in human teeth by X-ray fluorescence and multivariate analysis
NAM.P50 (Id: 353)	*C. Oprea, P. Szalanski, R. Ciofu, M. Gustova*	Trace element distribution in crops grown under industrial stress: a multidisciplinary approach
NAM.P51 (Id: 174)	*M. Kubešová, J. Kučera*	Optimisation of neutron flux parameters determination for k$_0$ standardization during irradiation at reactor LVR-15 in Řež
NAM.P52 (Id: 292)	*T. Räty, J. Lehto, X. Hou, G. Possnert, J. Paatero, J. Flinkman, H. Kankaanpää*	^{129}I in Finnish waters
NAM.P53 (Id: 371)	*P. Steier, M. Srncik, G. Wallner, M. Němec, J. John,*	^{236}U in well water - a tool for uranium prospection?
NAM.P54 (Id: 372)	*M. Němec, J. John*	(Radio)Chemistry in AMS – a Servant or a Partner?

Chemistry of Actinide and Trans-actinide Elements – TAN s158

Lectures s158

TAN.L01	(Id: 286)	*C. Düllmann, M. Schädel, A. Yakushev, A. Türler, K. Eberhardt, J. V. Kratz, B. Lommel, M. Maiti, H. Nitsche, J. P. Omtvedt, E. Parr, D. Rudolph, J. Runke, G. Schausten, E. Schimpf, A. Semchenkov, J. Steiner, P. Thörle-Pospiech, J. Uusitalo, M. Wegrzecki, N. Wiehl, D. Ackermann, L. L. Andersson, M. Block, W. Brüchle, J. Dvorak, H. Essel, P. Ellison, J. Even, J. M. Gates, A. Gorshkov, R. Graeger, K. E. Gregorich, W. Hartmann, R. D. Herzberg, F. P. Heßberger, D. Hild, A. Hübner, E. Jäger, J. Khuyagbaatar, B. Kindler, J. Krier, N. Kurz, S. Lahiri, D. Liebe*	New nucleus ^{277}Hs: in between two islands of stability
TAN.L02	(Id: 282)	*A. Yakushev*	Chemical identification of element 114 at TASCA
TAN.L03	(Id: 252)	*J. Dvorak, P. A. Ellison, I. Dragojević, M. A. Garcia, H. Nitsche, L. Stavsetra, K. E. Gregorich*	Independent verification of element 114
TAN.L04	(Id: 133)	*A. Serov*	Element 114 chemistry and what is next?
TAN.L05	(Id: 172)	*I. Zvara*	Vacuum thermochromatography - theory and Monte Carlo simulation
TAN.L06	(Id: 143)	*D. Wittwer, R. Eichler, R. Dressler, A. Serov, D. Piguet, Y. Lobanov, A. Polyakov, Y. Tsyganov, H. Gäggeler*	Gas phase chemical studies of superheavy elements using the Dubna Gas-Filled Recoil Separator - stopping range determination
TAN.L07	(Id: 7)	*K. Popa, P. E. Raison, R. J. M. Konings, D. Bregiroux, G. Wallez*	Progresses on the structural chemistry of the actinide phosphates
TAN.L08	(Id: 97)	*L. Sweet, K. Ziegelgruber, L. Arrigo, S. Peper*	Single crystal X-ray diffraction and Raman characterization of ternary Ce(IV) and Pu(IV) peroxo-carbonate compounds comprised of rare dimeric molecular units
TAN.L09	(Id: 128)	*W. Pemberton, K. Czerwinski, D. HaTchett*	A comparitive study of U(III) and U(IV) complexes in a room temperature ionic liquid system
TAN.L10	(Id: 177)	*A. Barkleit, H. Moll, G. Bernhard, B. Li, H. Foerstendorf, A. Rossberg*	Structural aspects of uranyl complexes with lipopolysaccharide
TAN.L11	(Id: 296)	*J. Narbutt, W. P. Ozimiński, S. Siekierski*	Theoretical study on „itinerant" properties of yttrium and americium(III) in the lanthanide series with respect to stability of their complexes

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TAN.P01	(Id: 132)	*A. Serov, R. Eichler, H. Gäggeler*	Thermochromatographic investigation of ^{113}In, ^{125}Sb and ^{125m}Te in quartz columns
TAN.P02	(Id: 209)	*N. Aksenov, S. Dmitriev, V. Lebedev, G. Bozhikov*	Application of coprecipitation for the study of chemistry of Rf
TAN.P03	(Id: 212)	*N. Aksenov, S. Dmitriev, G. Bozhikov, G. Starodub, D. Filosofov, J. Jon Sun, V. Radchenko, N. Lebedev, A. Novgorodov*	Sorption of Ti, Zr, Hf and Nb, Ta as homologues of Rf and Db from mixed aqueous-organic HF solutions
TAN.P04	(Id: 15)	*A. Kirishima, N. Sato*	Thermodynamic study on the U(VI) complexation with "aliphatic" and "aromatic" di-carboxylic acids by micro-calorimetry
TAN.P05	(Id: 64)	*O. Nipruk, N. Chernorukov, Y. Pykhova*	A study of heterogeneous equilibria in the saturated aqueous solutions of uranoarsenates and uranophosphates of alkaline and alkaline-earth elements

TAN.P06	(Id: 78)	*N. Chernorukov, O. Nipruk, Y. Pykhova*	Synthesis, structure and properties of synthetic trogerite $(UO_2)_3(AsO4)_2 . 2H_2O$
TAN.P07	(Id: 147)	*I. Dreissig, S. Weiss, H. Zanker, C. Hennig, E. Brendler, G. Bernhard*	Investigations on stability and structure of thorium colloids
TAN.P08	(Id: 321)	*N. Torapava, I. Persson, L. Eriksson, D. Lundberg*	Structures of hydrated and hydrolyzed thorium(IV) determined by EXAFS and LAXS
TAN.P09	(Id: 155)	*A. Osipenko, A. Maershin, M. Kormilitsyn, A. Bychkov*	Electrochemical behaviour of curium in chloride melts
TAN.P10	(Id: 156)	*A. Osipenko, A. Maershin, V. Volkovich, M. Kormilitsyn, A. Bychkov*	Study of reaction of CmO^+ and CmOCl formation in chloride melt by spectroscopic method
TAN.P11	(Id: 163)	*V. Chistyakov, A. Baranov, E. Erin, L. Nagajtseva, V. Momotov*	Kinetics of neptunium(V) conversion in strong nitric acid solutions containing potassium phosphotungstate, $K_{10}P_2W_{17}O_{61}$
TAN.P12	(Id: 219)	*A. Ölcer, J. Drebert, T. Reich*	Electrochemical and spectroscopical investigation of uranium in room-temperature ionic liquids
TAN.P13	(Id: 220)	*H. B. Tanh Jeazet, T. Doert, K. Gloe, S. Tsushima, G. Geipel, G. Bernhard, K. Gloe*	Novel U(VI) Schiff base complexes: synthesis, structural characterization, and extraction studies
TAN.P14	(Id: 117)	*V. Radchenko, M. Ryabinin, V. Stupin*	Production and investigation of thin films of metal actinides (Pu, Am, Cm, Bk, Cf)
TAN.P15	(Id: 118)	*V. Radchenko, T. Chernakova*	Radiographic examination of curium alloys with cobalt, iron, and carbon
TAN.P16	(Id: 119)	*V. Radchenko, Y. Pichuzhkina, K. Rotmanov, S. Tomilin*	Formation of ruthenium alloys with curium and technetium
TAN.P17	(Id: 139)	*V. Radchenko, M. Ryabinin, T. Chernakova, S. Tomilin, Y. Pichuzhkina*	Americium alloys with gold and copper
TAN.P18	(Id: 272)	*N. A. Kulagin*	High-resolution X ray study of electronic state of ions with non-stability nuclei in solids

Radionuclides in the Environment, Radioecology – REG s172

Lectures s172

REG.L01	(Id: 256)	*E. Steinnes, R. Gjelsvik, L. Skuterud, H. A. Thørring*	Impact of the Chernobyl accident on Norway: Lessons learnt
REG.L02	(Id: 112)	*Z. Köllő, L. Palcsu, Z. Major, M. Molnár, L. Papp, P. Dombóvári, L. Manga, T. Ranga*	Tritium in the rainwater around the nuclear power plant of Paks, Hungary
REG.L03	(Id: 331)	*D. Strumińska-Parulska, B. Skwarzec, A. Tuszkowska, A. Jahnz-Bielawska, A. Boryło*	Polonium (^{210}Po), uranium (^{234}U, ^{238}U) and plutonium (^{238}Pu, $^{239+240}$Pu) in the biggest Polish rivers
REG.L04	(Id: 169)	*G. Lujaniene, P. Beneš, K. Štamberg, K. Jokšas, D. Vopalka, E. Radžiūtė, J. Šapolaitė, B. Šilobritienė*	Experimental study and modelling of ^{137}Cs sorption behaviour in the Baltic Sea and the Curonian Lagoon
REG.L05	(Id: 11)	*M. Štrok, B. Smodiš, B. Petrinec*	Natural radionuclides in sediments and rocks from Adriatic Sea
REG.L06	(Id: 345)	*P. Povinec*	Recent trends in radiometrics and mass spectrometry technologies - synergy in environmental analyses
REG.L07	(Id: 270)	*F. Schoenhofer*	Radionuclides in drinking water - an overview: The EU directive, analysis, the Austrian standard, geographical distribution, removal and waste problems
REG.L08	(Id: 55)	*G. Wallner, T. Jabbar*	Natural radionuclide concentrations in Austrian mineral waters
REG.L09	(Id: 47)	*L. Benedik, Z. Jeran*	Radiological characterization of drinking and mineral waters in Slovenia

REG.L10	(Id: 276)	*N. Shandala, M. Sneve, S. Kiselev, V. Seregin, A. Titov, N. Novikova, E. Shchelkanova, A. Lukyanec*	Radiation and environmental monitoring at the sites for the RW and SNF temporary storage in Russia
REG.L11	(Id: 266)	*H. Nitsche*	Model compounds to understand bacterial surface functionality
REG.L12	(Id: 102)	*B. Raditzky, K. Schmeide, S. Sachs, G. Bernhard*	Complexation of U(VI) with nitrogen and phosphorous containing ligands in aqueous solution
REG.L13	(Id: 179)	*F. Morcillo de Amuedo, J. M. Arias Peñalver, M. R. Gonzalez Muñoz, C. Hernandez, T. Reitz, M. L. Merroun*	Molecular characterization of the speciation of uranium associated with the marine bacterium Idiomarina loihiensis MAH
REG.L14	(Id: 13)	*F. P. Carvalho*	Polonium-210 as a tracer of energy transfer in marine food chains
REG.L15	(Id: 32)	*M. Černe, B. Smodiš, M. Štrok*	Impact of U-mill tailings of the former uranium mine at Žirovski Vrh (Slovenia) on radionuclide accumulation by wetland plants
REG.L16	(Id: 90)	*H. Foerstendorf, K. Heim, K. Müller*	The sorption processes of Np(V) and U(VI) onto metal oxide phases. The formation of sorption complexes and mechanistic aspects studied by in situ ATR FT-IR spectroscopy
REG.L17	(Id: 194)	*M. C. J. Comarmond, T. Payne, J. Harrison, S. Thiruvoth, K. Müller*	Uranium sorption on various forms of TiO_2 - influence of surface area, surface charge and impurities
REG.L18	(Id: 28)	*Štefan Palágyi, K. Štamberg*	Transport of $^{125}I^-$, $^{137}Cs^+$ and $^{85}Sr^{2+}$ in granitoidic rocks and soil
REG.L19	(Id: 62)	*O. Anjolaiya*	Sorption behaviour of contaminants in soils: pH and humic acid effects on sorption of on CEC of clay soils and minerals, and the mobility of Cd, Cs, Ni and Sr.
REG.L20	(Id: 98)	*O. Zhukova*	Vertical and horizontal distribution of ^{137}Cs in zone of Chernobyl contamination in Russia
REG.L21	(Id: 347)	*S. Ovsiannikova, M. Papenia, K. Voinikava, J. Brown, L. Skipperud, G. Sokolik, S. Svirschevsky*	Migration ability of plutonium and americium in the soils of Polessie State Radiation-Ecological Reserve

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REG.P01	(Id: 8)	*C. Tanase, K. Popa, C. Birsan, A. Oprea, A. Pui*	Translocation of radioactivity from substrate to macromycetes in some mining areas
REG.P02	(Id: 14)	*F. P. Carvalho, J. M. Oliveira*	Radioactivity in soils and horticulture products near old uranium mining and milling sites
REG.P03	(Id: 21)	*A. Nurmukhanbetova, K. Kuterbekov, S. Lukashenko, V. Glushenko, V. Morenko, S. Makenbaeva*	Methods and results of the main negative factors of KOSHKAR-ATA tailing pond with impacts on environment
REG.P04	(Id: 29)	*Štefan Palágyi, P. Franta, H. Vodičková*	Determination of cation exchange capacity of fucoidic sands for Cs^+ and Sr^{2+} under dynamic column conditions
REG.P05	(Id: 36)	*A. Osterc, V. Stibilj*	Origin of ^{129}I in the environment of Slovenia
REG.P06	(Id: 37)	*R. Janovics, Á. Bihari, Z. Major, M. Molnár, L. Palcsu, L. Papp, M. Veres, I. Barnabás, Z. László*	Background level of radioactive isotopes around a L/ILW disposal facility before it started operation at Bátaapáti, Hungary
REG.P07	(Id: 38)	*R. Janovics, Á. Bihari, M. Molnár, Z. Stefánka, M. Veres, M. Braun, I. Somogyi*	Testing automatic groundwater sampling unit by the isotope analytical and dissolved ion tests
REG.P08	(Id: 50)	*J. Rachubik, A. Grosicki*	Radiocesium activity concentration measurements in game meat as a tool for monitoring radioactive contamination in wildlife and ensuring radiological safety for consumers
REG.P09	(Id: 65)	*I. Lopes, M. J. Madruga, A. Mourato, J. Abrantes*	Determination of strontium-90 in foodstuff
REG.P10	(Id: 68)	*Y. Y. Yoon, K. Y. Lee, S. G. Lee, T. K. Kim, T. J. Lee*	Age dating of the hot spring waters in Korea using natural radionuclides
REG.P11	(Id: 79)	*A. Grosicki, J. Rachubik*	Use of selected chelating agents to mobilize radiocesium from the body

REG.P12	(Id: 89)	*V. Hansen, P. Yi, X. Hou, P. Roos, A. Aldahan, G. Possnert*	Speciation of ^{129}I and ^{127}I in seawater profiles from the Kattegat and Baltic Sea
REG.P13	(Id: 93)	*D. Manzon*	Radiation monitoring method features of contamination areas ageing in landscape
REG.P14	(Id: 96)	*H. Papaefthymiou, D. Athanasopoulos, G. Papatheodorou, M. Iatrou, M. Geraga, D. Christodoulou, E. Fakiris*	Distribution of natural radionuclides and ^{137}Cs in the sediments of a Mediterranean fjord-like embayment, Amvrakikos Gulf, Greece
REG.P15	(Id: 105)	*F. Gharbi, S. Baccouche, M. Bentekaya, M. Oueslati, W. Abdelli, A. Trabelsi*	Natural radioactivity in Tunisian and in some imported phosphate fertilizers used in Tunisia
REG.P16	(Id: 107)	*Z. Hölgye, E. Schlesingerová*	Further results in search of transuranium isotopes in effluents discharged to air from nuclear power plants
REG.P17	(Id: 116)	*A. Vasidov*	Radioecological situation in areas of uranium legacy of Uzbekistan
REG.P18	(Id: 125)	*A. Natalia, H. Margarita, I. Raquel, L. Fernando*	Study of ^{7}Be concentration in the north of the Spain
REG.P19	(Id: 152)	*B. Štrbac, M. Milaković, J. Marinković*	Radioactivity concentration in soil samples taken from region of Banja Luka (Bosnia and Herzegovina)
REG.P20	(Id: 158)	*V. Hansen, X. Hou, P. Ross, A. Aldahan, K. G. Andersson*	Speciation of ^{129}I and ^{127}I in soil and sediment samples
REG.P21	(Id: 168)	*G. Lujaniene, P. Beneš, K. Štamberg, J. Šapolaitė, D. Vopalka, E. Radžiūtė, L. Levinskaitė, V. Aninkevičius*	Cs, Pu and Am sorption to natural clay and their various components: laboratory experiments and modeling
REG.P22	(Id: 188)	*M. Nisti, A. J. G. Santos*	Antropogenic radiotracers assessment as a tool to environmental compliance rules and management
REG.P23	(Id: 202)	*M. Buňata*	Plutonium 241 - importance and ways of determination
REG.P24	(Id: 226)	*S. Bister, F. Koenn, M. Bunka, J. Birkhan, T. Lüllau, B. Riebe, R. Michel*	Uranium and long-lived decay products in water of the Mulde river
REG.P25	(Id: 242)	*C. Saueia, B. Mazzilli, F. Le Bourlegat*	Leachability of natural radionuclides (^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Ra) in Brazilian phosphate fertilizers and phosphogypsum
REG.P26	(Id: 250)	*A. A. Pantelica, I. I. Georgescu, E. Pincovschi*	Natural and artificial radioactivity in soils from Romanian sites determined by γ-ray spectrometry
REG.P27	(Id: 251)	*B. J. Kim, D. M. Lee*	Aircraft-carried sampling system for aerial radioactivity monitoring
REG.P28	(Id: 267)	*N. Baumann, T. Arnold*	Investigations on seepage waters of the test site Gessenwiese by TRLFS
REG.P29	(Id: 277)	*N. Shandala, V. Seregin, M. Sneve, S. Kiselev, A. Kosnikov, G. Smith, A. Titov*	Radioecological criteria and norms during remediation of shore infrastructure of Russian nuclear fleet
REG.P30	(Id: 304)	*L. Ishchenko(Krivoshey), N. Karnaukh*	The estimation of radon activity in multistoreyed buildings of industrial iron-ore region
REG.P31	(Id: 316)	*E. Polyakov, N. Khlebnikov*	Fractionation of Sr(II), Th(IV), U(VI) in the natural water samples by nanocomposite track membranes
REG.P32	(Id: 325)	*E. Szeles, Z. Stefánka, Z. Varga*	Environmental monitoring of nuclear materials for safeguards purposes before decommissioning of nuclear facilities
REG.P33	(Id: 330)	*D. Strumińska-Parulska, B. Skwarzec, M. Pawlukowska, A. Boryło*	Plutonium speciation in the southern Baltic Sea sediments
REG.P34	(Id: 332)	*D. Strumińska-Parulska, B. Skwarzec, A. Boryło*	Naturally existing ^{210}Po in human and dog hair samples
REG.P35	(Id: 333)	*D. Boryło, B. Skwarzec, W. Nowicki, D. Strumińska-Parulska*	Polonium ^{210}Po and uranium ^{234}U and ^{238}U in phosphogypsum and surface water at waste dump in Wiślinka from Phosphoric Fertilizers Industry of Gdańsk and their accumulation in plants
REG.P36	(Id: 335)	*K. Y. Lee, Y. Y. Yoon, K. S. Ko*	Temperature effects on emanation of radon from rock to water and on its partition between water and air

REG.P37	(Id: 337)	*R. Druteikiene, V. Remeikis, A. Paškevičius, B. Lukšienė, D. Pečiulytė, D. Baltrūnas*	Microbial activity impact on the change of Pu oxidation states
REG.P38	(Id: 340)	*V. Jobbàgy, U. Wätjen, J. Meresova*	Current status of gross α/β activity analysis in water samples: A short overview of methods
REG.P39	(Id: 348)	*G. Sokolik, S. Ovsiannikova, K. Voinikava, M. Papenia*	State and migratory ability of uranium and radium in the soils of Belarus
REG.P40	(Id: 349)	*V. Jobbàgy, T. Kovàcs, N. Kàvàsi, J. Somlai, G. Szeiler*	Radioanalytical investigations of uranium concentrations in natural spring, mineral, spa and drinking waters in Hungary
REG.P41	(Id: 248)	*G. Jia*	Characteristics of concentration correlations for the pairs of ^{238}U/HCO$_3^-$, ^{234}U/HCO$_3^-$, ^{226}Ra/^{238}U, ^{228}Ra/^{226}Ra, ^{226}Ra/HCO$_3^-$, ^{228}Ra/HCO$_3^-$, ^{210}Po/^{238}U, ^{210}Po/^{226}Ra, ^{228}Th/^{232}Th, ^{228}Th/^{228}Ra, ^{228}Th/HCO$_3^-$ and ^{210}Po/^{210}Pb in drinking water
REG.P42	(Id: 67)	*S. Gordeev*	Long-term monitoring of radioactivity in environmental of the City of Moscow
REG.P43	(Id: 136)	*M. Puzan, V. Kudrjashov*	The accumulation of transuranic elements of the Chernobyl emission by meadow vegetation of the zone Chernobyl NPP
REG.P44	(Id: 339)	*A. Khater*	Distribution pattern of NORM in Red Sea shore sediments and their relation to non-nuclear industries
REG.P45	(Id: 367)	*T. Petrovic, M. Lipoglavšek, B. Zorko, M. Necemer, M. Korun*	Specific activity of ^{40}K in drinking water in Slovenia
REG.P46	(Id: 338)	*A. Khater*	Radioecological impacts of NORM fractionation in phosphate rock beneficiation processes

Production and Application of Radionuclides – PAR s198

Lectures s198

PAR.L01	(Id: 263)	*M. L. Bonardi, F. Groppi, S. Manenti*	High specific activity ^{177g}Lu for metabolic radiotherapy: deuteron cyclotron vs. nuclear reactor
PAR.L02	(Id: 9)	*A. Hermanne, F. Tarkanyi, S. Takacs, R. Adam Rebeles, S. Spellerberg, A. Ignatyuk, H. Schweickert*	Optimisation of the long lived ^{121}Te contaminant in production of ^{123}I trough the ^{124}Xe(p,x) route
PAR.L03	(Id: 16)	*M. Gustova, O. Maslov, A. Belov*	Determination of the valence state of ^{237}U, obtained in the photonuclear reaction ^{238}U (γ, n) ^{237}U using nanostructured material.
PAR.L04	(Id: 17)	*M. S. Uddin, B. Scholten, A. Hermanne, S. Sudar, H. H. Coenen, S. M. Qaim*	Radiochemical measurement of cross sections for the production of the therapeutic radionuclide ^{193m}Pt
PAR.L05	(Id: 44)	*R. Adam Rebeles, P. Van den Winkel, L. De Vis, R. Waegeneer*	PC-controlled radiochemistry system for preparation of NCA ^{64}Cu
PAR.L06	(Id: 30)	*M. Ayranov, D. Schumann*	Production of ^{26}Al, ^{59}Ni, ^{44}Ti, ^{53}Mn and ^{60}Fe from proton irradiated copper beam dump
PAR.L07	(Id: 99)	*M. Molnár, I. Major, I. Svetlík, L. Haszpra, M. Veres, É. Svingor*	Fossil fuel CO$_2$ detection by atmospheric ^{14}C and CO$_2$ mixing ratio measurements in the city of Debrecen, Hungary
PAR.L08	(Id: 101)	*O. Zhukova, V. Golosov, M. Marcelov, V. Belyaev*	Investigation of the dynamics of erosion-accumulating processes using ^{137}Cs and excess ^{210}Pb as a marker of soil material displacement
PAR.L09	(Id: 127)	*F. Monroy-guzman, V. E. Badillo-almaraz, C. J. Rosales*	Sorption of tungsten and rhenium on alumina in ^{188}W/^{188}Re generators
PAR.L10	(Id: 141)	*V. Luchnikov*	Programming of material's properties over extended time intervals via self-irradiation phenomena

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PAR.P01	(Id: 115)	*A. Vasidov, S. Khujaev*	Separation of ^{131}Cs from barium radionuclides
PAR.P02	(Id: 146)	*K. Abbas, N. Gibson, W. Horstmann, U. Holzwarth, F. Arroja, F. Simonelli, J. Kozempel, A. Bulgheroni, I. Cydzik*	An external cyclotron target system for nanoparticle sample activation
PAR.P03	(Id: 151)	*M. Fikrle, J. Kučera, F. Šebesta*	Preparation of ^{95m}Tc radiotracer
PAR.P04	(Id: 159)	*O. Lebeda, M. Fikrle*	Measurement of excitation functions for (d,x) reactions on natural molybdenum
PAR.P05	(Id: 200)	*P. Rajec, V. T. Csiba, M. . Leporis, E. Losonczi Pataky, M. Reich, J. Ometáková*	Preparation and characterization of target for ^{64}Cu production on the IBA 18/9 Cyclotron.
PAR.P06	(Id: 213)	*G. Bozhikov, N. Aksenov, S. Dmitriev, S. Karamian, G. Starodub, G. Vostokin, Y. Albin*	Recovery and isolation of hafnium isomers from ^{176}Yb target irradiated by ^{4}He ions in optimized conditions
PAR.P07	(Id: 239)	*S. Happel, C. Dirks, A. Zulauf, H. Jungclas*	Characterization of a Cu selective extraction chromatographic resin
PAR.P08	(Id: 354)	*J. Ráliš, O. Lebeda*	Isotopic effect observed by distillation of ^{123}I and ^{124}I from irradiated ^{124}TeO$_2$ target matrix

Radiation Chemistry – RCH s206

Lectures s206

RCH.L01	(Id: 290)	*M. Mariani, G. Consolati, F. Quasso, M. Galletta, E. Macerata*	Effects of γ irradiation on polyetylene isophthalate
RCH.L02	(Id: 280)	*M. Cabalka, B. Bartoníček*	Comparison of radiation and thermal ageing simulation procedures for NPP cables
RCH.L03	(Id: 189)	*J. Bárta, M. Pospíšil, V. Čuba*	Photo- and radiation-induced preparation of nanocrystalline copper and cuprous oxide catalysts
RCH.L04	(Id: 305)	*I. Špendlíková, J. John, V. Čuba, P. Lhoták, J. Kroupa*	Radiation stability and extraction properties of thiacalixarenes
RCH.L05	(Id: 243)	*T. Pavelková, V. Čuba, V. Múčka*	Effects of dose and dose rate of γ radiation on catalytic activity of catalase
RCH.L06	(Id: 180)	*V. Múčka, P. Bláha, V. Čuba*	Measurement of growth curves of microorganisms influenced by radiation

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RCH.P01	(Id: 135)	*A. Lizin, S. Tomilin, A. Lukinykh, S. Yudintsev*	Radiation and chemical durability of actinide crystalline matrices
RCH.P02	(Id: 356)	*B. Drtinova, M. Pospisil*	Behavior of cadmium(II) in irradiated aqueous solutions
RCH.P03	(Id: 357)	*E. Aguilar-Ovando, A. Negron-Mendoza, M. Colin*	Radiation chemistry approach to the study of sedimentary microenvironments as models for the protection of bio-organic molecules on the early Earth
RCH.P04	(Id: 358)	*A. Hernandez-Medina, S. Ramos-Bernal, A. Negron-Mendoza, M. Colin*	Behavior under γ irradiation of single crystals of NaCl doped with divalent cations
RCH.P05	(Id: 120)	*V. Radchenko, V. Stupin*	Radiographic effects during self-irradiation of curium intermetallic compounds

Nuclear Methods in Medicine, Radiopharmaceuticals and Diagnostics, Labelled Compounds – RFD s212

Lectures s212

RFD.L01	(Id: 319)	*V. Tolmachev*	Affibody molecules, a novel class of peptide-based radiopharmaceuticals. Optimising labelling chemistry for the best targeting
RFD.L02	(Id: 24)	*A. Popkov, M. Nádvorník, J. Hanusek, T. Weidlich, R. Jirásko, M. Holčapek, J. Čermák, A. Lyčka, V. Langer*	New amino acid synthons for preparation of [^{18}F]FDOPA and α-[^{11}C]methyl amino acids for positron emission tomography
RFD.L03	(Id: 140)	*K. Kersemans, J. Mertens, V. Caveliers*	Optimisation of the radiosynthesis of 4-[^{18}F]fluoromethyl-L-phenylalanine and [^{18}F]FET via a same Scintomics automated synthesis hotboxthree module.
RFD.L04	(Id: 186)	*M. Vlk, J. Sarek, M. Martinez M., T. Elbert, M. Hajduch*	Heterocyclic labelled derivatives of Betulinines
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