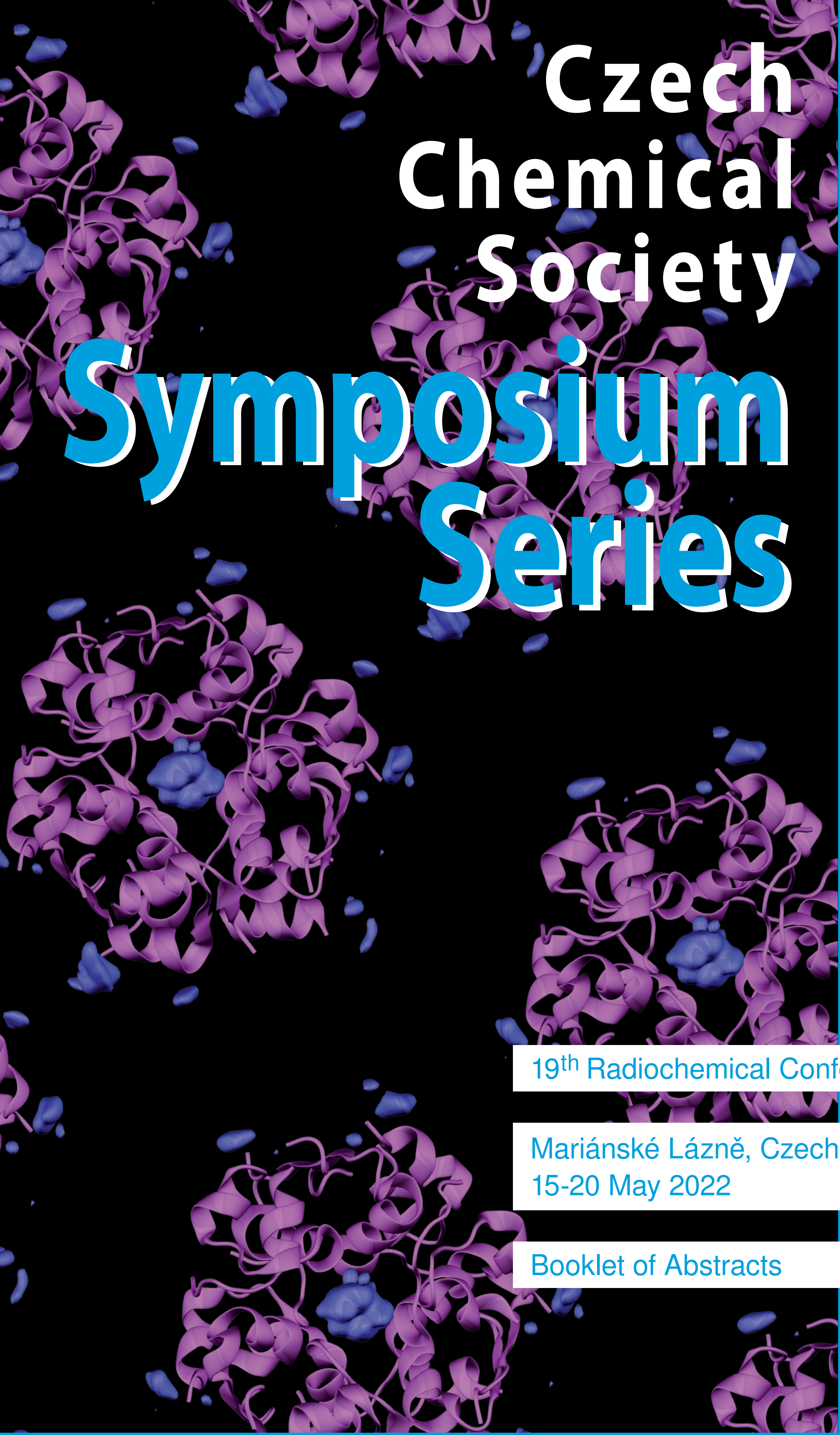

Czech Chemical Society Symposium Series

2

19th Radiochemical Conference

Mariánské Lázně, Czech Republic
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Booklet of Abstracts

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RADIOCHEMIE

19th Radiochemical Conference

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Welcome to the 19th Radiochemical Conference. The conference is held in Mariánské Lázně, the second largest Czech spa and one of the nicest city –gardens in the Czech Republic. RadChem 2022 aims at maintaining more than 60 year tradition of radiochemical conferences (the 1st Radiochemical Conference was held in 1961), dealing with all aspects of nuclear- and radiochemistry, organised in the Czech Republic (or formerly in Czechoslovakia). Despite of global Covid-19 pandemic in the previous months and years and current complicated geopolitical situation – and the associated restrictions and limitations, we strive to continue in the tradition of organising a fruitful and well attended platform for contacts between experts working in both basic and applied research in all aspects of nuclear- and radiochemistry in a face-to-face type event.

Regardless these obstacles, more than 200 scientists from 15 countries registered for the conference. The verbal sessions, where more than 100 invited and contributed presentations are scheduled to be presented, will be complemented by the corresponding poster sessions. Selected “hot topics” in nuclear- and radiochemistry will be covered in six plenary opening lectures delivered by invited recognized experts. The conference will be opened by the prestigious Hevesy Medal Award ceremony including the lecture given by its 2022 laureate, Univ-Prof. Dr. Frank Rösch (Institute of Nuclear Chemistry, Johannes Gutenberg-University Mainz, Germany). In conjunction with the technical programme, an exhibition covering technologies, equipment, technical and management services in areas pertaining to the theme of the conference will be held.

The extensive social programme is expected to provide an opportunity for informal contacts and discussions among the participants. Among others, RadChem 2022 is a proud host of EuChemS Division of Nuclear Chemistry annual meeting and of the European Network on Nuclear and Radiochemistry Education and Training (NRC Network) General Assembly. The interaction of all the above activities is hoped to maintain the high standard of this long-lasting series of conferences and through this contribute to the flourish of our Science.

RadChem 2022 Organising Committee

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Hevesy Medal Award Lecture

RADIOMETAL-THERANOSTICS: THE FIRST 20 YEARS.

FRANK ROESCH

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This HEVESY AWARD MEDAL lecture describes the dawn of radiometal-theranostics. In the 1990ies, a team of researches at the Research Center Juelich, Germany, realized the need to quantify the radiation dosimetry of ^{90}Y-labelled therapeutic compounds. They identified the positron emitter ^{86}Y as an appropriate isotope to quantify uptake kinetics of the radio-yttrium labelled pharmaceuticals by means of PET. Those data were turned into radiation doses of individual compounds in healthy organs and in tumors.

$^{86/90}$Y-EDTMP and $^{86/90}$Y-Citrate served as proof-of-principle theranostics. With the approach established, the $^{86/90}$Y-DOTA-TOC was investigated first in baboons, later in healthy volunteers and finally in patient. This paved the way to the clinical application of ^{90}Y-DOTA-TOC as a valuable treatment of neuroendocrine tumors. It already practiced the methodology of radiotheranostics in a period of time, when this wording did not yet exist. The same time it demonstrated the importance and fruitful interdisciplinary research and development between radio and nuclear chemists, radiopharmacists, medicinal physicists and nuclear medicine doctors.

1. Plenary

ID: 0899
SUPERHEAVY ELEMENT CHEMISTRY – FROM HISTORY TO FUTURE

Alexander Yakushev[a,b], **Ch. E. Düllmann**[a,b,c] **for the TASCA collaboration**

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Chemical studies of the transactinide elements chemistry began 60 years ago with studies of the first transactinide element, rutherfordium (Rf, Z = 104). An experimental method of gas chromatography was applied for chemical separations of many superheavy elements up to element 115, moscovium, the first chemical study of which was recently performed at the recoil separator TASCA at GSI Darmstadt, Germany. The chemical studies of the heaviest elements are subject to drastically decreasing production rates and lifetimes with increasing Z. This calls for developments of new highly efficient and fast techniques. This talk presents a brief historical overview of achievements in the chemical studies of the superheavy elements in the gas phase, recent results, and new developments for future experiments by the TASCA chemistry collaboration.

ID: 0980
RADIOCHEMISTRY – JUST A TOOL OR AN ESSENTIAL PART IN NUCLEAR SCIENCE?

Dorothea Schumann

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Nuclear chemistry works play an essential role in many scientific fields, be it safety of nuclear facilities, environmental science, radioanalytics, nuclear dating for climate reconstruction, nuclear astrophysics, basic nuclear physics or medicine, only to mention part of them. A closer look at all these branches of science reveals that the availability of well-characterised radioactive materials in sufficient quantities as well as the establishment of reliable and efficient measurement methods are indispensable prerequisites for a large number of successful experiments, applications and results. Nevertheless, the contribution that radiochemistry makes in all these cases is often underestimated. Many of our partners from other scientific fields are not aware that the characterisation of the radionuclide inventory of irradiated components to be disposed or the production of a target or a sample is often a lengthy development process. It is not uncommon for entire doctoral theses or larger scientific projects to be needed to develop the appropriate methods and to produce the required samples in the appropriate quantity and quality. This presentation will illustrate with some representative examples the important role of radiochemistry, starting from the determination of the radionuclide inventory of spallation targets foreseen for disposal to the preparation of radioactive targets for cross-section determinations of nuclear reactions and finally addressing sample preparation methods for half-life determinations. The importance of well-characterised radioactive samples for the quality of astrophysical experiments, the effort needed to declare radioactive waste comprehensively, and the power of radiochemistry to determine half-lives of long-lived radionuclides will be shown and explained in detail. Selected results as well as future needs and requirements will be discussed.

ID: 0990
TATTOOS - PLANS FOR THE NEW SWISS ISOTOPE PRODUCTION FACILITY @ PSI

Robert Eichler[a], **Aleksandar Stoyanov Ivanov**[b], **Christian Baumgarten**[b], **Nick, van der Meulen**[b], **Hui Zhang**[b], **Sven Jollet**[b], **Davide Reggiani**[b], **Patrick Steinegger**[c], **Jochem Snuverink**[b], **Daniela Kiselev**[b]

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With its continuous proton beam of world-leading power (1.4 MW), the HIPA facility at the Paul Scherrer Institute (PSI) represents one major asset in the line-up of Swiss large-scale research infrastructures. A recent Swiss Research Infrastructures Roadmap Application from PSI and the University of Zurich (UZ) for 2025-28 named Isotope and Muon Production with Advanced Cyclotron and Target Technology (IMPACT, https://www.psi.ch/en/impact) suggests the high-energy 590 MeV protons to be put towards use for intensified muon production (HIMB - High-Intensity Muon Beams) on one side and isotope production (TATTOOS - Targeted Alpha Tumor Therapy and Other Oncological Solutions) on the other side at unprecedented intensities and amounts. The aim of this presentation is to highlight the scope of the TATTOOS proposal, since it offers unique opportunities for isotope production for fundamental and applied radiochemistry as well as nuclear chemistry. The main goal of TATTOOS is the production of isotopically and radiochemically pure radionuclides for radiopharmaceutical use. However, the production route over high-energy nuclear spallation using 100 uA of the proton beam allows for assessing unprecedented activities of a plethora of common as well as exotic radionuclides. Their mass separation, applying the ISOL technology - pioneered at CERN (ISOLDE, https://isolde.cern) – in conjunction with fast and efficient radiochemical separation - a specialization of the Laboratory of Radiochemistry (LRC, https://www.psi.ch/en/lrc) at PSI - will determine the high isotopical and chemical purity of the products applicable for radiopharmaceutical use as well as fundamental research. Here, we present design challenges of the high power production facility and highlight selected scientific cases specifically of interest to the LRC and the closely collaborating Center of Radiopharmaceutical Sciences (CRS, https://www.psi.ch/de/zrw) at PSI.

ID: 1085

EUROPEAN R&D IN RADIOACTIVE WASTE MANAGEMENT: RELEVANT RESULTS AND JRC CONTRIBUTIONS

MOJMÍR NĚMEC[a], **STEFAAN VAN WINCKEL**[b], **LAURA ALDAVE DE LAS HERAS, STÉPHANE, BREMIER, VINCENZO V. RONDINELLA**

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The Euratom Treaty provides the legal basis for “promoting and facilitating nuclear research in the Member States and for complementing it by carrying out a Community research and training programme”[1]. The Treaty also established the Joint Research Centre (JRC), which is part of the European Commission. The EURATOM Research and Training (R&T) Programme defines the objectives and priorities along which research is being promoted and supported. The research activities are organized in multi annual framework programs (the present cycle covers 2021-2025). Collaborative projects (Indirect Actions) are co-financed by the European Commission Directorate General responsible for EU policy on Research, Science and Innovation (DG RTD); Direct Action research programs and contributions are implemented by the JRC using its own research infrastructure in the directorate for Nuclear Safety and Security. Safe management and disposal of spent nuclear fuel and high level waste constitute a relevant part of the R&T Programme. Moreover, Art. 8 of the Waste Directive[2] requires Member States to make arrangements for research and development activities to cover the needs of the national programme for spent fuel and radioactive waste management. The definition and distribution of R&D efforts is aligned to the strategic research agendas of relevant joint initiatives and platforms, such as the Implementing Geological Disposal Technology Platform, and the EURAD and PREDIS Joint Programmes. This presentation will provide a schematic description of some relevant scientific highlights from past research and an overview of needs / requirements for the research on nuclear waste in the coming years in Europe. In particular, it will present an overview of JRC research activities on safety of spent fuel and high level radioactive waste. Spent fuel management studies cover several stages: cooling in the spent fuel pool; handling, transport, interim storage (with particular emphasis on extended storage); retrieval and handling after storage; disposal in a deep geological repository, including long term safety aspects thereafter. The challenges and the opportunities for collaboration to perform experimental work on "real" spent fuel and waste forms using JRC's research infrastructure, which include hot cells and other shielded facilities, will be also mentioned.

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ID: 1065

THE RADIOLOGICAL RISK RELATED TO NEW NUCLEAR PROPULSION SYSTEMS RESULTING IN CHALLENGES TO MONITORING SYSTEMS

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The nuclear propulsion in application for space probes is developing constantly during last sixty years. At the beginning of those times the world superpowers were working also on nuclear jet propulsion for airplanes, where the heat from combustion of chemical fuel was substituted by the heat generated by nuclear reactors. Since the risk related with crash of airplane having nuclear reactor onboard was found too high those works were stopped in early sixties of XX Century together with atmospheric nuclear ban treaty. However, in March 2018 president of Russia Federation made a public announcement about successful test of nuclear jet propulsion applied to the unmanned cruise class missile. The lack of pilot removes the necessity of biological shielding and reduces the weight of the machine. A bit later, in August 2019 a mysterious radiation accident apparently related to nuclear propulsion happened in the Nyonoksa Bay killing seven person. All technical details remains disclosed, moreover, no evidence for nuclides released in this accident exists in open literature. Earlier, in October 2017, in air over vast European locations the traces of ^{106}Ru was found. Despite many efforts undertaken by researchers worldwide it remains unclear what was the site of release and its circumstances. The only sure fact is that the release point was in Russia. The temporary adopted explanation on release from Mayak is doubtful, since there was no high ^{106}Ru fond on site. It can be noticed, that ^{106}Ru is a useful nuclide for heat generation due to high energy emitted in beta decay (Emax=3.6 MeV) of his short lived daughter ^{106}Rh. On the other hand the practical usefulness is limited due to one year half time of ^{106}Ru. Very similar energetic properties can be found in the system of ^{42}Ar decaying to ^{42}K. This was proposed (but still not confirmed) as the possible nuclide system related to Nyonoksa accident[1]. Traces of those nuclides are very difficult to detect in remote sites. The another possible use of nuclear energy in propulsion of modern flying objects is the replacement of the classical chemical batteries used in so called “kamikaze drons” by the Radio-Thermo Generators (RTG). The advantage of such replacement is in invinitive extension of range of such weapon. During attack the RTG battery will be destroyed causing local high radioative contamination. The contemporary existing monitoring systems in many countries, based mostly on scenario of Chernobyl or Fukushima accidents or nuclear bomb fallout are blind for many variants of possible radioactive releases of nuclides suitable for RTG like ^{90}Sr, ^{238}Pu and especially the ^{42}Ar/^{42}K system. All this suggests that currently the strategies related to monitoring of radioactivity in air should be revised and adopted to new threads[1].

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ID: 1048

RADIOTHERANOSTICS OF THE TUMOR MICROENVIRONMENT: THE MAGIC BULLET?

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Different to specific molecular tumor targets such as the somatostatin receptor of neuroendocrine tumors and the prostate specific membrane antigen of prostate cancer more recently the tumor microenvironment has been identified as a suitable target for cancer diagnosis and therapy. Malignant tumors do not only consist of cancerous cells but also a vast majority of endogenous host stromal cells (e.g., fibroblasts, vascular and immune cells) and extracellular matrix (ECM) components, collectively known as tumor microenvironment (TME). Among all cells within the TME matrix, fibroblasts are dominant cells that have a strong association of their biological functions to all stages of cancer progression and metastasis. Cancer-associated fibroblasts (CAFs) have been implicated to have a strong tumor-modulating effect and are commonly found in most solid tumors. Generally, CAFs account for up to 80 % of all fibroblasts in the TME. FAP contains two types of enzymatic activity: dipeptidyl peptidase and endopeptidase. FAP appears to be a promising target in oncology due to its non-expression in normal fibroblasts and the stroma of benign epithelial tumors compared to its significantly high accumulation mainly on the stromal compartments of a variety of malignant tumors. The challenge to radiopharmaceutical chemistry is to develop molecular targeting vectors of high affinity to FAP und high selectivity to related proteases. Recently, molecular antibodies, small peptides and inhibitors have been turned into molecular imaging probes, with inhibitors utilizing the (4-quinolinoyl)glycyl-cyanopyrrolidine scaffold representing the most exciting class (nanomolar affinity to FAP compared to micromolar affinity to PREP such as the difluoro-version UAMC1110). Radiopharmaceutical chemists from the German Cancer Research Centre Heidelberg, Germany, pioneered in translating this FAP inhibitor into potent diagnostic radiopharmaceuticals by coupling chelators to the amine of the quinoline-part of the inhibitor introducing a number of linker and spacer motifs. The University of Mainz, Germany, utilized a squaric acid (SA)-based linker bridging between several bifunctional chelators (DATA, DOTA, and DOTAGA) to the inhibitor. Currently, ^{68}Ga labelled derivatives such as DOTA-FAPi-04, DOTA-FAPi-46 etc. and DATA5m. SA.FAPi or DOTA.SA.FAPi etc. are showing promising value in diagnosing many different kinds of tumors yielding new information even when compared to the ^{18}F-FDG glycolysis tracer or PET tracers specific to neuroendocrine tumors or prostate cancer. However, those FAPi-based derivatives show a relatively short retention times in the tumor lesions and seem to be unable to deliver a sufficient radiation dose profile. Recent work demonstrated that the tumor retention could be increased by going from a FAPi monomer to a FAPI dimer. The use of the DOTA-derivatives as chelator radiolabeled with attached to the first-in-human FAPi dimer ^{177}Lu-DOTAGA.(SA.FAPi)$_2$ resulted in an agent that was well tolerated in patients up to 4 cycles and demonstrated promising tumor doses and doses to healthy organs. This lecture summarizes the design of the theranostics compounds, the radiochemical and in vitro evaluations, and reports on the clinical application of the theranostic pair ^{68}Ga-DOTA.SA.FAPi monomer vs. ^{177}Lu-DOTAGA(SA.FAPi)$_2$ dimer.

2. Education

ID: 0927

A MOOC IN NUCLEAR- AND RADIO-CHEMISTRY: FROM THE DESIGN TO THE FEEDBACK

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In the recent years the loss of expertise in the nuclear- and radio-chemistry (NRC) is becoming an issue of concern in many aspects. Although this discipline is essential in many fields, few engaging curricula, few career prospects, along with a misperception of this subject, is causing a serious lack of NRC expertise in many parts of the world. In the attempt to contribute to limiting this phenomenon through raising nuclear awareness, the Massive Open Online Course (MOOC) “Essential Radiochemistry for Society” has been developed by the support of experts from all Europe within the H2020 MEET-CINCH project. The consortium was composed of 12 institutions from 8 European countries, England and Norway, with a variety of expertise in many NRC applications. The MOOC has been the result of a collaborative reflection about the external context and the most appropriate way to face the challenge. Furthermore, the proper target group, and the most suited pedagogical approach were discussed. The course is available since 2020 at the Polimi Open Knowledge platform (https://www.pok.polimi.it/) and yearly editions are available from September each year. The MOOC addresses Bachelor students in chemistry, physics, engineering, and general science and medical areas with the intent of improving their awareness of NRC. With a series of lessons that include videos, infographics, articles, quizzes, exercises, and links, the user is guided to discover all the benefits of these disciplines to society, far beyond what is strictly related to nuclear energy. Within the H2020 A-CINCH project, the MOOC editions are continuously monitored, maintained and enriched. Live webinars, held by experts from prestigious universities and research centers, have been organized for the purpose of keeping the course updated on both the most interesting aspects of NRC and cutting-edge scientific research. The present work would like to describe the MOOC development process and discuss strong points and weakness of the followed strategy, according to the feedbacks collected in the first editions among students worldwide.

This work was financially supported by the European Commission within the H2020 MEET-CINCH Project (grant agreement No 754972, https://www.cinch-project.eu) and the H2020 A-CINCH Project (grant agreement No 945301) funded by Euratom research and training programmes 2014-2018 and 2019-2020.

ID: 0942

USAGE MODEL DESIGN FOR BETTER EXPLOITING A MOOC IN NUCLEAR- AND RADIO-CHEMISTRY

<u>Elena Macerata</u>[a], Maddalena Negrin[a], Francesca Concia[a], Sudeep Das, Anu Airaksinen, Marko Štrok, Ioannis Pashalidis[b], Jon Petter Omtvedt[c], Mojmír Němec[d], Susanna Sancassani, Mario Mariani[a]

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In the last decades Massive Open Online Courses (MOOCS) have emerged and continued to grow, gaining recognition worldwide. MOOCs have the unique merit to enable to reach the widest number of users worldwide and offer them high-quality e-learning materials. However, even if completely free and always online, a MOOC could remain a treasure in the hands of few, especially when dealing with unique and specific disciplines. To prevent this occurrence, the development of models for using MOOCs could be of paramount importance, in order to make this resource a valuable tool to help teachers enrich their courses and adopting innovative teaching approaches. In this perspective, within the H2020 A-CINCH project, different usage models have been proposed for the MOOC “Essential Radiochemistry for Society”, developed few years ago within the previous H2020 MEET-CINCH project and currently available at the Polimi Open Knowledge platform (https://www.pok.polimi.it/). The MOOC is addressed to Bachelor students in scientific and medical areas, with the aim to improve their awareness about NRC, realize its involvement in various aspects of everyday life and highlight its multiple beneficial applications in different areas of modern society. The present work would like to describe some usage models developed on the basis of the MOOC usage experiences collected in different European countries, that exploited the whole MOOC or parts of it in different contexts, such as High Education courses, extracurricular initiatives and scientific fairs. The information and data acquired have been analyzed and used to prepare specific toolkits for teachers. The work done has gained even more relevance in the light of the recent pandemic situation, that has overwhelmingly demonstrated that e-learning resources developed by recognized institutions could be well-designed and flexible materials readily available to ensure high-level training.

This work was financially supported by the European Commission within the H2020 MEET-CINCH Project (grant agreement No 754972, https://www.cinch-project.eu) and the H2020 A-CINCH Project (grant agreement No 945301) funded by Euratom research and training programme 2014-2018 and 2019-2020.

ID: 0962

A VIRTUAL LABORATORY FOR EDUCATION AND TRAINING IN NUCLEAR AND RADIOCHEMISTRY

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Nuclear and radiochemistry contains key knowledge and techniques needed by a modern society in order to address societal challenges including the energy sector, medical treatment and diagnosis, material sciences and dating. In order to maintain a high level of expertise, continued education and training is required on all levels using current digital media and attractive, efficient teaching concepts. To this end, A-CINCH develops state of the art virtual and augmented reality tools. One of these is the highly innovative Virtual Laboratory that allows students to perform a broad variety of NRC experiments virtually and repeatedly with Augmented Reality, Virtual Reality and as a web application. This includes analytical radiochemistry, radiopharmacy, a neutron irradiation facility and radiochemistry preparatory work of the nuclear fuel cycle. In a 3D game fashion and by using elements of gamification, users can enter different laboratories, fulfill “tasks”, gain levels and enter the next room. In a most realistic way, labware is handled, samples are prepared and measured. Radiation fields can be visualized, which is an advantage even over a real laboratory rising awareness for the precautions necessary when handling radioactive substances. The Virtual Lab can be used on its own or as an introductory training in preparation for a real world hands on training. This Virtual Laboratory is created as a user-friendly and easy-to-navigate application. The A-CINCH project is the latest of four CINCH-based projects aiming on cooperation in education in nuclear chemistry and radiochemistry. In the first three projects, CINCH, CINCH-II and MEET-CINCH minimum requirements for bachelor, master and postgraduate programs to achieve approved NRC curricula were defined, a number of theoretical and practical courses were developed using hands-on and e-learning approaches and platforms. MEET-CINCH counteracted the massive lack of NRC expertise by building teaching package for high schools and a MOOC on NRC for the general public in order to attract young persons to the NRC field and convey them its fascination and relevance. New education and training approaches based on remote teaching, remote controlled RoboLab experiments, interactive screen experiments and the flipped classroom concept were developed and implemented. ECVET course modules are offered in an e-shop adapted to the needs of end-users. The wide mix of e-learning and presence teaching in nuclear and radiochemistry education aims to increase the number of students and trainees in the field. To address the new target groups directly and efficiently attract the attention of the secondary school students, new didactical tools, suitable for today’s youth, are used. These newly developed courses and tools and all previous CINCH results are accessible via the newly set up CINCH Hub platform.

ID: 0988

RADIOCHEMICAL EDUCATION OF UNIVERSITY STUDENTS USING A FLIPPED CLASSROOM

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At present, frontal-type teaching is still very widespread. Although it has many advantages, it also has major disadvantages. It is one of the least effective ways of teaching, where information is received only passively by students. In most cases, the quantity of information is preferred over quality. Another problem is the lack of space for internal differentiation, where for some students the subject matter may be too easy and for others too difficult. With this method of teaching, students' attention decreases with increasing time, which leads to their passivity. Flipped classroom-type teaching makes it possible to suppress or completely eliminate these shortcomings. Students get acquainted with the subject matter in an interactive form, the emphasis is on information retrieval and critical thinking, which is much more important today than simple memorization. A group discussion on a given subject/problem speeds up its understanding, as students more easily receive information from their schoolmates. Within the group, each student can also participate in activities according to his abilities and his own speed. The project was attended by 23 students of a radiochemistry course at the University of Chemistry and Technology, Prague, in 2022. Before each lesson, the students had the task of getting to know the topic. Tutorial videos were made for this purpose. At the beginning of the course, there was a brief summary of the subject matter covered by the teacher. The students then completed a 10-minute multichoice test containing questions from the subject matter, for which they could use any literature, including resources found online. Then they formed 3-4 member groups and completed an identical test again. Group activities followed, in which the students practiced the subject matter. At the end of the course, the students evaluated this type of teaching in a survey.

ID: 1015
ROBOLAB - PROVIDING REMOTE ACCESS TO A PHYSICAL RADIATION LABORATORY

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The ability to work remotely has been critical for the past two years due to the pandemic. Many students were hindered in developing their experimental skills due to restricted access to campus and laboratories. A partial remedy to such situations can be to provide remote access to physical laboratories. This enables a student to perform real experiments without being physically present. In addition, remote access laboratory experiments can be used as preparation for performing physical hands-on experiments in order to maximize the learning outcome of the very resource-intensive laboratory time in radiation laboratories. With new technologies constantly improving and being made available, one can say that remote is rapidly becoming the new “today” for future generations. “RoboLab” started in 2006 as a pilot project ran by University of Oslo used for teaching students about neutron activation and the effects of gamma absorption. Based on remotely controlled equipment in a real nuclear laboratory, the experiments can be performed without the usual safety training before entering such facilities. Furthermore, institutions without laboratories classified for radioactive work, or just laboratories with limited equipment and radioactive sources, can access experiments they otherwise would not be able to perform. Although hands-on training cannot be substituted with this type of experiments, RoboLab can highly increase awareness, preparation and knowledge prior to entering a laboratory. During the conditions of a restricted access to campus and teachings at many universities, the RoboLab has shown to be a viable alternative to hands-on exercises. The technical foundation of the RoboLabs is the LabView programming environment developed by National Instruments. LabView provides the possibility to quickly and easily develop web-based “virtual instruments”. A cloud-based NI platform establishes communication between the users and the server and allows the operator to securely access and interact with the setup located at UiO – or at any other institution hosting a RoboLab system – from any part of the world through a web browser, without installing additional plug-ins. The first representation of such system is a “Neutron Activation of Silver”-experiment at the University of Oslo (UiO). The application, accessed through a regular web browser, enables the students to irradiate a plate of natural silver with neutrons for a preselected time, make a transfer to a NaI detector and measure the disintegration rate as a function of time during the decay of two short-lived silver isotopes. Another experiment provided by UiO is absorption of gamma rays in matter. Lately an experiment to teach how a radionuclide generator can be made was developed and is ready for pilot runs. It absorbs ^{234}Th on a cation-exchange column and subsequently ^{234m}Pa can be eluted and a decay curve measured. This work is part of the A-CINCH EurAtom project and co-funded by the Norwegian Research Council. Based on the RoboLab concept developed at UiO three other remote controlled exercises was developed at the Leibniz University Hannover, also as part of the CINCH project series. These exercises are: HPGe gamma-spectrometry; Autodeposition on different metals; An ion exchange column with "on-line" detection. The RoboLab remote controlled experiments demonstrates the flexibility offered by adding digital teaching methods to the traditional toolbox.

ID: 1030
A-CINCH: AUGMENTED COOPERATION IN EDUCATION AND TRAINING IN NUCLEAR AND RADIOCHEMISTRY

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The A-CINCH (“Augmented Cooperation in education and training In Nuclear and radioCHemistry”) project is the latest project in the series of CINCH projects and it is corroborating and extending previous achievements. For such purpose, A-CINCH augments CINCH teaching tools developed in the three previous CINCH projects – CINCH, CINCH II and MEET-CINCH – the CINCH project series (www.cinch-project.eu), which were supported from FP7 and H2020. A-CINCH is a Horizon 2020 project No. 954301 that has started in October 2020 with 17 partners from 13 countries with a duration of 36 months. It primarily addresses the loss of the young generation's interest for nuclear knowledge by focusing on secondary, high school students and teachers and involving them by the “Learn through Play” concept. In addition, it focuses on coordination activities in both education and training field at all levels of academic and professional career in nuclear and radiochemistry (NRC). To address these target groups and to attract efficiently their attention, advanced educational techniques such as state of the art 3D virtual reality NRC laboratory, Massive Open Online Courses, RoboLab distance operated robotic experiments, Interactive Screen Experiments, NucWik database of teaching materials, or

Flipped Classroom, are being introduced into the NRC education. Nuclear awareness is strengthened by the High School Teaching Package, Summer Schools for high school students, Teach the Teacher package and others. Current A-CINCH progress will be presented in more detail including overview to e-learning tools designed to attract younger students to the field of radiochemistry and chemistry in general.

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ID: 1060

NEW STUDY PROGRAMMME AT THE CTU IN PRAGUE, FNSPE: DECOMMISSIONING OF NUCLEAR FACILITIES

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One of the strategic aims of the FNSPE is the education of experts for the Czech nuclear industry and research and for the state authorities which are dedicated to the peaceful employment of nuclear energy. By creating the new all-faculty specialized programme “Decommissioning of Nuclear Facilities” FNSPE flexibly react to the needs of the labour market in which a critical shortage of experts in these fields has been manifested. The current statute offers study in both – 3 years bachelor's and 2 years master's continuation degree programs. There is a possibility to study in Czech and English language. In this entirely multidisciplinary programme students will gain excellent knowledge of fuel cycle and chemistry of radionuclides, dosimetry (interaction of ionizing radiation (IR) – detection, monitoring, metrology), materials behavior in ionizing radiation field, waste management, construction and function of nuclear facilities and risk methodology. During the course students will be trained in measuring and analysis techniques and in addition, they will develop abilities for independent decision-making. The education programme is closely connected to its field of application, either in the form of internship or in the form of involvement of experts from industry in lectures.

ID: 1064

DECONTAMINATION LOOP – USEFUL EQUIPMENT FOR THE PRACTICAL TRAINING IN DECONTAMINATION

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Specific and unique equipment – decontamination loop - for the practical training of the students in the field of decontamination was devised and designed at the Department of Nuclear Chemistry, CTU in Prague, and it was manufactured by Wood Nuclear Slovakia s.r.o. The decontamination loop is a modular system intended for study of various decontamination methods and procedures. For this purpose, the system consisting of a number of individual modules such as stirred reactor, modular electrolytic cell for the electrochemical decontamination, sorption and filtration columns, flow-through heaters, solution circulation pumps, flow-through cell for radioactivity measurement, dual extraction unit etc. The loop is equipped with set of flow, temperature and pH sensors and as a whole system is logged and controlled with computer. The equipment is primarily intended for the students of the new study program „Decommissioning of Nuclear Facilities” but its use is much more extensive. For example, the loop will be used for tasks developed in the A-CINCH project (Augmented Cooperation in Education and Training in Nuclear and Radiochemistry) and its Hands-on Training in Decontamination and Decommissioning.

3. Nuclear Analytical Methods

ID: 0891

ACCUMULATION AND EFFECT OF SILVER NANOPARTICLES FUNCTIONALIZED WITH SPIRULINA PLATENSIS ON RATS

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The effect of unmodified and functionalized Spirulina platensis biomass silver nanoparticles on rats during prolonged oral administration was assessed. Silver nanoparticles were characterized by using transmission electron microscopy, while their uptake by the biomass was confirmed using scanning electron microscopy and energy dispersive analysis. The content of silver in the different organs of rats after a period of administration (28 days) or after an additional clearance period (28 days) was ascertained by using neutron activation analysis. In animals administrated with the unmodified nanoparticles, the highest content of silver was determined in the brain and kidneys, while in animals administrated with AgNP-Spirulina, silver was mainly accumulated in the brain and testicles. After the clearance period, silver was excreted rapidly from the spleen and kidneys; however, the excretion from the brain was very low, regardless of the type of nanoparticles. Hematological and biochemical tests were performed in order to reveal the effect of nanoparticles on rats. The difference in the content of eosinophils in the experimental and control groups was statistically significant. The hematological indices of the rats did not change significantly under the action of the silver nanoparticles except for the content of reticulocytes and eosinophils, which increased significantly. Changes in the biochemical parameters did not exceed the limits of normal values. Silver nanoparticles with the sizes of 8–20 nm can penetrate the blood–brain barrier, and their persistence after a period of clearance indicated the irreversibility of this process.

ID: 0897

DEVELOPMENT OF NEW LOW-LEVEL METHOD FOR THE ANALYSIS OF IODINE-129 AND THE ISOTOPIC RATIO ^{129}I/^{127}I DETERMINATION

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Iodine is an extremely volatile element naturally present under various organic or inorganic forms. These physico-chemical characteristics complicate its extraction and quantification. Among the 37 iodine isotopes, ^{127}I is the only stable one and ^{129}I is the radioisotope with the longest half-life (16.1 x 106 y). ^{129}I is chronically and regulatory released by fuel reprocessing plants e.g. Sellafield (UK) and La Hague (France) and also released in the atmosphere by nuclear power plants during normal operation process or in case of an accident. ^{129}I quantification and the determination of ^{129}I/^{127}I isotopic ratio is then essential in a post-accidental context, environmental monitoring field or transfer mechanism studies. ^{129}I quantification is usually performed by γ-spectroscopy or Liquid Scintillation Counting (LSC). These methods don't however allow the determination of the isotopic ratio (^{129}I/^{127}I) and obtained limit of detection (20 mBq/L and 10 mBq/L respectively after chemical treatment) are often higher than the environmental background. AMS allows also the quantification of ^{129}I and ^{129}I/^{127}I. Although performances of AMS no longer have to be demonstrated (excellent sensibility and selectivity), this device is not easily accessible. ICP-MS is an excellent alternative for the determination of ^{129}I and ^{129}I/^{127}I isotopic ratio. However, spectral and non-spectral interferences could disturb the measurement. In the present work, the ICP-MS measurements were performed using an ICP-MS/MS (8900 Agilent®). The measurement medium was meticulously studied to minimize non-spectral interferences such as memory and matrix effects. Concerning spectral interferences, the octopole collision/reaction cell and the two quadrupole mass filters allowed minimizing polyatomic interferences. The isobaric interference due to ^{129}Xe was eliminated after studying different reaction gas (O_2, N_2O and CO_2) with on-mass and mass-shift detection modes. The new method allows the measurement of iodine-129 either directly or after chemical treatment. The chemical treatment was developed, adapted to ICP-MS measurement medium and validated with reference materials. The turnaround time for the experiment is less than a day. A gain in sensitivity up to a factor 200 was observed and allowed reducing the detection limit up to 0.06 mBq/L after chemical treatment, which is 100 times lower than some current methods.

ID: 0909
THE NEW ELECTROPLATING SOLUTION BASED ON A CITRIC-OXALIC-SULFATE MATRIX

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The article proposed a new electrodeposition solution based on a citric-oxalic-sulfate matrix intended for high-resolution electroplating of actinides. The suggested method originated from the well-established oxalate-ammonium sulfate solution, but instead of DTPA and hydroxyl ammonium sulfate, citric acid was used. Since the volume of electrolyte, distance, and current density were discovered to be the same as for standard oxalate-ammonium sulfate solution, the focus was aimed to evaluate the optimal concentration of all compounds, the working range of pH, and the chemical yield, resolution, and the uniformity of deposited layer of the selected actinides, namely Am, Pu, U, and Cm. The obtained results were the same or better than using the standard plating solution while eliminating expensive and harmful chemicals.

ID: 0911
INFLUENCE OF VARIOUS ORGANIC SOLVENTS ON THE PERFORMANCE OF A NEWLY DESIGNED SCINTILLATION COCKTAIL

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The paper describes the influence of various organic solvents on the performance of a newly designed scintillation cocktail. The work mainly focuses on the sample load capacity of the scintillation cocktail in connection with the solvent component to be able to accept the largest possible volume of different types of samples, especially aqueous ones. Previous research has dealt with the ideal composition of the detergent component, which has a major influence on the sample load capacity, however, the solvent component also has a significant impact. The paper discusses properties of the individual components of the scintillation cocktail. New primary and secondary phosphors are discussed that show very promising results for use in liquid scintillation. One of the significant advantages of the proposed scintillation cocktail is the use of only the primary phosphor without the need for a secondary one. Individual properties characterizing the performance of the scintillation cocktail were tested, namely luminescence properties, alpha/beta discrimination ability, sample load capacity, quenching resistance, detection limit, FOM and detection efficiency.

ID: 0917
NEUTRON-IRRADIATED CONCRETE: STRUCTURAL CHARACTERISATION AND GAMMA DOSIMETRY.

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Increased reactivity of radiation-damaged quartz in the alkaline pore water of concrete and its role in radionuclide transport is a subject that is becoming increasingly important as nuclear power plants (NPPs) around the world reach end-of-life. Quartz contributes more than half the weight of most concretes and up to 40 % of the total radioactive waste volume associated with the dismantling of an NPP. The gamma-emitting radionuclides are the major contributors to the radiation dose rate in the concrete biological shield in the intermediate term after NPP shut-down. The appreciable activity and associated half-lives of these radionuclides pose a problem not only during the safe dismantling of NPPs, but also in terms of processing the concrete to reduce the active waste volume for final disposal in a repository. While thermal neutrons are captured by atomic nuclei according to their thermal neutron cross-sections, fast neutrons undergo collision cascades, which cause defects in the structure of materials. The concentration of defects accumulates as a function of neutron energy and fluence. The high covalency and the density of Si-O bonds in tectosilicates, primarily quartz, followed by feldspars, makes these minerals very susceptible to radiation damage. In this study, two inactive concrete powder samples were placed inside an instrumentation channel passing through the first biological shielding of an operating German NPP, respectively for a half- and a full fuel cycle. Using a plant-specific 3D-reactor model in conjunction with Monte Carlo neutron transport calculations, neutron fluences at the sample positions and the resulting gamma activities are calculated. We compare the measured and calculated activities of the gamma-emitting radionuclidesSynchrotron X-ray powder diffractometry was used to examine structural changes on sub-nanoscale. The local structure of the silicate aggregate minerals was investigated by solid-state magic angle spinning nuclear magnetic resonance (NMR) spectroscopy using ^{27}Al and ^{29}Si as probes. With this work we wish to gain insights into the potential for alkali-silica reaction (ASR) in radiation-damaged concrete and the role this plays in the transport or sorption of radionuclides either present in situ via neutron activation or ex situ due to transport of leaked reactor cooling water.

ID: 0919
ANALYSIS OF ISOTOPIC COMPOSITION OF INHOMOGENEOUS SAMPLES BY LA-MC-ICP-MS

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Uranium materials are strictly controlled by the international nuclear safeguards system. However, if such materials get out of regulatory control and subsequently seized, a comprehensive measurement is eventually required to assess the posed hazard, intended use and possible origin. Such nuclear forensic analysis focuses on the analysis of these intercepted nuclear or other radioactive materials to provide information for the investigating authority to avoid the diversion and subsequent malicious use[1-3]. Most nuclear forensic samples contain (in)visible inhomogeneity, which possibly provide suggestions for nuclear forensic conclusions. One of the few possible techniques for such measurements is laser ablation inductively coupled plasma multi-collector mass spectrometry (LA-MC-ICP-MS). As LA uses a focused laser beam scaled down to a few micron, LA-ICP-MS analysis can reveal sample inhomogeneity in the material in question and is able to measure the spatial isotopic composition. However, the precise measurement can be hindered by the instrumental parameters (e.g. laser beam size or scan speed) or by the sample characteristics (e.g. grain size or differences in U isotope enrichment)[4]. The present work investigates the qualitative measurement of U isotopic inhomogeneity for nuclear forensics by LA-MC-ICP-MS. To study the important parameters, a synthetic sample was prepared by the mixing of two solid certified standard materials (SRM 010U and SRM 030U) to mimic an inhomogeneous U sample. The developed LA-MC-ICP-MS method was also measured and compared with the large-geometry secondary ion mass spectrometry. By a rough LA measurement using a line scan or a 2-D area (map) on the sample surface (pre-screening),the points-of-interest in the inhomogeneous sample can be found and selected. After finding the required sample position, a more precise measurement can be performed on the designated small locations. The procedure allows the accurate analysis of the isotopic composition at the relevant spots and significant and the proper identification of the end-products (i.e. the constituting starting materials).

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ID: 0922
CONTRIBUTION OF MASS SPECTROMETRY TO THE ENHANCEMENT OF OBJECTIVES OF RADIOLOGICAL CHARACTERIZATION IN ENVIRONMENTAL SAMPLES

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Turnaround time and performances of standard analytical protocols may not be compatible with the objectives (contributing to knowledge of the radiological state, detecting an abnormal rise in radioactivity as early as possible…) and the levels of radioactivity present in the environment. Research and expertise programs, implemented to explain or predict radioactivity transfers between the different compartments of the environment, require a rapid and consistent identification of isotopes (U, Pu, Th) in order to allow experts to determine the origin of radionuclides. Developments engaged by IRSN in order to meet these requirements have made possible to develop reliable methods for the rapid processing and analysis of solid matrices, including chemical treatment - separation and measurement by mass spectrometry. Some of these techniques have been automated and can be used for emergency response. Others have been deployed as part of regular monitoring metrology, where they help to increase productivity and sensitivity. This communication will be focused on various examples to compare the performances of standard methods with those of the new techniques of preparation and analysis by ICP-MS. This overview of new methods will explain how IRSN's needs are met through faster, more precise and more exhaustive characterizations of environmental samples, both routinely and during radiological emergencies.

ID: 0930

INVESTIGATION OF A NEW APPROACH FOR ^{36}CL DETERMINATION USING PLASTIC SCINTILLATORS

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^{36}Cl is one of the important radionuclides to be controlled in view of radioactive waste disposal from nuclear infrastructures. Determination of ^{36}Cl is challenging due to its volatility, low activity concentration in decommissioning samples and the lack of solid reference materials for method validation. In this paper, a new approach to determine ^{36}Cl in solid samples by using plastic scintillator microspheres (PSm)/resin (PSresin) was examined. The main advantages of using PSm/PSresin are the decrease of the turnaround time (TAT) of the analysis procedure since the PSm/PSresin with the radionuclide adsorbed can be directly measured by liquid scintillation counting (LSC) and avoiding mixed wastes. Samples were pyrolysed to release ^{36}Cl with the Trio-Furnace Pyrolyser from RADDEC. Different set-ups were investigated for trapping and/or separating this ^{36}Cl. Two different approaches were investigated using respectively plastic scintillation microspheres (PSm) and plastic scintillating resins (PSresin). With the PSm, two different set-ups for trapping ^{36}Cl from the pyrolyser were tested:

- using the PSm directly as a trapping material and
- mixing the PSm with the trapping solutions (H_2SO_4 and Na_2CO_3) after applying the combustion procedure.

With the PSresin (TK-TcScint), a selective scintillating resin normally used for ^{99}Tc determination, was tested using a set-up where the trapping solutions (H_2SO_4 and Na_2CO_3) were loaded into the cartridge containing the PSresin, which was placed into a LS vial and directly measured by LSC. This paper will present and discuss a comparison of the chemical recoveries of ^{36}Cl obtained using the various approaches and set ups.

ID: 0932

DEVELOPMENT OF A PSKIT FOR THE FAST AND SELECTIVE ANALYSIS OF ^{99}TC IN DECOMMISSIONING SAMPLES

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Just in Western Europe, more than 160 nuclear facilities should be closed by 2025. This process requires the management of all the elements with a potential content of radioactivity, in order to determine if there are long live radionuclides present and need to be stored in a deep geological repository, if short live radionuclides are present and need to be temporary stored for its decay or if the levels are low enough to be treated as a conventional waste. The measurement of gamma emitters is already solved, but for alpha and beta emitters, it is necessary to develop fast and selective methods of analysis. Among the beta emitters that should be determined, ^{99}Tc (a fission product of uranium) is a key element due to its long half-life, environmental mobility, and high presence in the radioactive wastes. The in-situ measurement of these radioactive wastes would allow a fast decision about the classification of the materials, so more measurement of the samples can be performed and therefore a better management can be done. In this sense, the objective of this work is to develop a PSkit for in-situ fast measurement of ^{99}Tc. PSkits are scintillation vials with a foil of plastic scintillator at the bottom. Also, this foil is coated with a selective extractant, in this case Aliquat·336, a known selective extractant for ^{99}Tc. In this way, the sample is added into the vial, stirred and then poured to finally measure the PSkit directly into a scintillation detector. In this work, first, the synthesis of the foil was optimized in terms of kind of vial, proportion of crosslinker and the porogen. The results obtained shows that the use of a plastic vial, proportions 32:1 and 2:1 (St:DVB in mols) and the use of heptane as porogen produce the best results in terms of detection efficiency and also foil shape and adherence in the vial. Also, the conditions for the PSkit use were also studied. These included the contact time of the solution in the PSkit, the shaking method and the rinsing conditions, being a gentle stirring and a contact time of the sample in the PSkit of 10 minutes enough to obtain retentions around 80 % and detection efficiencies around 50 % with low retentions of the common radiometric interferences of ^{99}Tc. The established procedure was applied to the measurement of simulated samples that reproduces the matrix of real wastes, studying the effect that this matrix had on the retention and detection efficiency. Finally, the quantification errors of the method was determined with the measurement of the simulated samples.

ID: 0936

NUCLEAR ANALYTICAL FACILITIES ON A COLD NEUTRON BEAM AT MLZ, GARCHING

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The cold neutron beam at MLZ offers unique conditions for nuclear analytical chemistry. The analytical facility accommodates several instruments being under development or recently upgraded. This paper summarizes the progress, and the possibilities these instruments offer. The focusing guide is being replaced with a truly-curved elliptical guide ensuring a more homogeneous beam with a smaller divergence and with coinciding focal points, which serves all instrumental setups much better. The detectors and their cooling systems have been renovated and upgraded. At the same time, new spectrometers are employed enabling better Compton suppression, improved timing for coincidence measurement together with list-mode acquisition. These feature will be used in new dynamic in-beam activation analytical measurements. Prompt Gamma Activation Analysis (PGAA) is a routine technique used since the reactor start. It exploits the advantages of the strong neutron beam with measuring small samples (with masses less than a mg), or activating them in the nearly parallel cold neutron beam with counting the activity in a dedicated low-background chamber. In-beam activation analysis (ibNAA) proved to be an important addition to PGAA. This technique can be further developed with many repeated irradiation and counting cycles using a transfer system to a low-background position on the top of the PGAA setup. The irradiation and counting times could be as short as 1 s, while the transfer times a few tenths of a second. With this cyclic in-beam setup, several hard-to-measure elements, like F, Ag, Pb, even ^{19}O isotope become available with much better sensitivities. PGAI is a possible alternative setup enabling imaging combined with PGAA. It is still in development phase. Detecting the neutron-induced charged particles offers a further broadening of the analytical horizon. Activation products emitting no gamma rays can be detected using scintillator detectors, e.g. liquid scintillation counting directly irradiating the sample in liquid form. Another hard-to-measure element P can thus be determined even on trace levels. A new instrument has recently been introduced at the PGAA facility; Neutron Depth Profiling (NDP) also detects the neutron-induced charged particles (protons, tritons and alpha particles) emitted in the surface layers (max 50 um). This allows for the determination of the concentration profiles of certain light nuclides, like ^{6}Li, ^{10}B, or ^{3}He. The FRM II reactor has not been operational for more than year. Its restart is planned for the fall this year in thermal mode. The guide system, optimized for cold neutrons, will provide a lower flux. The new setups can be tested in this operational mode. The instruments will be available for external users with limited capacities.

ID: 0955

ANALYSIS FOR RADIOACTIVITY OF NEGATIVE ION POWDER

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Lots of reports about inhalation risk and external exposure caused by negative ion technology products which include toiletry, wristbands, clothing, and bed mattresses. Negative ion technology can involve the use of minerals that contain a naturally occurring source of radiation such as thorium or uranium. Hassan et al. investigated thirteen NORM-added negative-ion products in terms of organ equivalent and annual effective dose and one of them would contribute about 2 mSv per year annual effective doses. Negative ion powder, which is the raw material of negative ions products containing wallpaper and textile, is available for purchase in several countries. To investigate the radioactive level of negative ion powder in the Chinese market, radioactivity monitoring of negative ion powder was carried out, 12 samples purchased online from different manufacturers were analyzed through γ-spectrometry and α-spectrometry. Samples were measured by HPGe at first, then samples were digested using a microwave to prepare the solution for ^{226}Ra and ^{232}Th measurement. ^{232}Th isolated from other metal ions using an anion exchange column. ^{226}Ra was isolated from other metal ions using a cation exchange column ^{226}Ra and ^{232}Th used ^{225}Ra and ^{229}Th as a tracer to assess chemical yield. After the purified step, the thorium and radium were electrodeposited on stainless steel disc, then insert into the PIPS detector for counting. The determination of 12 kinds of negative-ion powder through gamma spectroscopy was shown in Table 1. From the analysis results, this batch of material contains natural long-lived radionuclides from the uranium and thorium series and without artificial radionuclides. According to the provisions of the IAEA recommended exemption radionuclides activity limit of 1 Bq/g for each radionuclide in the ^{238}U and ^{232}Th series. This batch of samples, except NM12, were seriously exceeded the activity concentration, and some of them have high radioactivity of thorium and it is necessary to evaluate the external dose. Comparing the activity concentrated of ^{234}Th and ^{214}Bi, ^{228}Ac and ^{208}Tl, we found that long-living radionuclides, ^{238}U and ^{232}Th, did not reach the equilibrium balance. If the equilibrium balance of living radionuclide, U-series and Th-series, was destroyed, only measuring the activity concentration of a single daughter cannot guarantee that the activity of ^{238}U and ^{232}Th activity concentration do not exceed the standards. To assess the radioactivity risk of negative ion powder, we used α-spectrometry to determine the activity of ^{226}Ra and ^{232}Th. To determine elemental compositions of the negative ion powder, 5 samples from different manufacturers were analyzed via EDS spectrometer, and the result showed the 5 analyzed samples mainly contained C, O, Si, Al, Ag, Th, and a small number of rare earth elements. The negative ion is mainly released from radionuclides rather than tourmaline powder and rare earth mineral powder. According to the elemental compositions of the sample, negative ion powder is associated with ore or waste residue of associated ore. α-spectrometry was used to determine the radioactive of ^{226}Ra and ^{232}Th. The source for alpha spectrometer analysis was prepared by the electrodeposition technique. The electrodeposited yield over 90 % for both thorium and radium can be achieved by using 2~3 mL of 0.5M HNO_3 and 17 mL of ethanol as electrolyte with 0.4 mA in 60 min deposition time. The next step is to develop an optimal radiochemical procedure to separate thorium and radium from the matrix.

ID: 0956

MEASUREMENT OF ALPHA-EMITTING THERAPEUTIC RADIOPHARMACEUTICAL RA-223 DICHLORIDE INJECTION BY DIFFERENT SOURCE-DETECTOR DISTANCES Γ-SPECTRUM METHOD

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^{223}Ra dichloride injection is a new type of alpha-emitting therapeutic radiopharmaceutical. To accurately measure the radioactivity of ^{223}Ra is very important for drug dose control and assessment of various potential contamination events. Gamma spectrometry, with the advantage of simple sample pretreatment and high accuracy, is the first choice for ^{223}Ra activity measurement. The most important step in the analysis using HPGe is the determination of the number of photons emitted by the source and received by the detector, therefore, the detection efficiency, coincidence effect, and γ-ray emissivity have a great effect on results. The efficiency calibration of the HPGe detector is commonly measured by using the mixed mono-energy γ nuclide solution, however, the price of the mixed mono-energy γ nuclide solution is pretty high. Efficiency calibration also can conduct using the standard sources that emit similar gamma rays with that nuclide to be analyzed. To accurately analyze ^{223}Ra, the present study determined the efficiency calibration curve of BE5030 for different source-detector distances by using mixed mono-energy γ nuclide solution, ^{133}Ba simulative standard solution, and ^{226}Ra simulative standard solution. Multiple γ -rays emitted by ^{223}Ra and its daughters have been used to measure and analyze the activity of ^{223}Ra dichloride injection. The experimental results show that when S-D=17 cm, the deviation of the efficiency which is calibrated by ^{133}Ba standard solution (ƐBa-133), and ^{226}Ra standard solution (ƐBa-133), from the efficiency calibrated by the mixed mono-energy γ nuclide solution (ƐMixed) range from 1 % to 5 %, but when S-D=0 cm, the maximum deviation reach to 31 %. When S-D=17 cm, using the full-energy peak of ^{223}Ra, 351.03 keV, to calculate the activity of ^{223}Ra, the result calculated based on the efficiency calibration curve of ^{133}Ba (RBa-133), the result calculated based on the efficiency calibration curve of ^{226}Ra (RRa-226) and the result calculated based on the efficiency calibration curve of mixed mono-energy γ nuclide solution (RMixed) have a good agreement, the deviation is less than 3 %. Using other full-energy peaks, 269.48 KeV and 271.23 KeV, the deviation reaches to 5 %. When S-D=0 cm, the deviation of the activity between RRa-226 and RMixed is still very small, but the deviation between RBa-133 and RMixed range from 19 % to 31 %. There is a large deviation between the γ -ray emissivity data of ^{223}Ra and daughter nuclides, and the maximum deviation can reach 32 % compared with ENSDF data. The sample preparation process of ^{223}Ra solution does not release ^{219}Rn, nor does it break the radioactive equilibrium between ^{223}Ra and daughter nuclides in the sample. For the BE5030 HPGe detector, the influence of the coincidence effect of 404.83 keV, 427.15 keV, and 831.98 keV on the analysis results are 7.6 %, 12.8 %, and 9.4 %, respectively. To determine high activity level samples, we advise choosing large source-to-detector distances, and the efficiency calibration can be measured by both ^{133}Ba simulative standard solution and ^{226}Ra simulative standard. To determine low activity level samples, we advise choosing small source-to-detector distances, and the efficiency calibration would be better measured by mixed mono-energy γ nuclide solution or ^{226}Ra simulative standard.

ID: 0960

USE OF RADIOTRACERS TO EVALUATE THE BIOACCESSIBILITY OF ESSENTIAL MINERALS FROM DAIRY PRODUCTS

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Globally, it is estimated that more than two billion people suffer from micronutrient deficiencies. Nutrient-rich foods are at the center of creating healthy and sustainable diets, but supplementation approaches based only on the amount of nutrients fail to consider the bioavailability and thus the uptake potential of these nutrients. Therefore, food matrix-based approaches are essential to understand and improve nutrition. In this study we focus on the bioaccessibility of two important minerals, calcium and iron, in dairy products. Calcium stimulates healthy bone growth and prevents osteoporosis, and with anemia affecting a third of the women in reproductive age worldwide increasing dietary iron uptake is now more important than ever. Fortification of staple foods such as milk with these minerals makes for relatively easy distribution amongst the populations, as these products are commonly consumed. However, although dairy products can be an excellent source of many minerals, the body is not always able to absorb them from the product. The aim of this research is to assess the bioaccessibility and exchangeability of calcium and iron in skim milk. The radiotracers ^{45}Ca and ^{55}Fe were used to follow the phase distribution and exchange behavior between casein micelles, serum proteins, and soluble calcium/iron. Fortification salts, including calcium chloride, calcium carbonate and iron chloride, were intrinsically labelled with ^{45}Ca and ^{55}Fe respectively. It was concluded that the addition of $^{45}CaCl_2$ led to an increase of calcium in the soluble phase, indicating an increase in bioavailability, whereas fortification of milk with $^{45}CaCO_3$ was shown to be unlikely to increase calcium bioavailability. Since Fe^{2+} is known to be more bioavailable than Fe^{3+}, Mössbauer spectroscopy was used to assess the oxidation state of iron in milk. Furthermore, using ^{55}Fe as tracer it was shown that the addition of an iron absorption enhancer (ascorbic acid) increases the bioaccessibility of iron, whereas the addition of the iron absorption inhibitor (tannic acid) resulted in a significant decrease in bioaccessibility. Taken together, these results show that rather than focusing just on the amount of supplemented mineral, their initial chemical state as well as conditions present in the digestive system (such as pH) play an extremely important role which cannot be neglected when using supplements. Through the use of radiotracers, a better understanding of the exchange behavior of Ca and Fe in milk was obtained, which is essential to develop effective milk products containing bioaccessible nutrients to combat micronutrient deficiencies.

ID: 0963

COMPREHENSIVE CHARACTERIZATION OF SINGLE HOT PARTICLES: FROM ISOTOPIC COMPOSITION TO BIOAVAILABILITY

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When on April 26, 1986 the reactor of the Chernobyl nuclear power plant exploded, nuclear fuel was in part released as microscopic solid particles. Structurally intact hot particles pose a risk to humans mainly by the (unlikely) path of inhalation. However, over the decades, weathering may lead to considerable release of radionuclides that subsequently can enter the human food chain. Since hot particles differ strongly with respect to morphology, chemical composition and stability, investigations need to be performed on single particles, rather than on bulk samples. We separate and extract the micrometer sized particles from the environmental matrix in a scanning electron microscope (SEM) equipped with a micro manipulator. Subsequently, the particles are fixed by SEM glue on tungsten needles made in-house. Element composition is imaged by EDX-measurements, and radioactive isotopes are detected by gamma spectrometry. To selectively image the elemental and isotopic composition at below 100 nm spatial resolution, we combine secondary-ion mass spectrometry with resonant laser ionization. Identification of actinide elements and fission products is possible at ultra-trace levels, including ^{242m}Am and Cm. Isobaric interferences, particularly ^{238}U/^{238}Pu and ^{241}Pu/^{241}Am, are suppressed by up to five orders of magnitude. In contrast to most mass-spectrometric techniques, only negligible mass is consumed, leaving the particle intact for further studies. Selected individual particles were sequentially leached in ammonium acetate solution, hydrochloric acid, oxalic acid, nitric acid and aqua regia. The solutions are then measured via gamma spectrometry and ICP-MS to analyze their uranium content and the amounts of leached fission and breeding products. Fuel particles from the Chernobyl accident are usually classified according to their appearance, dissolution rates and oxidation state: the further oxidized and brittle UO_2+Z, non-oxidized UO_2 and zirconium merged bearing U_X-ZrY-OZ particles, of which we investigated at least one particle each. The first leaching steps did not attack the structure of the particles, and only small fractions of ^{241}Am and ^{137}Cs were leached under mild conditions, indicating a low bioavailability. Leaching behaviors were mostly in line with the predictions based on the visually assigned classes and oxidation states. From these data, conclusions on potential risks to human health and environment are drawn. It also demonstrates the potential of a technique that is applicable to almost all elements and opens up new scientific applications beyond the nuclear field.

ID: 0975

RADIOMETRIC ANALYSIS OF POTASSIUM, RADIUM AND URANIUM LEVELS IN BRAZIL NUTS

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The paper describes an alternative approach for the determination of uranium, radium-226 and potassium levels in Brazil nuts. Potassium analysis was performed by a NaI detector using 1-gram of nut's ashes, radium analysis was carried out by radon emanation measurements after EDTA-mediated ash dissolution, and uranium levels were determined by alpha spectroscopy after acidic dissolution and liquid extraction of the element. A total of eight different brazil nut samples obtained from local markets has been investigated and their potassium, radium-226 and uranium levels have been compared to literature values, indicating the applicability of the proposed methods.

ID: 0977

FEASIBILITY STUDY ON THE USE OF NEUTRON ACTIVATION ANALYSIS FOR NUCLEAR FORENSICS: DETERMINATION OF ELEMENTAL IMPURITIES IN URANIUM MATERIALS AFTER PRE-IRRADIATION REMOVAL OF URANIUM

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In the framework of Collaborative Materials Exercises (CMX) organized by the Nuclear Forensics International Technical Working Group (ITWG) we received 4 radioactive samples S1 – S4 of unknown origin in two different physical forms (2 powders, 2 metal pieces), which contained unknown content of uranium with unknown enrichment and were asked to answer the following questions: 1) Do samples have the same material properties? 2) Is the material regulated by national law or international agreement? 3) Is a sample consistent with material having a known set of technical specifications? 4) Is a sample consistent with material resulting from a known activity or process? e.g. a particular application or step in the fuel cycle? 5) Is a sample consistent with materials information contained in a database or library? 6) Is the sample consistent with material from a particular origin? In addition to classical forensic examinations, e.g., fingerprints on packaging materials, a variety of analytical methods, namely micro-Raman spectroscopy, XRF analysis, SEM-EDX analysis, XRD analysis, alpha-spectrometry, gamma-spectrometry, SIMS, and ICP-MS were used in the early stage of the exercise to determine the basic physical properties and chemical composition. To extend knowledge about elemental composition of the samples and to gain information about a possible linkage among the samples, NAA was also used in the later stage of the exercise for determination of minor and trace elements. Since neutron irradiation of the samples in a nuclear reactor would yield overwhelming activities of ^{239}U, its daughter ^{239}Np, and fission products, which would mask gamma-ray peaks of other nuclides arisen from neutron activation of minor and trace elements, a procedure of neutron activation analysis (NAA) with pre-irradiation separation of U using UTEVA resin (Triskem International, France) has been developed. The procedure consists in passing the samples dissolved in 3 M HNO_3 through three columns of UTEVA resin packed in 2 mL cartridges (pre-conditioned with 3 M HNO_3) in succession, washing the columns with 30 mL of 3 M HNO_3, reduction of the effluent volume to 5 mL, pipetting effluent aliquots into polyethylene (PE) irradiation capsules, and their evaporation to dryness. A uranium decontamination factor of ~106 was achieved. Thus, using NAA with both short- and long-time irradiation (90 s and 3 h, respectively), contents of the elements Na, Cl, Mn, Ca, Sc, V, Cr, Mn, Fe, Co, Zn, As, Sb, Ba, La, Ce, Sm, Eu, W, and U could be determined, while only limits of detection (LOD) could be evaluated for other 10 – 15 elements. Simultaneously with the aliquots obtained after U removal, two types of blanks (PE capsules and a process blank) were analysed. Most operations with the samples during their preparation for NAA were carried out in a clean laboratory (Class ISO 6 - ISO 5) using plastic ware pre-cleaned by washing in 3 M HNO_3 and rinsed with demi-water. For quality control of NAA (check of the multielemental calibrators), NIST SRM 1547 Peach Leaves was also co-irradiated. The usefulness of the NAA procedure developed for the collaborative exercise is discussed with special emphasis on accuracy and uncertainty of results and element LODs. The results for lanthanoids are also compared with those obtained by ICP-MS.

ID: 0987

SEPARATION AND DETERMINATION OF MOLYBDENUM-93 IN CONCRETE AND STEEL LEACHATE AND OTHER WASTE FROM NUCLEAR POWER PLANTS

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Molybdenum-93 ($T½ = (4.0±0.8)·10^3$ years) is significant long-lived radionuclide contained in the treatment of nuclear waste. ^{93}Mo is formed when a molybdenum rich material is exposed to thermal radiation, especially by neutron capture by ^{92}Mo. Molybdenum occurs as an impurity e.g., in steels in the core area, in the coolant, etc. Several studies were published with procedures of separation of molybdenum in a simple matrix such as water, but only few of them studied more complex matrices as concrete or steel which are the main components in decommissioning of nuclear power plant. The aim of this study was to develop a procedure to separate molybdenum from concrete and steel leachate and other radioactive waste. There is no possibility to obtain a reference material containing ^{93}Mo. That’s why simulated samples were prepared. They were composed of three parts, the first part contained the matrix itself i.e., a pure water, or a leachate of concrete or steel, the second part contained a liquid radioactive waste (concentrate) containing high activity of ^{14}C, ^{54}Mn, ^{60}Co, ^{63}Ni, ^{90}Sr, ^{134}Cs, ^{137}Cs and the third part was addition of known amount of natural Mo tracer. Only two methods achieved satisfactory results i.e., high radiochemical purity and high separation yield. The first method is based on ion exchange chromatography, where is the first step separation on anion exchange resin and the second step is separation on Aliquat 336 based resin. The second method is based on extraction, where molybdenum is extracted from the aqueous phase by a complex with alpha-benzoin oxime into the organic phase of chloroform. The last step for both methods is gravimetric determination by precipitation of lead molybdate which leads to 75 % yield in average for first method for concrete and 85 % for steel and 90 % for pure concentrate, and 85 % for second method in average for concrete and 90 % for steel and 95 % for pure concentrate respectively. Prepared samples on a thin layer on filter were measured by low energy gamma ray spectrometry. ^{93}Mo has two emission lines at 16.5 keV (62 % emission yield) and 18.6 keV (9 % emission yield). All prepared samples were radiochemically pure, which was one of the aims of this work.

ID: 0992

UTILIZATION OF FLUORIDE TARGET SAMPLE MATERIAL FOR ^{239}Pu MEASUREMENT WITH MILEA AMS

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Accelerator mass spectrometry (AMS) is a very sensitive analytical method for determination of ultra-trace concentrations of long-lived radionuclides in environmental samples. Plutonium isotopes $^{239/240}$Pu are anthropogenic alpha emitters with a half-life of 24 and 64 thousand years, a commonly monitored in the environment due to plutonium radiotoxicity. In practice, AMS measurement of $^{239/240}$Pu ratio enables to identify the origin of plutonium emission in the environment, whether it originates from global fallout or releases from nuclear facilities. The aim of this research was to evaluate the performance of prepared plutonium fluoride matrices with the 300 kV MILEA AMS system at ETH Zürich in order to lower the detection limit. The measurements included tuning the system for ^{239}Pu determination using fluoride and oxide target samples, background analyses of both target matrices, focusing on the ^{238}U interference, and ionization efficiency tests for plutonium extracted in the form of PuF_4^-, PuF_5^- and PuO^-. The influence of the different matrices on separation of ^{239}Pu from interfering ^{238}U was investigated by doping selected samples with different amounts of uranium. Plutonium fluoride matrices based on lanthanide co-precipitation have been tested similarly to uranium fluorides as potential target materials in previous studies. Compared to commonly used oxides, these materials of plutonium can possibly suppress isobaric interferences and provide significantly more intense beams and higher ionization yield values over shorter sputtering periods. Analysed samples were prepared by co-precipitation of plutonium from hydrochloric or nitric acid solutions by adding praseodymium chloride carrier and hydrofluoric acid. After washing and drying the material was mixed with lead fluoride to increase the extracted ion currents. Plutonium oxide incorporated into iron oxide carrier mixed with niobium powder was used as a reference material. In both cases, the chemical analysis confirmed successful incorporation of plutonium into PrF_3 and Fe_2O_3, respectively, with co-precipitation yields over 99 %.

ID: 0993

RADIOCHEMICAL ANALYSIS FOR RADIOLOGICAL CHARACTERIZATION OF DECOMMISSIONING WASTE - DETERMINATION OF ^{126}Sn AND ^{93}Mo

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With increased number of nuclear facilities to be decommissioned, radiological characterization of various radioactive waste generated during decommissioning and operation of nuclear facilities is needed for estimation of the total inventory of radioactivity and its variation with time. Among them, determination of hard-to-measure radionuclides is the main challenge. ^{93}Mo is a long-lived (4000 years) radionuclide and decay by electron capture, it is mainly produced by neutron activation of stable molybdenum (^{92}Mo) presented in the construction materials and fuel elements. ^{126}Sn is a long-lived (2.35×10^5 years) fission radionuclide with beta decay and emission of low energy and low intensity gamma rays. Both ^{93}Mo and ^{126}Sn are high mobile radionuclides in the environment, and therefore important for waste repository. Although some methods for determination of ^{93}Mo in waste samples were reported, but not effective to accurately determine them in radioactive samples from nuclear facilities. The method for determination of ^{126}Sn in radioactive waste is not available. In this work, we report methods for determination ^{93}Mo and ^{126}Sn in radioactive materials, such as spent ion exchange resin, metals and concrete. The key strategy is highly remove the main radionuclides in the radioactive samples such as ^{60}Co and ^{137}Cs by multi-steps chemical separation using chromatographic techniques, and purification from the radionuclides with similar chemical properties, such as ^{125}Sb, ^{51}Cr, ^{99}Tc, ^{93}Nb for ^{93}Mo determination. An ICP-MS measurement technique was developed to measure ^{126}Sn after chemical separation, the interference from ^{126}Xe and ^{126}Te was eliminated and suppressed by using sequential mass separator and dynamic reaction cell technique using NH_3 as reaction gas. The developed methods have been successfully applied for the analysis of samples from the decommissioning of nuclear power plants.

ID: 0995

DETERMINATION OF NANOGRAM LEVELS OF COPPER IN A VARIETY OF MATRICES USING INSTRUMENTAL, PRECONCENTRATION AND RADIOCHEMICAL NEUTRON ACTIVATION ANALYSIS

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Copper at low levels is generally considered an essential element. It can be determined by neutron activation analysis (NAA) via $^{63}\mathrm{Cu}(n,\gamma)\rightarrow{}^{64}\mathrm{Cu}$ and $^{65}\mathrm{Cu}(n,\gamma)\rightarrow{}^{66}\mathrm{Cu}$ reactions. The positron-emitting nuclide ^{64}Cu (half-life=12.7 h) is not commonly used because of interferences. The short-lived ^{66}Cu (half-life=5.09 min) can possibly be used in instrumental NAA (INAA). However, it is seldom done in practice due to the Compton background interferences from nuclides such as ^{28}Al, ^{38}Cl, and ^{24}Na. The ^{66}Cu nuclide decays by β-emission and a single non-coincident gamma-ray of 1039.2 keV. The peak efficiency reduction factor (PERF) of this peak was measured as 1.03. INAA in conjunction with anticoincidence counting (INAA-AC) technique was used for analyzing a number of reference materials with varying salt content for copper concentrations as low as 0.06 mg/kg. Most elements of environmental and toxicological importance in natural waters are typically present at µg/kg or lower levels. A reversed-phase extraction chromatographic (RPEC) preconcentration NAA (RPEC-PNAA) method using a porous inert support, namely Amberlite XAD-4 resin coated with the chelating agent 1-(2thiazolylazo)-2-naphthol (TAN) was developed in our laboratory for the extraction of copper from aqueous samples including seawater with an absolute detection limit of 0.106 µg. A radiochemical NAA (RNAA) method was also developed for the determination of very low levels of copper in diets and foods. The method involved irradiation of a sample for 15-20 min followed by rapid dissolution in a mixture of nitric and hydrochloric acids at low heat and coprecipitation of ^{66}Cu with thioacetamide in presence of 10 mg copper carrier. The precipitate was filtered through a polycarbonate membrane, dried, and counted.

ID: 0996

DEVELOPMENT OF MODIFIED GRAPHENE BASED ELECTRODE FOR ANALYSIS OF URANIUM IN AQUEOUS SOLUTIONS

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Presence of uranium in both natural and anthropogenic waters creates demand for the new ways of systematic monitoring of its concentration. New analytical electrode based on powdered graphene and modified with specifically designed zeolite sorbent and similar to well-known and widely used graphite paste electrode, has been tested for the ability to analyze the uranium content in aqueous systems by differential pulse voltametry. The main aim of the work is to assess development opportunities for analytical electrodes usege at typical concentrations for mining sludge and lagoons formed after mining, as well as for adjacent natural waters. The electrode was therefore tested in concentration areas of ppm order and below (up to the order of ppb). Electrode shows pure response with linear calibration curve. Electrochemical measurements with electrodes of this type can serve as fast, easy to use and relatively cheap alternative to commonly used methods requiring complicated and expensive instrumentation.

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ID: 1000

NEUTRON ACTIVATION ANALYSIS AT MLZ

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Recently, the NAA instrument has been fully integrated into the MLZ user system. The FRM II reactor features various short and long high-flux irradiation positions in a well-thermalized neutron field in the range of 1013–1014 cm^{2}s^{-1} with a thermal-to-epithermal ratio up to 12,000. The counting of the irradiated samples take place in a near laboratory of the Radiochemie München (RCM), where currently three HPGe detectors with digital spectrometers are available for NAA. We plan to upgrade the data acquisition by establishing a new smart-controlled sample handling in the near future. This includes the application of list mode for optimum adaption to time dynamics. The current status of the instrument and developments will be discussed in the presentation, as well as NAA applications at MLZ coming from a wide field of science. They are focused on cultural heritage, geology, cosmochemistry, recycling technologies, as well as biology. In particular, we have observed an increasing demand for the development of new standard reference materials needed for other analytical methods and specialized applications like the characterization of recyclables, or the mobile analysis of cultural-heritage objects. Several works are closely connected to projects at the PGAA instrument, like the planned smart-control for NAA, which can be applied for both instruments. A cyclic in-beam NAA at the PGAA instrument will be shown in the presentation of Zs. Révay. The combination of NAA and

PGAA results is also used in various user projects since it increases the number of the elements significantly.

ID: 1001

DETERMINATION OF THE ALPHA-ACTIVITY FROM SAMPLES WITH AN EXTREMELY STRONG BACKGROUND OF BETA-EMITTING RADIONUCLIDES BY LIQUID SCINTILLATION COUNTING

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One of the biggest challenges regarding the exploitation of nuclear energy concerns the proper management of its high-level radioactive waste. Accelerator Driven Systems (ADS) offer a new pathway to address this issue by transmuting the produced nuclear waste. Lead-Bismuth-Eutectic (LBE) represents an attractive choice of coolant for an ADS, since it can serve simultaneously as a spallation source. In such a case, significant amounts of polonium are generated in the coolant, alongside many additional radionuclides. For the safe operation of such systems, the release behaviour of various radionuclides from the coolant needs to be thoroughly investigated. Earlier studies on this subject have been carried out by means of the transpiration technique, using LBE samples containing only one single radionuclide. However, in a running ADS many different impurities will be present in the coolant simultaneously. It is thus important to study the influence of possible chemical interactions between impurities on the evaporation behaviour of hazardous volatile radionuclides. For such studies, we intend to make use of LBE samples from the prototype high power liquid LBE spallation target MEGAPIE. This material is worldwide unique, because exhibits a similar radionuclide inventory as the coolant of an ADS, since it has been irradiated with a high proton current for several months at PSI in 2006. In particular, we are interested in studying whether the evaporation behaviour of polonium is influenced by the presence of other impurities. In order to conduct such experiments, the initial amount of alpha-emitting Po needs to be carefully analysed in the presence of a large quantity of beta-emitters. In this talk, we will present a fast and straightforward pathway to accurately quantify the activity of an alpha-emitting radionuclide in the presence of an extremely high beta background using the technique of liquid scintillation counting. The presented approach is based on the pulse shape discrimination feature of modern TRI-CARB liquid scintillation counters.

ID: 1005

SIMULTANEOUS DETERMINATION OF ORGANICALLY BOUND TRITIUM AND RADIOCARBON BY LIQUID SCINTILLATION METHOD

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Krško Nuclear Power Plant (KNPP) is located in south – eastern part of Slovenia, on the left bank of the Sava River. Emissions of the operational Pressurized Water Reactor (PWR) are mainly ^{14}C and ^{3}H. The dam for the Brežice Hydroelectric Power Plant (BHPP) constructed few kilometres downstream has caused changes in the local climate parameters and hydrology around KNPP. The simultaneous method for determination of OBT and ^{14}C seems efficient tool for the study of the BHPP impact on the distribution, dynamics, and retention time of the ^{3}H and ^{14}C in biota in the vicinity of KNPP. Simultaneous methods for determination of ^{3}H and ^{14}C in organic samples have been already developed. Pyrolizer or oxidizer are commonly used for this purpose because they allow well controlled and fully automated procedure, but detection limits are rather high due to limitations of allowed sample quantity. Paar 1121 oxygen vessel allows combustion of larger sample quantities and is therefore more suitable for the analysis of environmental samples from the vicinity of PWR reactor. On the other hand, the procedure cannot be automated and sample size is limited by its caloric value which might cause big pressure leap during the combustion. The presented method consists of combustion of the sample in a Paar vessel in an oxygen atmosphere where it is converted to CO_2 and H_2O. Formed CO_2 is released through a cleaning system containing two chemicals 0,1M CrO_3 and 0,1M $AgNO_3$ and trapped in the mixture of CarbonTrap and CarbonCount. Formed water is neutralized using Na_2O_2 and $KMnO_4$ followed by liophylisation and mixed with scintillation cocktail. Critical point in this method is determining the right amount of sample for combusting that will provide enough CO_2 for complete saturation of trapping medium and enough water for determination of OBT. Usually, depending on the sample, from 15 to 20 g is enough to satisfy this criterion. The accuracy, trueness, and precision of the simultaneous method will be presented through inter-laboratory comparisons and results of international inter-comparison tests organized by International OBT group. We took part in 4th, 5th and 6th international inter-laboratory OBT exercise organized on grass, fish and quince where we achieved excellent results. For ^{14}C, we compared our results with the official values from the regular KNPP monitoring report. By these comparisons we proved the suitability of the implemented inexpensive, yet accurate and precise simultaneous method for analysis of both radionuclides in the environmental samples.

ID: 1025
NOVEL SAMPLE PREPARATION APPROACH TO INVESTIGATE ^{14}C FROM IRON MATERAIL

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In this study, we present a novel approach for ^{14}C analses of iron material. ^{14}C contamination of iron materail could happen in nuclear reactors, where the neutron flux could produce significant amount of ^{14}C atoms inside the iron based construction elements (vessel wall, tubes, etc). If those elements are going to be disposed in waste, than the ^{14}C concentration should be measured, reported. On the other hand, artefacts made from wrought iron, could also incorporate some C content from the applied heating material (charcoal, wood), which also deliver a measureble ^{14}C content into the iron. This C and ^{14}C might allow the ^{14}C based dating of the production of the iron tool. For all the above listed applications, we need a good sample preparation method, to extract the C from the iron, without any external contamination. As the typical concentration is maximum a few percent C in the iron (m/m %), a complete combustion/oxidation of 0.1 - 1 g iron for this purpose is neccessary. We present an elegant way of this preparation, using a LECO C744 type iron - C analyser. The exhaust gas of this automatized oxidizer is applied for trapping the produced CO_2, for further isotope analyses. About 1 g of iron is completely oxidized within 1 minute by the C744 unit, and the exhaust gas is collected. C yield, and reproducibility of this preparation method is investigated by AMS ^{14}C analyses of known age iron artefacts, and several ^{14}C reference materials. This method could play a major role, when nuclear power plants are decommissioned and huge amount of iron waste has to be classified according their ^{14}C isotope content.

ID: 1034
URANIUM-ISOTOPES IN AUSTRIAN GROUNDWATER MEASURED BY QQQ-ICP-MS AND ALPHASPECTROSCOPY

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The Federal Ministry of Climate Action, Environment, Energy, Mobility, Innovation and Technology funded a monitoring project of the current status of the activity concentration in Austrian groundwater. Measurements of uranium-isotopes (^{238}U, ^{235}U and ^{234}U) were part of this project. The uranium isotopes concentration was determined using two different analytical methods - inductively coupled plasma mass spectrometry (ICP-MS) and alpha-spectrometry as reference method. To increase the detection limit for ^{234}U measured by ICP-MS a concentration step was conducted. This concentration step bases on the already existing, in house validated method Separation of Natural Radionuclides ^{226}Ra, ^{228}Ra, ^{210}Pb, ^{210}Po in water to get an overall method. This method involves after evaporation, the separation into a ^{210}Pb, ^{210}Po fraction by sulfate precipitation and a ^{226}Ra, ^{228}Ra fraction. The chemical recovery is determined by ICP-MS using an added amount of Pb++ and Ba++. The process is followed by two separate measurements with LSC. Uranium isotopes are located in Pb/Po fraction- they are co-precipitated mainly by adsorption and are usually removed by washing the precipitate with nitric acid. This solution was collected and used to determine uranium isotopes. We were able to get a concentration factor of 20 and remove parts of the matrix. We were able to achieve a concentration factor of 20 and remove interfering matrices. ^{235}U was used to calculate a recovery factor for uranium ^{234}. The details of the method development and the reference measurements with alpha spectrometry will be presented.

ID: 1039
DETERMINATION OF ^{129}I BY ACCELERATOR MASS SPECTROMETRY IN FORENSIC ANALYSIS

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The nuclear forensic analysis deals with tracing the origin of nuclear materials found outside the regulatory control. Each material contains a different amount and type of radionuclides that are characteristic of each type of material. This unique composition and characteristic parameters are used to identify unknown samples. They are referred to as “fingerprints” due to their uniqueness. Nowadays, ^{129}I is one of the most observed isotopes of iodine in the environment. The main part of ^{129}I in the environment comes from nuclear power plants, leakage from spent nuclear fuel reprocessing facilities, from nuclear weapon tests, from nuclear accidents. It can be used for age dating, as an oceanic tracer, for water mass exchange monitoring, for research of the geochemical cycle of stable iodine. Iodine exists in nature mainly in trace amounts, so it is necessary to separate and pre-concentrate it from the samples before measuring. The separation of iodine itself is a difficult task, as it is a volatile form of iodine. Three methods are the most commonly used for solid sample preparation – alkaline leaching, acid digestion, and combustion. All methods are time-consuming and require special instrumentation or chemicals. For this purpose, there is a goal to find a new, simple, and fast technique for iodine sample preparation from solid matrices. There are many methods available for iodine determination. The most used and most accurate ones are low energy gamma-ray spectrometry, liquid scintillation counting, inductively coupled plasma mass spectrometry, and accelerator mass spectrometry. AMS is the most used method for the determination of trace amounts of iodine in the samples. It can detect an isotopic ratio of ^{129}I/^{127}I in order of magnitude 10^{-15}. As a part of the RAMSES project, new laboratory with MILEA-type AMS was built in Řež, on which the samples will be measured.

ID: 1058

APPLICATION OF SIMS AND SEM TO ANALYSIS OF ENVIRONMENTAL SWIPES FOR NUCLEAR SAFEGUARDS

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Secondary ion mass spectrometry (SIMS) is one of the analytical techniques used by the International Atomic Energy Agency for the analysis of environmental swipes (ES) for uncovering undeclared nuclear materials and activities. A combination of imaging and mass separation capabilities of SIMS are used for finding particles of interest (containing U/Pu) amidst large amount of non-specific dust and for the determination of isotopic composition. The advantage of analyzing individual particles, compared to processing the whole swipe, is in the impossibility to mask the technology or history of inspected nuclear material by mixing or dilution with a material of varying enrichment. However, the ES often contain more than one enrichment populations and the situations when two uranium particles with different enrichment are too close to be distinguished by SIMS imaging are not exceptional. Consequently, the results for some particles may be affected by isotopic mixing of two or more enrichments. In this work, we used a combination of SIMS and SEM (Scanning Electron Microscopy) for analyses of inspection swipes, and employed the procedures of correlative microscopy, which enabled observation of identical areas by both instruments. First, fast uranium particle search was carried out using SIMS and particles that could produce mixed isotopic compositions were identified. Next, the sample was transferred to SEM and the morphology of those particles inspected. The particles that were a part of a cluster or were within the distance of 10 m or less from other particles were excluded from the next step of the procedure – the SIMS measurement of isotopic ratios at high precision and accuracy. The procedure enabled for more accurate interpretation of the results of the inspection swipe analyses.

ID: 1068

NEUTRON ACTIVATION ANALYSIS AND X-RAY FLUORESCENCE ANALYSIS OF VEHICLE GLASSES FOR FORENSIC PURPOSES

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Vehicle crimes are often associated with the production of glass fragments. Shards found on the crime scene and microfragments attached on suspect clothing are traces available for Law Enforcement Agencies. The refractive index is often used as a parameter for glass profiling. As an additional characterization, chemical composition of glass fragments was studied by instrumental neutron activation analysis and X-ray fluorescence analysis. A set of 75 samples of vehicle glass was procured for the study. The collection included materials with known origin from two vehicle manufacturers in the Czech Republic as well as an arbitrary set of samples from several car brands collected at scrapyards. The combination of the two nuclear analytical methods provided results for more than 33 elements. The distribution of determined values for some elements was rather uniform. However, several elements are promising for forensic applications due to significant differences of their mass fractions in the studied samples (e.g., K, Mn, Se, Rb, Zr, Ce).

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ID: 1072
STATUS REPORT OF THE FIRST AMS LABORATORY IN THE CZECH REPUBLIC AT NPI, ŘEŽ

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The first Accelerator Mass Spectrometry (AMS) laboratory in the Czech Republic has recently been established within a consortium of Nuclear Physics Institute (NPI) of the Czech Academy of Sciences (CAS), Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague (CTU in Prague – FNSPE) and the Institute of Archaeology of the Czech Academy of Sciences, Prague. The laboratory has been built in new premises of NPI, Řež. It is equipped with a 300 kV Multi-Isotope Low-Energy AMS system (MILEA) developed in a collaboration of Ionplus AG and ETH Zurich, Switzerland. MILEA has been designed, but not limited to, for determination of ^{10}Be, ^{14}C, ^{26}Al, ^{41}Ca, ^{129}I, isotopes of U, Pu and other actinoides. This device serves for a national project “Ultra-trace isotope research in social and environmental studies using accelerator mass spectrometry”, acronym RAMSES. Factory and on-site acceptance tests (FAT and SAT, respectively) of MILEA carried out or assisted by Ionplus specialists were finished in summer of 2021 and showed favourable parameters, e.g., blank ratios for the individual rare/stable isotopes. SAT values for the main measured nuclides listed below show that MILEA system at NPI is competitive with large AMS systems. Noteworthy, MILEA is equipped with the Ionplus Gas Interface System (GIS) for direct measurements of $^{14}CO_2$ allowing analysis of ultra-small samples containing only 3 to 100 µg of carbon.

- ^{10}Be/^{9}Be: < 1.4e-15
- ^{14}C/^{12}C (graphite samples): 8e-16 – 9e-16
- ^{14}C/^{12}C (gaseous samples): < 2e-14
- ^{26}Al/^{27}Al: < 2.6e-15
- ^{129}I/^{127}I: 1.9e-14 – 7.1e-14
- ^{236}U/^{238}U: 4.5e-13

as abundance sensitivity. Here we present our first results, mainly ^{14}C measurements and uncertainties of blanks and standards and sample preparation procedures for determination of ^{10}Be, ^{26}Al, and actinoids.

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ID: 1074
POTENTIAL OF INAA IN ELEMENTAL ANALYSIS OF HEROIN, COCAINE, AND METHAMPHETAMINE

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The analysis of illicit drugs composition is required for effective actions of Law Enforcement Agencies. Determination of major as well as trace elements provides additional parameters that could help in identification of drugs origin. Heroin, cocaine and methamphetamine samples were assayed by instrumental neutron activation analysis (INAA) for determination of mass fractions of several elements. For completeness, a set of adulterants used for drugs cutting was also included in the study. The results suggested that INAA with short time irradiation was especially attractive due to its simplicity and short turnaround time. Some adulterants are specific in significantly different content of chlorine in comparison to heroin, cocaine, and methamphetamine (all three as hydrochlorides). Therefore, chlorine analysis has the potential to estimate adulterant used and its quantity. Iodine was quantified in all methamphetamine samples, and mass fractions span four orders of magnitude. This could indicate specific compounds and procedures used in production. Despite INAA unsatisfactory detection limit of sulfur for most materials, its mass fraction was determined in several drug samples. High sulfur content most probably indicated that methylsulfonylmethane and levamisole were used as cutting agents.

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ID: 1076
DEVELOPMENT OF AN INTEGRATED ON-LINE MONITORING SYSTEM FOR TRITIUM AND RADIOACTIVE CARBON USING ROBOTICS TECHNOLOGY

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Tritium and radioactive carbon generated by the operation of nuclear facilities are radionuclides with half-life of 12.3 and 5730 years, respectively, so if leaked, it will have a significant long-term impact on the environment and humans, so continuous monitoring is required. In the case of radioactive carbon dioxide, it is fixed in living organisms through respiration of animal and plant or carbon assimilation when it is emitted to the environment. In addition, since it can affect the human body through the path of the food chain, continuous monitoring in terms of the source of occurrence is necessary. Tritium can also be released into the environment from nuclear facilities in gaseous or liquid form, affecting nearby residents. In Korea, tritium in urine samples of residents around heavy water reactors was detected higher than those of other regions, making it a social issue. Radionuclides discharged into the environment must be analyzed and monitored periodically and continuously, so it is most effective to apply automated monitoring devices. However, it has not yet been automated because pre-treatment such as distillation, heat treatment, and fluorescent liquid mixing are essential to analyze beta nuclides such as tritium and radioactive carbon. In this study, an integrated ^{3}H-^{14}C on-line monitoring system that automates all processes of analysis, including collection of gas samples, pre-treatment, sample injection, mixing of scintillation liquid, and measurement, to automatically monitor tritium and radioactive carbon in gases released into the environment without analyst was developed. The ^{3}H-^{14}C online monitoring system largely consists of a collection part, a mixing part, and a measuring part. The collection part consists of six collection bottles, a cooling device with a peltier thermoelectric module, a catalytic reactor, a peristaltic pump and a solenoid valve, enabling separation and collection of tritium, organic and inorganic radioactive carbon. The mixing part consists of a combination of robot arm, peristaltic pump, and solenoid valve to sequentially perform opening and closing of the vial cap, position and operation control, and injection of a certain amount of collection liquid into the measurement vial. A TDCR type low-level liquid scintillation counter (HIDEX 300SL) was used in the measurement unit, and measurement was configured to automatically start when a robot arm put a vial into the liquid scintillation counter. The tritium and radioactive carbon on-line monitoring system developed in this study is configured to automatically operate all processes necessary for analysis, such as gas sample collection, pre-treatment, injection of collection liquid, mixing of scintillation liquid, and measurement, for at least four months. If the storage and liquid tank are expanded, long-term monitoring is also possible. Through this study, procedures and methods for automatic analysis of tritium and radioactive carbon were established, and the applicability and usefulness of robotics technology could be confirmed when manufacturing automatic radiation monitoring system.

ID: 1080

COMPOUND SPECIFIC RADIOCARBON DATING OF POTTERY VESSELS FROM THE LATE LA TENE SITE SKLARSKE VALLEY (SUMAVA MOUNTAINS, CZECH REPUBLIC)

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Fragments of archaeological pottery are one of the most common artefacts excavated in archaeological sites. However, radiocarbon dating of these artefacts nor their total lipid extracts have not brought reliable radiocarbon results due to wide spectrum of carbonaceous compounds from various reservoirs which contributed to the overall lipid composition. The only possibility is to use a compound-specific radiocarbon dating analysis (CSRA) approach to date specific biomarkers from the sample. In the case of pottery dating, the solution came with the advent of robust analytical chromatographic techniques. By using preparative gas chromatography to isolate the most concentrated fatty acids in pottery lipid extracts and subsequent AMS radiocarbon dating of these acids we are nowadays able to obtain reliable radiocarbon dates of archaeological pottery. In this study, we extracted lipids from 24 fragments of the Late La Tène pottery excavated at the site of Sklarske Valley (Sumava Mountains, Czech Republic) to contribute an overall picture of dietary strategy at the site and to possibly obtain some AMS radiocarbon dates from the concentrated lipid extracts using CSRA approach. Total lipid extracts dominated by long saturated fatty acids, mostly by palmitic (C16:0) and stearic (C18:0) fatty acids. Plant lipids were confirmed by the presence of fatty alcohols, very long fatty acids, and alkanes. Further, resinous lipophilic biomarkers were confirmed by the presence of abietic acid derivatives, and the presence of long mid-chain ketones revealed high-temperature heating of some of the original vessels. The detected fatty acids and their distribution suggested an animal fat type source of the extracted compounds. This was also confirmed by the δ^{13}C and Δ^{13}C values of the palmitic and stearic fatty acids confirming mostly ruminant fat origin. Two of the very concentrated lipid extracts were chosen for compound-specific radiocarbon analysis and were, thus, sampled for preparative GC technique to isolate C16 and C18 FAMEs which were then combusted to CO_2, graphitized, and AMS radiocarbon dated. We were not able to get radiocarbon dates from C16 fatty acids as the amount of isolated carbon was not sufficient for subsequent radiocarbon dating (below 200 µg of carbon). However, radiocarbon dates of C18 fatty acids showed a good agreement with the other dates obtained from the Sklarske Valley site and fitted well with the site chronology. Still, resulting intervals were quite broad and dates could be possibly improved by applying a solvent-less trapping technique to preparative GC process to maximally eliminate any introduction of contaminating exogenous carbon to the isolated sample. This improvement has been undergoing further research.

ID: 1081

COSMIC-RAY BURIAL DATING OF AN EARLY TO MIDDLE PLEISTOCENE HOMININ SITE: PHYSICAL AND CHEMICAL SAMPLE TREATMENT AND PURITY CONTROL VIA AD-HOC MASS SPECTROMETRY SCANS

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The assay of in-situ terrestrial cosmogenic nuclides (TCN) ^{10}Be and ^{26}Al is central to a geo-archaeological project aimed at determining the age of cultural Layer VII. representing a possible Early to Middle Pleistocene (Lower Palaeolithic) hominin occupation at Korolevo, Transcarpathia, western Ukraine. The cobbles (quartz, quartzite and sandstone) from the Layer VII. fluvial terrace on the Tisza River excavated in the 1980's and quartz pebbles of modern Tisza riverbed were selected for processing. Sample crushing, magnetic separation, quartz purification and Be and Al extraction were conducted following standard methods and Be and Al isotope ratios were measured on the DREAMS accelerator mass spectrometer (Helmholz-Zentrum Dresden-Rossendorf). The samples exhibited various levels of weathering, lithology, and mass. To examine potential effects of heterogeneous sample composition or incomplete chemistry on the determination of ^{10}Be and ^{26}Al abundances, ad-hoc mass spectrometry scans were performed following the AMS measurement. Here we summarize the challenges and performance of our methods together with a description of the deficiencies and their impact on the AMS results.

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ID: 1084

ELEMENTAL PROFILE OF TAIL HAIR BY NEUTRON ACTIVATION ANALYSIS FOR BEEF GENETIC AUTHENTICATION

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The growth in demand for quality beef has stimulated the production of animals with superior genetics. Genetic breedings gave rise to cuts of meat with high added value. Cattle breed certification programs have been created in Brazil to assist in the transparency of the production chain and attest to specific quality attributes of certain breeds. However, this mechanism is not enough to prevent fraud that brings economic losses to producers and consumers. Different analytical techniques have been used to discriminate genetic breeds through the intrinsic characteristics of animals to guarantee genetic origin. For this study, neutron activation analysis was used to assess whether the chemical profile of bovine tail hairs allows for the genetic authentication of animals. Tail hairs were collected from 20 high performance animals from Angus and Wagyu breeds, raised in a certified Brazilian farm that operates in a vertical production system, being responsible for the processes of insemination, breeding, rearing, fattening, slaughtering and marketing of the final product, facilitating the traceability of the production chain. Samples were cleaned with deionized water and analytical standard acetone and cut into small pieces with titanium blade scissors. Analytical portions of approximately 100 mg were placed into high purity polyethylene capsules and irradiated in a thermal neutron flux of 10^{13} $cm^{-2}s^{-1}$ at the nuclear research reactor of IPEN/CNEN. Measurement of induced radioactivity was performed by high resolution gamma ray spectrometry. NIST SRM8414 Bovine Muscle and NIST SRM1577c Bovine Liver certified reference materials were used for analytical quality control. The mass fractions of Br, Ca, Co, Fe, K, La, Na, Rb, Sc, Se and Zn were measured in tail hair samples. Statistically significant interactions ($p < 0.05$) were found between gender and genetic breed for the elements Ca, Co, Fe, Na, Rb, Sc and Se. Machine learning algorithms (Multilayer Perceptron, Naïve Bayes, Random Forest, Random Tree and Support Vector Machine) were used to classify Angus male, Angus female, Wagyu male and Wagyu female. Statistically significant differences ($p<0.05$) were found between these four categories. It was possible to discriminate categories with 88 % accuracy, achieving best classification performance for the Random Forest algorithm. The results indicate that the elemental profile of bovine tail hair can be a potential tool for genetic authentication of animals.

ID: 1086

APPLICATION OF FLUORIDE TARGET MATERIALS IN AMS MEASUREMENT OF URANIUM ISOTOPES

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Accelerator mass spectrometry (AMS) is arguably the most sensitive method for determination of isotopic ratios for a variety of trace nuclides in many different fields of interest, including uranium-236 and since recently also uranium-233. Both these radionuclides, being almost solely anthropogenic, occur in the environment at extremely low concentrations, usually reaching well below 10^{-10} for uranium-236 and even lower for uranium-233. Related analyses of these isotopes serve not only as a reliable fingerprint of nuclear contamination for safeguards monitoring purposes but also allow for distinguishing between sources of emission, including different types of fission reactors and global fallout[1]. Besides, an ever-growing demand for uranium-236 determination persists in marine sciences, where it can be used as a highly conservative tracer. Due to low concentrations, a high overall sample-to-detector efficiency is required, which depends strongly on initial chemical separation yield as well as subsequent chemical form used for negative ion production during measurement. To improve the ionization efficiency, a sample preparation method based on lanthanide fluoride co-precipitation and mixing with lead fluoride has been developed[2] and further modified. Applicability of uranium-fluoride target materials has been evaluated by series of ionization efficiency and isotopic ratios measurement realised in collaboration with Laboratory of Ion Beams Physics, ETH Zurich and VERA AMS Laboratory, University of Vienna.

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ID: 1087

DISCRIMINATION OF CAT AND DOG FOODS USING NEUTRON ACTIVATION ANALYSIS AND MACHINE LEARNING

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Domestication of dogs and cats took place thousands of years ago, altering the behavior and physiology of animals. Since then, the number of pets has increased year after year, reaching nowadays 471 million dogs and 373 million cats in the world. On average, half of a pet's cost of living is related to food, mainly dry kibble. The existence of exclusive pet foods is relatively recent. The first dog food dates back to the late 19th century, until then pets were fed what was available to the owners, such as bread, bones and pieces of meat. Foods for cats and dogs are similar in visual aspects but different in composition. Despite being mammals living in the same environment, dogs and cats have physiological differences that require specific nutritional needs. Dog and cat food follows distinct formulations, manufacturing processes and legal requirements. Cats that have access to dog food, or vice versa, can suffer from various diseases and malnutrition. Given the multitude of commercial brands available in the Brazilian market, we sought to evaluate their chemical profiles, as well as compliance with the recommendations of the Association of American Feed Control Officials (AAFCO). The study was carried out with representative samples of food for dogs (n=96) and for cats (n=103). Samples categorized by dogs, puppies, cats and kitten were submitted to neutron activation analysis to determine the chemical elements Br, Ca, Co, Cr, Cs, Fe, K, La, Na, Rb, Sb, Sc, Se, U and Zn. Data analysis were performed by univariate and multivariate non-parametric statistics and supervised machine learning algorithms (Random Forest, Multilayer Perceptron, Adaboost, Kneighbors and Support Vector Machine) to discriminate the categories. Significant differences were found for the elements Fe, K, Na, Se that are nutritionally important in diets, according to AAFCO recommendations. Non-parametric multivariate tests showed significant differences for all the food categories. The results indicated that dog food has a greater number of non-conformities with nutritional recommendations, presenting more samples with higher levels of toxic elements (U and Sb). The differences between the dog and cat foods were substantial enough to allow discrimination with supervised machine learning algorithms, achieving a maximum accuracy of 81.3 % by the Multilayer Perceptron.

ID: 1088
SUGARCANE QUALITY IN THE SUGAR-ENERGY AGRIBUSINESS ASSESSED BY NEUTRON ACTIVATION ANALYSIS

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Sugarcane has been a versatile crop in Brazil and an integral part of its economy by providing key components such as sugar, ethanol and energy. It is the number one source of renewable energy, accounting for 17 % of total energy consumption. Sugar is the country´s fourth largest export commodity. Brazil is the world’s largest sugar producer with 34.65 million tons in 2021, maintaining its worldwide share of 20 % in production and 40 % in exports. Brazilian sugarcane has a leading role in the global sugar market and in the biofuel energy market. The country has stood out in the last 50 years for having continuously implemented a biofuel program for its entire fleet of vehicles, through the use of a hybrid fuel of ethanol and gasoline. A milestone was the National Alcohol Program (Proálcool), launched in 1975 with the objective of stimulating the production of ethanol to meet the needs of the domestic market and the development of an environmentally and economically sustainable automotive fuel policy. However, a challenging aspect of ethanol and sugar production depended on the harvesting system, a previously manual process after burning the cane standing in the field, which increased the amount of mineral impurities adhering to the delivered stalks to the sugar and ethanol industry. The mechanized harvesting system with sophisticated technology was then consistently deployed to solve the problem, increasing productivity and minimizing production costs. For this study, a series of sugarcane truckloads were sampled at the entrance to the sugar mill industry, using a sampling probe at various positions of the load. Distinct textures of soil (clayey and sandy) and climatic conditions (dry and rainy seasons) were observed. Sugarcane and soil samples were processed (drying, milling and packaging) for irradiation in the nuclear research reactor of the Nuclear and Energy Research Institute (IPEN/CNEN), São Paulo, SP. The elemental profiles of the samples were obtained by neutron activation analysis allowing to estimate the amount of mineral impurities in the raw material with high precision, using the tracers Sc, Th, Fe, and Hf, found in much higher levels in soils than in sugarcane. Data processing was performed using robust multivariate data science (NPMANOVA) and artificial intelligence as supervised machine learning for classification (Neural Networks, Support Vector Machine and Deep Learning). We also applied supervised machine learning for prediction (Random Forest, Logistic Function and Robust Regression) and unsupervised machine learning (Principal Components Analysis with Biplot and Cluster Analysis), canonical functions for dimension reduction and visual analytics. The SAS language and the WEKA program were used. The adopted methodology worked efficiently to detect differences in the content of mineral impurities in different conditions of environmental humidity and type of soil where the sugarcane was grown. It was possible to demonstrate probabilistically ($p\text{-value} < 0.01$) and by inductive artificial intelligence the differences in field origin and quality of labor work in the sugarcane harvesting, for possible corrective actions. The discriminating functions inferred by artificial intelligence could be applied in a bot, in order to optimize the quality and productivity of the system, as this technological package allows detecting potential for improvement in the raw material that enters the industry.

ID: 997
COMBINED RADIOCHEMICAL SEPARATION AND MULTICOLLECTOR ICP-MS APPROACH TO DETERMINE ^{135}CS AND ^{135}CS/^{137}CS ISOTOPIC RATIO

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The radiological characterization process conducted during the lifecycle of a nuclear facility is paramount in all operative phases of decommissioning, waste management, and site environmental monitoring to enhance safety of workers and public and mitigate the environmental footprint of nuclear technology. Most of radiocesium contamination can be easily detected like ^{134}Cs and ^{137}Cs, and continuously monitored because of the short/medium-term risk of radiation exposure. Besides, the nuclide vector includes hard-to-measure radionuclides like ^{135}Cs ($t_{1/2}$ above 10e6 y), one of the major radionuclides responsible for the long-term environmental impact of a waste repository because of its high mobility. The recent development of advanced non-radiometric methods coupled with selective radiochemical separations enables the detection of the low abundant pure beta emitter ^{135}Cs by the evaluation of ^{135}Cs/^{137}Cs isotopic ratio that could identify different contamination sources and monitor their dispersion downstream of plant operations. The low abundance of ^{135}Cs needs a high recovery yield from the matrix and an effective separation from potential interfering elements before accurately assessing ^{135}Cs/^{137}Cs isotopic ratio. The removal of polyatomic (^{95,97}Mo) and especially of isobaric (^{135,137}Ba) interferences is the most challenging issue. Notably, the procedure of Cs recovery based on the selective ammonium molybdophosphate (AMP) ion-exchanger and its subsequent complete dissolution introduces a large amount of Mo and Ba contaminants in the eluted Cs solution. This approach requires a complicated purification step. So far, several strategies have been attempted by using combined anion and cation chromatography systems following the Cs pre-concentration.

In this work, a chromatographic system based on the ammonium molybdophosphate polyacrylonitrile (AMP-PAN) resin is being developed and combined with a Multi-Collector ICP-MS (Plasma 3 – Nu Instruments). Firstly, the conditioning, loading, and stripping conditions of AMP-PAN resin have been verified with a surrogate waste containing 200 ppm Cs and 80 ppm Co, Sr, Ni in 1 M HNO_3. The effluent concentrations measured at different time intervals showed full breakthrough of Co, Sr and Ni and high retention of Cs (> 90%). Thereafter, the use of a 5 M NH_4NO_3 stripping solution has led to high Cs recovery (~ 90%) and a negligible amount of Mo. The optimization of selectivity towards interfering elements, especially Ba, is being conducted with further experiments on a multi-element feed solution of 1 – 4 M HNO_3 containing 20 ppm Cs and 2 ppm Ba, Mg, Al, K, Ca, Mn, Fe, Co, Ni, Zn, Sr, Pb, Bi. To this purpose, several purification strategies are being investigated. Removal of contaminants has been attempted by preliminary co-precipitation (i.e. calcium phosphate), extraction chromatography (i.e. Sr-resin), and washing (i.e. 5 M HNO_3) from the AMP-PAN resin to improve Cs selectivity and decontamination factors. To evaluate the proposed method, Cs recovery yield and decontamination factors of interfering elements have been assessed by Q-ICP-MS, while Ba pattern has been detected by magnet scanning of the MC-ICP-MS.

The optimized radiochemical method is being tested and validated with some real samples and reference materials (IAEA 385, 446), where the concentration of interfering elements could be orders of magnitude higher. In these cases, ^{134,137}Cs have been also monitored by gamma spectrometry. Moreover, laboratory intercomparison could be fostered for the calculation of ^{135}Cs/^{137}Cs isotopic ratio.

4. Chemistry of Nuclear Fuel Cycle, Radiochemical Problems in Nuclear Waste Management

ID: 0878

STATICS AND KINETICS OF SORPTION OF CO-60 WITH COMPOSITE SORBENT T35 IN THE PRESENCE OF HUMIC ACIDS

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To elucidate the impact of humic acids (HA) on chemical affinity of Co(II) to inorganic sorbents experiments on statics of sorption Co (II) by the Thermoxide T35 (composition ZrO_2-K_2Ni [Fe $(CN)_6$]) in the chloride-acetate solution are carried out. The dependance of the Co (II) sorption (S) as a function of contacting time, the dependance of distribution coefficient (Kd) on the pH, the concentration of humic acids (HA) and temperature was established. Within the framework of the surface complexation model, the model of distribution of Co_2^+, $CoAc^+$, $CoCl^+$, $Co(OH)^+$, $Co(OH)_2$, $Co(OH)_3^-$ complexes between the solution and the sorbent was analyzed. The found stability constants of the $Co(OH)^+$, $Co(OH)_2$ complexes coincide with thermodynamic quantities. In the presence of HA (10.0 mg/l), the best coincidence of the model dependence Kd - pH with the experimental data corresponds to the sorption of the complex of the composition Co(OH)A0, A - is an anion of humic acid. Kinetic analysis of sorption isotherms performed by the batch sorption with solution recirculation showed that the transfer of Co(II) ions to the sorbent corresponds to the mixed-diffusion model. It includes at least two stages of film and gel diffusion in macro-micropores of the sorbent grain. The velocity coefficients of film (b, m/s) and gel diffusion (Dg, m^2/s) do not depend on the pH of the solution, and the concentration of HA. The average value of Dg is (6.0±4.5)·10-10, m^2/s, the activation energy of gel diffusion (20.7±10.0) kJ/mol at a pH of 7.5. The values found are close to the parameters of Co_2^+ self-diffusion in ultra-dilute aqueous solutions. The results showed the possibility of effective extraction of Co(II) from natural aqueous solutions by the sorbent Thermoxide 35 in the presence of HA.

ID: 0884

STUDIES IN DUAL DOPING OF URANIUM NITRIDE FOR USE AS ADVANCE TECHNOLOGY FUELS

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Uranium Nitride (UN) is a potential candidate for the advanced technology fuels (ATF) concept, as it has numerous benefits compared to conventional uranium oxide (UO_2) used today. Namely, higher thermal conductivity and uranium density. UN main drawback is its poor corrosion resistance in oxidizing environments. Doping of UN with oxide scale forming elements such as chromium could improve the resistance towards oxidation. In this work, the addition of chromium and aluminum in the UN matrix was accomplished using a sol-gel method to obtain a homogenous mixture of the metals. The effect caused by the introduction of these dopants in the properties of the UN material was studied. It was observed that addition of aluminum caused a change in the microstructure of the spheres. These changes are suspected to be responsible for the decrease in the reaction temperatures needed to oxidize the UN with air during the thermogravimetric analysis. Chromium content appeared to decrease during the synthesis of UN. Nonetheless, oxidation temperatures were increased only when chromium alone was added as dopant.

ID: 0888

DISSOLUTION OF IRRADIATED MOX FUEL WITH HIGH Pu CONTENT

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The use of MOx with high plutonium contents as a fast reactor fuel is under consideration to help manage plutonium inventory. These plutonium contents are above those typically reprocessing and posse a number of challenges. Increased plutonium content makes the fuel dissolve more slowly and leads to an increase in plutonium-rich residues. Increasing the dissolver cycle time to allow for the slower dissolution kinetics will impact upon reprocessing throughputs. In this study dissolution experiments on irradiated MOx fuel containing high amount of plutonium was done, the irradiation experiments had been conducted in the High Flux Reactor in Petten, the Netherlands and aimed to assess the irradiation behaviour of high Pu content mixed oxide fuels, meant for operation in fast reactors. The MOx fuel had been fabricated by hybrid sol-gel method, all pellets have annular form, with Pu content Pu/(Pu+U) = 0.45. The fuel has been irradiated for 74.1 effective full power days. The samples made available for the dissolution study are slices of about 2 mm thickness with a weight of fuel about 0.37 g each. Due to its high plutonium content, the fuel was expected to dissolve slowly with high residues poorly in standard nitric acid (PUREX process) dissolution steps. Therefore, a series of dissolution experiments with varying conditions were proposed:

- Dissolution in hot concentrated nitric acid,
- Dissolution in hot concentrated nitric acid with fluoride as a catalyst and
- Oxidation with in-situ chemically generated Ag(II).

All methods have been applied and the samples have been dissolved. During the dissolution experiments sub-samples have been taken and analysed for U and Pu content using TIMS method to get information on dissolution rate.

ID: 0904
EXPERIMENTAL ACTIVATION DETERMINATION IN AND ON COMPONENTS OF NUCLEAR POWER PLANTS AND COMPARISON WITH ACTIVITY CALCULATIONS

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Due to the “German Energiewende”, all nuclear power plants (NPPs) in Germany will have been shut down by the end of 2022. Consequently, a safe, economical, and efficient dismantling of the NPPs will be an important challenge for the next decades. This includes to progress with methods for optimal planning and implementation of decommissioning. Several studies have been conducted to develop a standardized method for evaluating the specific and temporal progression of the activation in the reactor components, near-reactor concrete, and construction elements based on the reactor's power history. This will serve as an early non-destructive tool for the radiological characterization of the NPP's components. Such essential knowledge can significantly minimize the radioactive waste and the radiation exposure of the operating personnel during the NPP's decommissioning. The studies considered two strategies. In the first one, the radionuclide inventory in the material of an NPP already under dismantling was investigated. Among others, ^{60}Co and ^{14}C in steel samples from the reactor pressure vessel and ^{152}Eu and ^{133}Ba in concrete drill cores from biological shielding were determined. In the second strategy, the neutron flux in NPPs still under operation was determined with activation monitors (small metal foils, e.g., In, Sn, Zn). The experimental data from both strategies were compared with results from Monte-Carlo simulations.

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ID: 0906
CHARACTERISATION OF CONTAMINATED STAINLESS STEEL FROM ACIDIC REPROCESSING ENVIRONMENTS

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The Thermal Oxide Reprocessing Plant (THORP) at Sellafield has reached its end-of-life and is about to be decommissioned. One of the first steps to reducing the hazards associated with the plant is Post-Operational Clean Out (POCO). As part of this process, a large amount of radioactive metallic waste is required to be managed through the waste hierarchy, with one of the key stages being decontamination. One of the major components of this metallic waste is stainless steel. Work has been ongoing to identify the extent, nature and depth of contamination of stainless steel by various problematic radioisotopes present in hazardous aqueous waste streams. Developing our understanding of the mechanisms by which contaminants interact with stainless steel surfaces is key to the development of tailored decontamination approaches, which can address challenges associated with waste management, such as volume minimisation. Cost savings upwards of £500 million are possible through re-categorisation of the UK’s current stainless steel waste inventory. Simulation of contamination is an important avenue of investigation as analyses of plant samples pose a number of challenges. Our work has targeted conditions similar to those observed in a number of processes at THORP, which involve dissolution of components in nitric acid. Work has explored how concentrated nitric acid at elevated temperatures affects contamination of stainless steel by high yield fission products over extended time periods. Solution analysis, as well as characterisation of stainless steel coupons has been undertaken. Uptake kinetics are fast, with equilibrium being achieved within two weeks. Effects of corrosion are significant, as observed through Scanning Electron Microscopy, with significant levels of intergranular corrosion and grain dropping. Non-destructive surface-based analysis techniques, such as Laser Induced Breakdown Spectroscopy (LIBS) and X-ray Photoelectron Spectroscopy (XPS) have been employed to identify the presence of contamination in the very thin oxide layers (< 10 nm), as well as potential binding mechanisms. Depth profiling through Glow Discharge Optical Emission Spectroscopy (GD-OES) has identified the presence of contaminants in the bulk substrate, due to diffusion down corroded grain boundaries. With this knowledge, decontamination strategies tailored to minimise both primary and secondary waste streams, subsequently minimising the associated economic and environmental burdens.

ID: 0913
EFFECT OF BACKGROUND ELECTROLYTE COMPOSITION ON THE FORMATION OF TH(IV) NANOPARTICLES ON THE MUSCOVITE (001) BASAL PLANE

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Actinides are known to form nanoparticles (NP), which were shown to enhance radionuclide transport in the environment. Understanding these processes on the molecular level is therefore of particular interest. Previous results showed a strong and unusual influence of the background electrolyte on Th sorption on the mica (001) basal plane based on surface x-ray diffraction (SXD) data. Uptake was shown to be below the detection limit for $NaClO_4$ solution but not NaCl (0.4 Th/AUC). Additionally an exceptional high coverage was detected for $LiClO_4$ (4.9 Th/AUC) and surprisingly intermediate sorption occurs for $KClO_4$ (~0.1 Th/AUC) under otherwise identical solution conditions. We combined Surface X-ray Diffraction (SXRD) and in situ AFM to investigate this previously reported unusual electrolyte effects on Th uptake on mica for a broader range of electrolytes and two different Th concentrations. At low [Th] (0.1 mM), interfacial structures show a broad Th electron density (~50 Å). A linear decrease of Th uptake with decreasing hydration enthalpy of the electrolyte cation (Li^+, K^+, NH_4^+, Cs^+) indicates a competitive effect between Th and the electrolyte cation. Na^+ is a clear outlier from this trend, with significantly lower Th uptake. In situ AFM imaging confirms the results. Particles show a vertical size of ~1 – 2 nm and larger lateral dimensions of ~10 – 20 nm, which is typical for particles formed at interfaces by a heterogeneous nucleation mechanism. At high [Th] = 1 and 3 mM, all investigated electrolytes (ACl, A = Li^+, Na^+, K^+) show similar Th uptake, indicating a much smaller impact of electrolyte composition. The interfacial structures are dominated by a high Th loading at a distinct distance (~6.5 Å) from the surface. Therefore, the main retention mechanism at high [Th] is suggested to be the sorption of Th NPs aggregated from Th oligomers present in solution via homogeneous nucleation. Preliminary investigations suggest a connection of the observed behavior to the complexation behavior of nitrate, which was present in our solutions in low concentrations due to the use of $Th(NO_3)_4$ as a starting material.

ID: 0921
SCREENING OF MODIFIED DIGLYCOLAMIDE DIASTEREOMERS TO INCREASE AM(III) SELECTIVITY DURING THE PARTITIONING OF PUREX RAFFINATE SOLUTION

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Multiple strategies have been developed to separate lanthanides (Ln) and actinides (An) from Spent Nuclear Fuel (SNF) and PUREX raffinate through solvent extraction. Current research is focusing on the development of new ligands to separate Am(III) from Cm(III) and other fission Ln(III). Diglycolamides (DGA) such as N,N,N′,N′-tetra-n-octyl diglycolamide (TODGA) are promising extractants for this task. Recently, methyl substitutions in the backbone of TODGA with different orientation were studied, and interesting effects on the selectivity for Am(III) over Cm(III) and other Ln(III) were observed.[1] The orientation on the alkyl groups has shown different behavior reflected on the affinity for Am(III) and efficiency of the system. Here, we study the selectivity and affinity of two related extractants (TODGA and N,N,N′,N′-tetra-n-decyl diglycolamide, TDDGA) with dipropyl and ethyl-methyl substitutions in the backbone, and their respective diastereomers (syn or anti 0.1 M in n-dodecane). An inverse selectivity is observed for Am(III) over Cm(III) and other Ln(III) at high nitric acid concentration (7 - 10 M). The steric hindrance gives low distribution ratios D < 0.5 for trivalent metal ions. Pu (IV) was also tested for the interest in the complexation behavior with tetravalent ions. Pu(IV) shows high distribution values at the same HNO_3 concentration (D > 10). The separation factors are inversely proportional to the HNO_3 concentration. The syn-ethyl-methyl substitutions for both analogues showed the highest D values for Am(III) among the other ligands, giving a promising molecule for further studies. The results of the extraction studies will be presented and discussed in the poster.

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ID: 0928

PAW PROCESS: ASSESSING LIPOPHILICITY IN MIXTURES OF CONTAMINANTS AND RADIOACTIVE WASTE

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The management of radioactive waste requires an accurate description of source termsand their behavior in natural or engineered barriers. Many radioactive waste involve organiccontaminants, either included in the source term (e.g. ^{14}C bearing lixiviates of graphite waste) or released simultaneously with radionuclides (e.g. degradation products of plastics oradjuvents in cementitious materials). The migration of organic compounds is largelyinvestigated in cementitious and geological environments, as well as their effect onradionuclides confinement[1]. This behavior strongly depends on the lipophilicity ofcompounds, even for ionized species[2]. This property is therefore an interesting parameterto quantify, useful for further performance assessments. However, some hazardous waste anddegradation mixtures are sometimes too complex to allow an exhaustive characterization oforganic compounds and their lipophilicity. In this context, a new process "Partition of AqueousWaste" (Paw) was developed. It quantifies and classifies organic compounds from unknownmixtures regarding their lipophilicity. The process uses sequential liquid-liquid partitionequilibria. Its experimental validation will be illustrated with different kinds of analyses, e.g.organic carbon, UV-Vis, ^{14}C. Using a specific mathematical model, a "lipophilic" profile of thesample is obtained. Hence, the Paw process provides insights on the constituting solutes of themixtures and their environmental behavior. Such data may be relevant for performance assessments in the context of radioactive waste storage.[1]

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ID: 0934

COPRECIPITATION OF THE ACTINIDES Th(IV), U(IV) AND ACTINIDE HOMOLOGUES Y(III), La(III), CE(III) AND Yb(III) IN SECONDARY MINERALS FROM CRYSTALLINE FRACTURE SYSTEMS: A NATURAL ANALOGUE STUDY FROM FORSMARK, SIMPEVARP AND LAXEMAR (SWEDEN)

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As studies on the retention of radionuclides in natural systems over geological times are not feasible, it is common practice to resort to natural analogues. The coprecipitated actinides and homologues of the equivalent oxidation state in secondary mineral phases are a good approximation to estimate the radionuclide retention potential of such minerals. Fracture fillings formed within crystalline host rock, as e.g. the calcites studied in this work, preserve the element content of the precipitation solution. The specific element composition of single growth zones thus allows conclusions to be drawn about the evolution of the precipitation solution and the precipitation conditions. Fe for example is sensitive to changes in redox conditions and was found in elevated concentrations in connection with paleo-biofilms[1]. Sr contents on the other hand variate between e.g. meteoric, marine, glacial and hydrothermal fluids, hence are specific to the origin. To obtain highly spatially resolved information at ppm–mg range, we applied 2D element analysis (µXRF and LA-ICP-MS) to scan for the trace elements Na, Mn, Fe, Sr, Y, La, Ce, Yb, Pb, Th and U accumulated in low-temperature calcite fracture fillings handpicked from exploration drill cores at Forsmark, Simpevarp and Laxemar (Sweden)[2]. Based on the element maps, we found several growth zones and other microstructures (e.g. point accumulations, microfractures, gradual element enrichment) within the fracture fillings. Particular structures from the Laxemar area (ca. 400-430 core meters) indicate paleo-biofilms along which extremely high trace and rare earth element concentrations were observed. Further line scans with an extended element spectrum were performed to illustrate the high retention potential of such biogenic structures. At one sample from the Simpevarp site (ca. 200 core meters) face depending partitioning within a single growth zone could be visualized. It occurred in three clusters with major enrichment in Mn-Fe-Sr, Y-Yb-U and Na-La-Ce-Pb-Th. Furthermore, partition coefficient DTE based on the element assemblage of the most recent overgrowth and connected formation waters were determined. These ratios were compared to experimentally generated coefficients[3] and values obtained by medium-term precipitation experiment conducted at the Äspö Hard Rock Laboratory[4].

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ID: 0938

BARIUM REACTIONS ON CALCITE-WATER INTERFACE

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High-level radioactive waste (HLW) and spent nuclear fuel (SNF) are planned to be disposed of in many countries in deep-lying geological multi-barrier systems. Groundwater intrusion into deep geological repositories represents a considerable risk of long-lived radionuclides being released from the deposited HLW and SNF. Therefore, the risk assessment of such a scenario plays an integral role in current research. Furthermore, flowing groundwater in fractures will be the primary medium for radionuclides transport into the surface environment in case of their release from the nuclear waste repository. Thus, the primary solid/liquid interface reactions, such as sorption and co-precipitation on mineral surfaces, will affect the mobilization of radioactive contaminants from deep geological storage systems. In addition, groundwaters can consist of significant concentrations of dissolved compounds, for example, relatively high concentrations of carbonates or sulphates that affect the behavior of the radioactive contaminants. At the same time, relatively high salinity was also discovered. Gamma emitter ^{133}Ba was used in our experiments as a chemical analogue to investigate ^{226}Ra behavior since they have similar properties and behavior. ^{226}Ra is a significant hazardous element in radioactive waste disposal due to its comparatively long half-life (1600 years), high radiotoxicity, and high solubility, and it is one of the contaminants in groundwater systems. It is also presented in the petrochemical, mining, and geothermal industries[1]. This research aimed to examine theoretically and experimentally the barium sorption and co-precipitation processes in model waters containing different concentrations of carbonates, sulphates, and calcium at pH 8 under saline and anoxic conditions. In addition, barium reactions on the calcite-water interface were studied by the batch sorption method. The activity of ^{133}Ba a was measured with an HPGe detector. The SEM and zeta sizer-nano was used to examine the morphology and size of precipitates. We discovered significant removal of ^{133}Ba from the model waters, notably in sulphate pre-concentrated solutions. An interaction with calcite provided the effect of the solid phase on the model water system. Moreover, a geochemical model PHREEQC was used to assess the reaction mechanism of barium on calcite.

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ID: 0939

HLW EVAPORATION TECHNOLOGY WHICH INCLUDES DECOMPOSITION OF BOTTOM SOLUTION COMPONENTS USING REDUCTANTS AS APPLIED TO THE HIGHLY BURNT-UP SPENT NUCLEAR FUEL REPROCESSING

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Introduction Evaporation of tail solutions from spent nuclear fuel reprocessing is a required activity before high-level waste (HLW) solidification. Evaporation of HLW according to the traditional flow sheets of the 2nd generation reprocessing plants is complicated by an increase of the sediment-forming fission products amount due to a 100-fold increase in SNF burnup, as well as the presence of fluoride ion in rafinates. Decomposition of bottom solution components using reductants during evaporationTo solve the problems of precipitation, a process of highly active rafinate evaporation with continuous decomposition of nitric acid was developed and laboratory testing in the evaporator with external heating chamber and natural circulation of bottoms was carried out. The decomposition of nitric acid during evaporation was carried out when a solution containing a reductant is supplied to the evaporator bottom part, the process was carried out when the solution is retained in the bottom part. Mixture of formaldehyde and formic acid was used as a reducing agent, or a solution of formic acid is used after the start of the process using a mixture of formaldehyde and formic acid. The composition of the denitrating reagent was determined, which ensures the minimization of N_2O formation during the evaporation process with simultaneous denitration. A technological flow sheet was developed for

HLW concentrating with the purification of nitric acid from fluoride ion at the rectification stage by distillation into the vapor phase and localization at the alkaline absorption stage. The main results on the HLW model solutions evaporation with denitration achieved at the laboratory bench were reproduced at the SverdNIIKhimmash JSC full-scale bench facility. As a result of distillation purification, regenerated nitric acid with the concentration of 11 to 12 mol/L and with its purification from fluoride ion from 6 to 7.5 times was obtained. Verification of a previously developed mathematical model of rectification with purification of nitric acid from hydrofluoric acid was also carried out. The data obtained as a result of a bench test formed the basis for creating a mathematical model of the process for the evaporation of HLW with the continuous decomposition of nitric acid. Conclusion HLW evaporation technology which includes decomposition of bottom solution components using reductants as applied to the highly burnt-up spent nuclear fuel reprocessing was successfully devel-oped and tested at the SverdNIIKhimmash JSC full-scale bench facility.

ID: 0940
SORPTION OF NICKEL ON CALCITE BASED ON SURFACE CHEMISTRY STUDIES

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Calcite is a common mineral in the environment, comprising approximately 4 % of the earth's crust. The surface reactions of calcite play an important role in many geological and environmental systems, including the production of oil and gas, the geological storage of nuclear waste and CO_2 etc. For example, the sorption of ^{63}Ni on calcite affects significantly its mobility and removal from aqueous phases. ^{63}Ni is one of the most important radionuclides that will determine the radiation risk of spent nuclear fuel for the first several hundred years of the final disposal. A clear understanding of the interactions between ^{63}Ni and calcite surfaces is vital for the safety assessment of a spent nuclear fuel repository. However, despite numerous studies that have been performed, the fundamental parameters of calcite surface charge in aqueous solutions, especially at conditions relevant to natural systems, remain poorly understood. The calcite surface composition and charge are linked to its reactivity and sorption properties, and thus, they are the primary data source to understand the sorption properties of ^{63}Ni on calcite surfaces. In this work, the surface properties of Bukov calcite are studied by a combination of experimental and basic-stern surface complexation modelling methods. The surface properties and cross-sections of crushed calcite samples are characterized experimentally by cation exchange capacity (CEC), B.E.T. (Kr) for specific surface area (SSA), scanning electron microscope (SEM) and zeta potential measurements (ZP). The structural and surface characterization measurements will answer several uncertainties associated with understanding calcite surface structures and dissolutions, including the dissolution kinetics, surface charges, the contribution of grain boundaries, potential determining ions etc. The next step in the work is to develop a basic Stern surface complexation model together with our colleagues in Gesellschaft für Anlagen- und Reaktorsicherheit (GRS), Germany, to provide data for the surface species and the Stern layer thickness. With the understanding of the surface properties, three sets of sorption experiments are performed. The titration experiments are carried out in a batch-wise manner to measure the calcite surface site capacities and intrinsic surface protonation and deprotonation constants. A backtitration approach is performed to compensate for the effect of dissolution. Sorption edge experiments are carried out by measuring the sorption of 10^{-9} M ^{63}Ni on calcite from pH 7 to 11. Sorption isotherm experiments are carried out at pH around 8 with Ni concentrations from 10^{-9} M and 10^{-3} M. A multi-site surface complexation model of ^{63}Ni sorption on Bukov calcite is used to provide explanation and prediction of ^{63}Ni sorption behaviour under various conditions. The model is based on the above experimental data (titration data, sorption edge data and sorption isotherm data) and molecular DFT modelling which will provide basic model parameters like surface site types and surface site densities.

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ID: 0944
PLUTONIUM LOSSES REDUCTION DURING THE SEPARATION OF PRECIPITATING ELEMENTS FROM SNF SOLUTIONS BEFORE EXTRACTION PROCESSING

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IntroductionWhen dissolving highly burnt spent nuclear fuel (SNF) with obtaining of solutions containing heavy metals (HM) more than 300 g/L, there is a high probability of secondary precipitation, the composition of precipitants depends on the degree of nuclear fuel burnup and the conditions of its dissolution. When a zirconium molybdate precipitate is formed, a certain amount of plutonium is included in its composition. The amount of captured plutonium can exceed the allowable losses of nuclear materials (NM) and increase the radioactive waste class. Plutonium losses reduction. When handling sediments containing molybdenum, you can use the operation to decompose zirconium molybdate in an oxidizing agent - hydrogen peroxide. For the complete extraction of plutonium, the sediment should be treated in this way at least three times, the residual plutonium content in the sedi-ment being 0.01 %. Leaching of plutonium (IV) from the zirconium molybdate precipitate with a solution of electrogenerated silver (II) does not lead to the extraction of plutonium from the precipitate. A method was developed to reduce the amount of sediment-forming elements (molybdenum and zirco-nium) in SNF solutions by preliminary electrochemical oxidation of Pu(IV) to Pu(VI) in the presence of silver (II). Reduction of NM (plutonium) losses with the zirconium molybdate precipitate is achieved by preliminary electrochemical oxidation of Pu(IV) to Pu(VI) in the presence of silver (II), followed by the precipitation of zirconium molybdate precipitate with continued electrochemical oxidation. This method was tested on an installation designed to dissolve plutonium dioxide, located in one radiation-shielding chamber at the radiochemical plant of JSC SKhK using gram quantities of NM. A method that allows reducing the amount of sediment-forming elements (molybdenum and zirconium) in SNF solutions by preliminary electrochemical oxidation of Pu (IV) to Pu (VI) in the presence of silver (II) was successfully developed.

ID: 0949
Tc(VII) REDUCTIVE IMMOBILIZATION BY Sn(II) PRE-SORBED ON ALUMINA NANOPARTICLES.

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The interaction of highly mobile radioactive elements in the spent fuel with the different technical and geological barriers of a nuclear waste repository needs quantification and mechanistic understanding to allow a reliable safety assessment. One of the most concerning mobile fission products is ^{99}Tc. It is a long-lived radionuclide (half-life of 0.213 million years) that is expected to occur as Tc(VII) under oxidizing conditions and as Tc(IV) under reducing conditions. The anion pertechnetate (TcO_4^-) is the main species of Tc(VII) and it is known to be a highly mobile species since it barely interacts with mineral surfaces. On the contrary, TcO_2 is the main species of Tc(IV) and it is a hardly soluble solid. Therefore, the reduction of Tc(VII) to Tc(IV) limits the mobility of Tc in water and is triggered by reducing agents such as Fe(II) or Sn(II).[1] In a previous work, we have observed that pre-sorption of Fe(II) on alumina enabled the Tc(VII) reduction at the interface, even at low pH values when Tc(VII) reduction by Fe(II) was expected to be limited due to the low sorption of Fe(II) on alumina.[2] In this study we focus on the impact of Sn(II). We have performed sorption experiments following a stepwise strategy to ensure that Tc(VII) reduction by Sn(II) occurred at the interface (heteroreduction).

- Sn(II) was sorbed on alumina,
- the Sn(II) pre-sorbed on alumina solid was isolated and dried,
- a solution of Tc(VII) was added to this modified alumina, and
- the yield of Tc removal by Sn(II) pre-sorbed on alumina was analyzed

The resulting Tc-containing solid was analyzed by X-ray absorption spectroscopy (XAS) at the Rossendorf Beamline (ROBL) at the European Synchrotron Radiation Facility in Grenoble (France). Re-oxidation experiments were performed in samples where Tc(VII) reduction by Sn(II) was obtained by different pathways:

- Tc(VII) direct reduction by dissolved Sn(II) (homoreduction) and
- Tc(VII) reduction by Sn(II) pre-sorbed on alumina (heteroreduction)

The results show that Tc(VII) is reduced to Tc(IV) with a high yield (85-100 % removal from solution), being maximum at pH values between 3.5 and 9.5, and minimum at pH 10. Reoxidation studies show that Tc(IV) obtained by heteroreduction presents lower oxidation kinetics than Tc(IV) obtained by homoreduction. These results support that the presence of alumina plays an important role by preventing Tc(IV) reoxidation.

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ID: 0950

CHEMICAL BEHAVIOR OF LONG-LIVED FISSION AND ACTIVATION PRODUCTS IN THE NEAR FIELD OF A NUCLEAR WASTE REPOSITORY AND THE POSSIBILITIES OF THEIR RETENTION (VESPA II)

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To ensure a reliable and long-term safety assessment of high-level radioactive waste disposal, it is essential to study the physico-chemical properties of the radionuclides within spent nuclear fuel as well as their transport behavior expected under conditions of the near- and far-field of a nuclear waste repository. Among the radionuclide inventory, long-lived mobile fission products are of high concern since they can strongly contribute to the total biosphere dose from spent nuclear fuel disposal. The collaborative project VESPA "Chemical Behavior of Long-Lived Fission and Activation Products in the Near Field of a Nuclear Waste Repository and the Possibilities of their Retention - Phase II" aims to investigate the solubility and the immobilization of ^{99}Tc, ^{129}I, ^{137}Cs, and ^{79}Se. In particular, the focus is set on (1) the source term, evaluating, e.g., the ^{129}I inventory together with the instant release fraction and its speciation; (2) the effect of geochemical conditions in the near-field, i.e. T, p, Eh, pH, on the processes of surface redox-mediation and secondary mineral phase formation; (3) the solution chemistry, determining solubility products, complex formation and activity coefficients of Tc(IV) in presence of anions and small organic molecules, and Se(IV), Se(0), Cs(I) and I(-I) at elevated temperature; and (4) the retention behavior of I, Se and Tc on layered double hydroxides (LDH) and Fe-corrosion products. Finally, safety analysis calculations link the obtained results and provide an enhanced confidence in predictive risk assessments.

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ID: 0958

Gd-DOPED UO_2 SURFACE OXIDATION BY H_2O(G) IN THE PRESENCE OF H_2(G) AT DIFFERENT TEMPERATURES

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To improve the efficiency of nuclear reactors by extending the fuel cycle, burnable neutron absorbers, such as Gd_2O_3, have been extensively added in the PWR and BWR fuels to control the reactivity due to their large neutron absorption cross-section. Doped fuels must be safely stored after their life cycle in a deep geological repository, therefore their behaviour in case of canister failure must be examined. To assess the safety of a deep geological repository, redox conditions of the spent nuclear fuel are of a great main concern because of the more soluble U(VI) compared to U(IV), which would result of significant release of radionuclides to the environment. Such conditions depend on the formation of both oxidizing and reducing species. Water vapour has a significant impact on the oxidation of uranium dioxide and might be present in the near-field due to residual water coming from the cooling pools or groundwater evaporation. Conversely, reducing species such as molecular hydrogen could be present because of the anoxic corrosion of the metallic canister. However, there is no information regarding the interaction among Gd-doped UO_2, water vapour and hydrogen. For this reason, in this work, the effect of hydrogen stream saturated with water vapour on the surface oxidation of Gd-doped UO_2 at different temperatures was studied. In situ experiments were conducted in a high pressure chamber (HPC) connected to an X-ray Photoelectron Spectroscopy (XPS) analysis chamber. Hydrogen stream was saturated with water vapour at room temperature and introduced into the HPC, where the sample was located. Then, the temperature of the chamber was increased to the desired value and maintained for a specific time. Prior to perform any experiment, a reducing treatment with only H_2 at high temperature was conducted to completely reduce the sample surface. The XPS spectra obtained after each experiment were used to determine the uranium oxidation state in the surface by the de-convolution of the U_4F_7 band into U(IV), U(V) and U(VI) contributions. Preliminary results show that the surface of Gd-doped UO_2

sample was less oxidized than previous studies conducted under similar conditions on non-doped samples, where UO_2 was found to be oxidized in the hydrogen/water vapor experiments. The addition of gadolinium oxide to uranium oxide could strongly reduce the water oxidizing effect on the UO_2 matrix in the presence of $H_2(g)$.

ID: 0961

MICROBIOLOGICALLY INFLUENCED CORROSION OF CAST IRON CONTAINERS FOR HLW STORAGE

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Ductile and corrosion resistant cast iron is investigated as a potential container material to store high-level nuclear waste (HLW) in deep geological repositories (DGR) in claystone bedrock. The dynamic corrosion process is dependent on the conditions present in the DGR which are influenced and/or controlled by geochemical parameters (e.g., redox potential, pH, presence of and ionic concentration in (pore-)water), physical parameters (e.g., pressure), and the influence of metabolically active microorganisms. Cast iron corrosion will occur at the intersection of container and its decontaminable coating with the bentonite backfill material which contains natural microbial populations. The conditions in a DGR are simulated in microcosm experiments to investigate the impact of microbiologically influenced corrosion (MIC); the microcosms contain: B_{27} bentonite, synthetic pore water, N_2 or N_2-CO_2 atmosphere, cast iron coupons, as well as the bacterium Desulfosporosinus burensis (isolated from repository depth in Buré, France). Three coupon configurations will be used: untreated, coated with decontaminable coating, and coated with decontaminable coating which has been damaged to simulate possible damages. The microcosms will be examined for bio- and geochemical parameters, such as pH, redox potential, mineral phases, sulphate concentration, Fe(II):Fe(III), changes in microbial populations, and the corrosion process for formation of corrosion products, and potential microbial influence, after a 270-day incubation period at 25 °C under anaerobic conditions. In subsequent experiments, the sorption behavior of lanthanides and actinides onto the membranes of viable cells and spores of D. burensis, as well as the surface of corroded cast iron coupons will be investigated.

ID: 0964

INFLUENCE OF SURFACE NANOTOPOGRAPHY ON THE ADSORPTION OF EUROPIUM ON MUSCOVITE (001)

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Radionuclide migration is one of the key problems for the long-term safety of nuclear waste repositories. One possible mechanism to retard or prevent the migration of radionuclides from the repository to the biosphere is the adsorption onto mineral surfaces of the surrounding host rock. Clay rock formations such as the Opalinus Clay are being considered for potential sites for nuclear waste repositories, partly due to the strong sorption potential of clay minerals. Phyllosilicates, such as clay minerals or mica, have shown a high affinity for adsorption of various radionuclides in several experimental studies. However, mineral surfaces in natural environments are often subjected to reactions (e.g., dissolution) that may alter the surface nanotopography and, consequently, affect the overall adsorption process. Recently, it has been reported that the nanotopography of calcite surfaces leads to heterogonous sorption of europium due to differences in the atomic configuration of the adsorption sites[1]. In this study, we investigate the influence of surface site coordination on the adsorption energy barrier and the resulting overall distribution of radionuclide adsorption on the mineral surface. We utilize numerical methods to study the adsorption of $Eu(OH)_3$ on a muscovite surface with different nanotopographic structures. Density Functional Theory (DFT) calculations are performed to obtain the adsorption energy barriers of several surface sites present on muscovite. For each site, the adsorption energy is calculated based on a series of geometry optimizations with increasing Eu–site distance. The values of the site-specific adsorption energy barriers are then implemented in a Kinetic Monte Carlo (KMC) model based on a previous study[2]. In the KMC model, larger surface structures, such as steps or etch pits, are placed on the muscovite surface and a dissolution simulation is performed to create a realistic nanotopography. Based on the adsorption energy barriers obtained with DFT, $Eu(OH)_3$ is adsorbed on the generated muscovite surface in a second KMC model step. The KMC model is then used to predict the distribution of adsorbed $Eu(OH)_3$ and the temporal evolution of the adsorption. Using this combined numerical approach, we show the effect of surface site coordination on radionuclide adsorption reactions and the resulting adsorption heterogeneity on mineral surfaces at large scales.

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ID: 0965

THE PRODUCTS OF RADIOLYSIS AND HYDROLYSIS OF POLYACRYLONITRILE

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The products of radiolysis and hydrolysis of polyacrylonitrile (PAN) in strongly alkaline conditions were studied using several methods — HRMS, FT-IR and LCMS analysis, combinable HPLC with UV / VIS and refractometry. The studied conditions correspond to the environment formed by cementitious materials utilized as the engineering barrier in the disposals of radioactive waste. Degradation products from materials such as PAN, which are part of the waste, can significantly affect the migration of radionuclides present. The data obtained from infrared spectroscopy show the common trends: in the region of 3400-3200 cm^{-1} bands are visible, which correspond to the valence N-H vibrations of the amine or amide functional group. Furthermore, bands of valence symmetric and antisymmetric CH_2 and CH_3 groups are visible in the region 2950-2850 cm^{-1}. The band around 2240 cm^{-1} corresponds to the valence vibration of the nitrile group ν (C≡N). The degradation proceeds via intermediates (-C = N) and amide ($-CONH_2$) and leads to the formation of a carboxyl functional group (-COOH). The presence of intermediates in the reaction mixture is manifested by characteristic bands of vibrations in the infrared spectrum of the samples. The presence of PAN fragments after hydrolysis was also confirmed by MS spectra.

ID: 0966

SORPTION OF URANIUM AND LEAD ON CSH

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To evaluate the long-term safety of low/intermediate level radioactive waste repositories and future deep geological repositories for spent nuclear fuel, it is necessary to investigate the behaviour of radionuclides that will be present, such as uranium or lead. Radionuclide migration is affected by interactions with engineering barriers of the repositories, which may also include sorption of radionuclides on the cementitious materials. In this study, the sorption of selected elements on cement phase CSH (Calcium-Silicate-Hydrate, main sorbing phase in cements) was carried out within different conditions (temperature, addition of EDTA, as a selected representative of the organic substances present in the waste). The sorption behaviour of element is described by the distribution ratio (R_d) between liquid and solid phase (L/S) or with sorption isotherm. Sorption of U and Pb on synthetic cement phase CSH with Ca/Si ratio 1 was studied in a set of equilibrium and kinetic sorption experiments held within range of L/S = 100–800 L kg^{-1} and temperature range 22-80 °C with ^{233}U and stable Pb in an inert atmosphere. The aim of the study was to compare behaviour of these elements with previous study which was done on real cementitious materials and describe the sorption from several point of view: distribution ratio or the isotherm shape under the influence of temperature or addition of EDTA. The measurement of ^{233}U was done on LSC detector. Pb concentration in solution was measured on AAS. Where possible with regard to experimental setup reaction enthalpy, entropy and apparent activation energy were also determined. Sorption experiments with a synthesised CSH phase in comparison with real cementitious material within several conditions confirmed the dominant role of CSH for sorption of selected radionuclides in cementitious materials corresponding with the higher R_d values of sorption on CSH than on real cementitious materials. Sorption of uranium and lead is strong with high R_d values (in the order about 10^5 L kg^{-1}). Pb sorption is more sensitive to presence of EDTA.

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ID: 0968
SPECTROSCOPIC INVESTIGATION OF THE COMPLEXATION OF U(VI) WITH 2-PHOSPHONOBUTANE-1,2,4-TRICARBOXYLIC ACID (PBTC) IN THE PH RANGE OF 2 TO 10

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Cement-based materials are used for waste conditioning and as technical barriers in potential nuclear waste repositories. In order to adjust the physico-chemical and mechanical properties of concretes, several additives are added to the cement. Various polycarboxylates with phosphonic acid groups, of which 2-Phosphonobutane-1,2,4-tricarboxylic acid, PBTC, is the most prominent representative, are used as modern long-term retarders for the hardening of cement. Water ingress into a repository is considered a worst-case scenario. The concrete decomposes and the cement additives can be leached out, enabling complexation with radionuclides. Therefore, the prediction and modelling of aqueous complexation reactions between actinides and various inorganic and organic ligands, such as PBTC, is of significant interest. Although PBTC is already known in industrial applications as a strong complexing agent of various met-al ions (e.g. Ca_2^+, Zn_2^+, Al_3^+, Fe_3^+), there are no studies on the complexation behaviour with radionuclides. In contrast to these well-investigated spherical ions, in case of the linear uranyl ion (UO(VI)) ligand coordination is limited to the plane perpendicular to the O=U=O axis. For this reason, the complexation of PBTC with U(VI) in the pH range from 2 to 12 in NaCl medium was investigated spectroscopically. The aim is the structural and thermodynamic characterization of possible U(VI)-PBTC complexes. Here we present some results of the structural investigations by NMR spectroscopy. First, the protonation constants of the commercially available PBTC ligand were determined by ^{13}C- and ^{31}P-NMR spectroscopy in the pH range from 0.5 to 12.5 in 0.5 m NaCl. The obtained NMR signals of the uncomplexed PBTC further served as a reference for the shift of the signals of the PBTC complexed with U(VI). This complexation was investigated with an U(VI) concentration of 50 mM and different metal-to-ligand ratios (U(VI)-to-PBTC ratio = 1:0.5 / 1:1 and 1:5) as well as a fixed ionic strength of 0.5 m NaCl. The results show, that at U(VI)-to-PBTC ratio ≥ 1:1 one U(VI)-PBTC species dominates and this is the case over the whole investigated pH range. The ^{31}P- and ^{13}C-NMR spectra indicate that U(VI) is chelated by the phosphonate group and one of the carboxyl groups. Based on the NMR results various structural proposals of this U(VI)-PBTC complex were postulated and supported using DFT calculations.

ID: 0969
COMPLEXATION STUDIES OF Eu(III) WITH NTA AT HIGH IONIC STRENGTHS

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Radionuclide speciation inside long-term radioactive waste repositories needs to be understood in order to ensure effective containment of the waste. Organic ligands originating from the degradation of organic components inside such a repository can possibly affect the mobility of radionuclides in solution. The present study focuses on nitrilotriacetic acid, NTA, as a model molecule and europium, Eu(III), as a nonradioactive analog with outstanding luminescence and magnetic properties. The complexation of NTA with Eu(III) (in ratios of 1:2 and 1:1 Eu:NTA) as a function of pH was studied using nuclear magnetic resonance (NMR) spectroscopy in 1 M NaCl D_2O solution. The ^{1}H and ^{13}C NMR spectra of the NTA solutions with Eu(III) show clearly distinguishable signals for the free NTA and two Eu-NTA complexes, which is indicative of a 1:1 and a 1:2 Eu-NTA complex. The interaction of Eu(III) with NTA is relatively strong and favors the 1:2 Eu-NTA complex even in solution containing 1:1 Eu-NTA ratio, unless in very acidic solutions. As a repository relevant cationic groundwater components, the influence of Ca(II), Al(III) on Eu(III) complexation is studied in detail. A combination of NMR spectroscopy and time-resolved laser-induced fluorescence spectroscopy yields qualitative and quantitative information on the coordination environment from the ligand's and the metal ion's perspective, respectively. In subsequent studies focusing on ternary systems comprising repository relevant solid phases, radionuclides and organic ligands this will allow the identification of radionuclide speciation in solution and their sorption to solid phases.

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ID: 0970

CHARACTERIZATION OF GRAPHITE REFLECTOR SHIELDING EMERGED FROM NPP A1

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The decommissioning NPP A1 project is reaching preparation stage of dismantling the reactor KS 150. Graphite reflector shielding, is notable part of reactor internals which must be dismantled, managed and treated. Radiological characterizations were performed in order to determine 19 limited radionuclides and ^{36}Cl for LLW repository. Material characterisation was conducted by means of EDS – SEM and differential scanning calorimetry measurements were performed for Wigner Energy investigation. The last stage of graphite characterization are encapsulation experiments with non-activated A1 graphite. Encapsulation of graphite into cementitious grouts experiments were conducted in order to select suitable encapsulations recipes. During experiments were observed rheological properties of grouts, influence of graphite loading into cementitious matrix, curing and heat generation. After selection of suitable solidification recipes were conducted encapsulation experiments which lead to product quality evaluation (compressive strength and leachability).

ID: 0971

URANIUM(VI) RETENTION BY CALCIUM ALUMINOSILICATE HYDRATES (C-A-S-H) – IMPACT OF TEMPERATURE, IONIC STRENGTH, AND ORGANIC LIGANDS

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Concepts for the safe disposal of high-level radioactive waste in deep geological formations to ensure isolation from the biosphere are based on a multi-barrier system. Cementitious materials are one component of the geotechnical barrier, used as backfill material, for borehole sealing and to enforce the mechanical stability of tunnels. Calcium silicate hydrate (C-S-H), the principal binding phase of conventional concrete, is known to provide a high sorption capacity for trivalent to hexavalent actinides[1-3]. In modern concretes, Al-containing compounds such as blast furnace slag or fly ash are added, leading to formation of calcium aluminosilicate hydrate (C-A-S-H). To systematically study the influence of Al on both the structure of C-A-S-H phases and their U(VI) retention properties in comparison to C-S-H phases, three series of samples were synthesized in the absence and presence of U(VI). They comprise samples with Ca/Si molar ratios of 0.8, 1.2 and 1.6, representing different alteration stages of concrete, and increasing Al/Si molar ratios of 0, 0.06 and 0.18 within each series. Furthermore, the impact of temperature (25 °C, 100 °C, 200 °C) on the structure of C-A-S-H phases and on the U(VI) retention mechanism was studied. Structural characteristics of the cementitious phases were obtained from powder X-ray diffraction as well as ^{27}Al and ^{29}Si solid-state magic angle spinning nuclear magnetic resonance spectroscopy. Al tetrahedra were identified to occupy bridging positions of the Si chain and cross-linking positions. Enhanced temperatures were found to increase the crystallinity of the material with the appearance of neoformed crystalline phases. U(VI) luminescence spectroscopy was applied to characterize the U(VI) binding. Several U(VI) species (surface-sorbed or C-A-S-H interlayer absorbed) are forming in different amounts, depending on the composition of the C-A-S-H phases. Finally, to study the stability (or reversibility) of the U(VI) retention by the C-A-S-H phases in the presence of (i) high ionic strength pore waters and (ii) organics originally present as admixtures in cement-based materials, leaching experiments were conducted. In these experiments, simulated pore waters of complex solution composition and solutions containing gluconate as a representative of organic ligands that may be present under repository conditions, respectively, were used. The leaching experiments were conducted over extended timescales of up to 6 months. The results showed a high retention of U(VI) in the C-A-S-H phases under both abovementioned solution conditions. In conclusion, the immobilization of U(VI) by cementitious material via sorption and/or incorporation processes has important positive implications for limiting uranium migration during geological disposal of radioactive waste.

The German Federal Ministry for Economic Affairs and Energy (BMWi) is thanked for financial support within the GRaZ II project, no. 02E11860B.

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ID: 0984
CORROSION BEHAVIOUR OF IRRADIATED MIXED OXIDE (MOX) FUELS: EFFECTS OF ENVIRONMENTAL CONDITIONS ON RADIONUCLIDE LEACHING

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In the context of spent nuclear fuel (SNF) disposition, a deep geological repository (DGR) based on a multi barrier concept is considered in many countries as the safest and most sustainable disposal option. The licensing of such a DGR is challenging and demands amongst others, safety assessments considering time frames of up to one million years. Moreover, a profound understanding of the corrosion behaviour of SNF coming into contact with groundwater, when the waste canisters are eventually breached, is crucial. During the last decades, a number of studies addressed this topic, leading to a good phenomenological understanding of the long-term behaviour of SNF in a DGR. However, these studies addressed mainly uranium dioxide-based fuels, and various processes contributing to the (radiolytic) matrix corrosion of SNF in the generally reducing repository environment are still not fully understood. Since corrosion data on irradiated mixed oxide (MOX) fuels under reducing conditions are scarce to date, the SF-ALE project (Spent Fuel Autoclave Leaching Experiments) was initiated. MOX fuel with a well-known irradiation history was characterised by optical microscopy, SEM and EPMA before leaching experiments were performed. The latter aim at elucidating the impact of the environmental conditions on SNF corrosion, addressing the instant release of radionuclides as well as the (long-term) matrix corrosion. Three cladded fuel rod segments with burn-ups ranging between 29 GWd/t_{HM} and 52 GWd/t_{HM} were selected for leaching experiments and were exposed to bicarbonate water as reference groundwater at neutral pH, and a synthetic cementitious water (pH 13.5), to address repository conditions for disposal concepts with different engineered barrier systems. Since repository conditions are expected to be reducing as a consequence of hydrogen generation due to anoxic corrosion of metallic waste canisters, the autoclave leaching experiments, which lasted for about 3.5 years, were performed under a gas phase consisting of an argon/hydrogen mixture at a pressure of 40 bar. During the leaching experiments, both the leachates and the gas phases were regularly sampled in order to determine the release of more than 30 relevant radionuclides from the irradiated MOX by various analytical techniques. Results regarding the initial release fractions of the fission products caesium and iodine observed within the first two years of leaching were published recently[1]. In this contribution, we report results on the release behaviour of further selected fission products and actinides obtained throughout the whole duration of SF-ALE.

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ID: 0986
SORPTION OF Ni ON NATURAL CALCITE

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Safety assessment of deep geological repository (DGR) of radioactive waste considers safety functions of all the barriers, including the host rock. In case of Czech DGR the host rock is considered to be crystalline rock, as granite or migmatite. Migration within such a rock is driven mainly by advection in the rock fracture which can be coated with secondary minerals, as calcite or hydrothermal clays that can possess strong sorption properties. In the case of presented study, sorption of ^{63}Ni as one of the safety relevant elements on calcite and surrounding host rock has been studied. Calcite samples were gained in Bukov URF (90 % purity). Sorption isotherm for both materials has been determined using batch sorption methodology, interaction the rock materials with synthetic granitic groundwater SGW_2 and ^{63}Ni 10-5 mol NiCl carrier. The experiments showed that calcite possess low sorption capacity Kd varying in the first tens of ml/g in comparison with the host rock. Interesting potential influence of CO_2 presence has been spotted, so as the inconsistency in geochemical databases, used for geochemical modelling of Ni speciation.

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ID: 0991

EVALUATION OF THE NEW EURO-GANEX PROCESS RESISTANCE BY GAMMA IRRADIATION TEST LOOP

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The reprocessing of spent nuclear fuel is currently moving towards advanced cycles that contemplate the recycling of minor actinides (MAs: Am, Cm, Np), as a strategy to minimise the radiotoxicity of the waste that must be stored[1]. One of the constraining points for the development of extraction processes from the point of view of safety is their resistance to the highly radioactive field and acid concentration where they must be used. For that, resistance studies of extraction systems are very useful to know and predict the long-term behaviour during a normal or mal-operation in a future reprocessing plant. Within the homogeneous strategy, which addresses the recycling of U and the transuranic elements (TRU= Np, Pu, Am, Cm) contained within a single fuel type and distributed homogeneously throughout the reactor core, GANEX (Group ActiNide EXtraction) is the most promising process to recover all of them. In the GANEX concept, bulk uranium is removed in the first cycle, followed by the co-extraction of all actinides in a second cycle. Three options exist for this second cycle CEA-GANEX, EURO-GANEX and CHALMEX processes[1]. EURO-GANEX process was tested by an irradiation loop[2] and successfully demonstrated with high plutonium content in centrifugal contactors for the first time[3], obtaining excellent results. However, EURO-GANEX also has various drawbacks: one of them is that the sulphonated BTP reagent employed in the aqueous phase does not accomplish the “CHON principle” increasing the waste volume; another one is that the combination of two extractants in the organic phase (TODGA and DMDOHEMA) complicates solvent formulation, clean up and the process itself. Therefore, the process needs to be further optimized to meet the above mentioned criteria. Improvements made to EURO-GANEX system have resulted in the emergence of the so-called New EURO-GANEX process, where the composition of the solvent has been modified by replacing TODGA and DMDOHEMA with cis-mTDDGA in the organic phase and SO_3-Ph-BTP with PyTri-Diol in the aqueous phase[4]. In this work, the Náyade irradiation loop has been configured to simulate the two main steps of the new EURO-GANEX process, i.e. An + Ln co-extraction and TRU stripping steps, under an interinstitutional study between CEA, INL and CIEMAT. For that, a high accumulated dose over the organic solvent in contact with nitric acid was supplied, simulating the recycling of the organic phase; but a relatively low dose over the aqueous phase containing PTD was supplied since it is not expected to be recycled. The performance and changes in the composition have been analysed along the irradiation experiment by different techniques: gamma spectrometry and ICP-MS for the extraction and corrosion behaviour of the full system, and HPLC-MS to determine the degradation of the organic and aqueous solvent.

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ID: 1003

Sr BEHAVIOUR FLOW-THROUGH COLUMNS REPRESENTATIVE OF NEAR-SURFACE DISPOSAL OF VERY LOW LEVEL RADIOACTIVE WASTES

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Short-lived, very low-level radioactive wastes (VLLW) will be stored in near surface repositories in many countries, including Finland, Sweden and the UK. Very low-level radioactive wastes may be produced whilst operating or decommissioning a nuclear power plant, as medical waste or as naturally occurring radioactive materials. Here, most radionuclides have half-lives less than 30 years and the burial site is designed with a multi-level barrier that will isolate the radionuclides from the surrounding biosphere for several hundreds of years. Despite careful planning, there are still uncertainties regarding the long-term performance of the barriers and therefore the long-term transport of the radioactive contaminants must be studied. Using flow-through column experiments our work assesses the mobility of a key risk-driving radionuclide (Sr), under conditions that would prevail in a Finnish near-surface repository. Specifically, we focus the potential retention mechanisms of Sr transport with respect to the evolving repository studying the effects of the bentonite barrier, steel corrosion, and microbial activity. Preliminary results suggest that Sr is well retained across all systems and the addition of bentonite increases Sr sorption capacity. Post-mortem analyses, including X-ray Absorption Spectroscopy, help elucidate the mechanisms that control Sr mobility.

ID: 1008

RADIOLYTIC AND HYDROLYTIC DEGRADATION OF A POLYCARBOXYLATE SUPERPLASTICIZER

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Superplasticizers are organic cement additives often used in construction as they improve the properties of concrete. The engineered barriers in repositories for radioactive waste often include cementitious materials. Superplasticizers and their radiolytic and hydrolytic breakdown products and how they might affect the transport properties of radionuclides represent a concern in the long-term safety assessment of radioactive waste disposal. This study presents the radiolytic and hydrolytic degradation of a commercially available polycarboxylate (PCE) superplasticizer. Superplasticizer samples were irradiated in the Jožef Stefan Institute TRIGA reactor in shutdown conditions to gamma doses up to 3.2 MGy, to identify the degradation products that could affect the mobility of the radionuclides. In addition, a hydrolytic degradation study of the superplasticizer was performed in 0.1 M NaOH. FTIR, NMR and SEC were used to study the degradation of the PCE superplasticizer. The results indicate that backbone crosslinking occurs at an accumulated dose of 0.6 – 0.8 MGy, followed by cleavage of PEG side chains at 3.2 MGy. Similarly, hydrolytic degradation showed cleavage of ester bonds.

ID: 1012

DEVELOPMENT OF TRITIUM REMOVAL TECHNOLOGY FROM LARGE AMOUNT OF CONTAMINATED WATER USING A HYBRID PROCESS.

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Tritium is a radioactive beta emission isotope of hydrogen with a mass of 3.0 and is mainly present as tritiated water(HTO) in wastewater discharged from nuclear facilities. However, since HTO and H_2O have very similar physical and chemical properties, it is very difficult to separate a trace amount of HTO mixed in a large amount of water (H_2O). There is a commercialized isotope exchange and hydrogen distillation technology that can remove tritium, but this technology is a process to remove HTO from pure water quality of D_2O, so it is impossible to apply to large amounts of contaminated water such as contaminated water in Fukushima, tritium contaminated water generated when decommissioning nuclear power plants, and contaminated environmental water. In the case of Korea, tritium was detected higher than that of residents of other regions in urine samples of residents near Wolsong Nuclear Power Plant, a heavy-water nuclear power plant, and was reported in the media. Therefore, in preparation for the generation of tritium contaminated water, it is essential to develop a technology that is larger in capacity and more efficiently capable of removing tritium than the current level of tritium removal technology. However, at the current technology level, it is very difficult to remove tritium from large amounts of contaminated water such as Fukushima contaminated water. Elim-Global Research Institute and a research team from four universities in Korea are developing different tritium removal technologies separately and developing large-capacity tritium removal technologies through a hybrid complex process based on the characteristics of the developed technology. Pre-treatment technologies consisting of MF(Micro-Filtration), UF(Ultra-Filtration), RO(Reverse Osmosis), CDI(Capacitive Deionization), and EDI(Electro Deionization) were developed and configured to operate flexibly according to the water quality of the target contaminated water. Particulate impurities and ionic impurities in contaminated water are removed through the developed pre-treatment process and only the unremoved tritium and pure water is introduced into the tritium removal device. Currently, research is being conducted to confirm the mass balance through the analysis of the tritium concentration in incoming, treated, and concentrated water during the operation of each pre-treatment process, and research on the behavior of tritium in each process. Multi-stage zeolite membranes showed 60 % of tritium removal efficiency and metal oxide & electrochemical technologies showed 30 % removal efficiency. When a hydrophilic inorganic adsorbent was used, it was confirmed that the tritium removal efficiency was 43 %, the functional ion exchange resin may be removed up to 8 %, and research for increasing the removal efficiency for each technology is continuously being conducted. In this research, in order to obtain maximized tritium removal performance, it is designed as a hybrid process in consideration of the advantages and characteristics of the four processes, and it is expected that tritium removal performance could be obtained better than a single process. In addition, a process for treating tritium concentrated water discharged from each tritium removal technology is also being developed, and a hybrid type tritium removal system with a 10 L/hr, 70 % tritium removal efficiency is planned to be developed by combining pre-treatment, tritium removal technology and concentrated water treatment technology.

ID: 1013

INVESTIGATIONS ON THE CHEMICAL BEHAVIOR OF CESIUM AND IODINE IN LEAD-BISMUTH LIQUID METAL SOLUTION

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Lead-bismuth eutectic (LBE) is a eutectic alloy composed of 44.5 % Pb and 55.5 % Bi. It is of particular interest as a possible coolant for the Generation IV nuclear reactors and accelerator-driven systems (ADS) given its good thermophysical properties (low melting point, low vapor pressure, and good thermal conductivity) and inertness towards reactions with water and air. However, the behavior of radionuclides present in the coolant, originating from the activation of the liquid metal itself and the potential contamination with fission products, are not well understood. In particular, the release of radionuclides from LBE is a key issue for its safe application and a significant challenge for the licensing of LBE-cooled reactors. In the framework of the “Horizon 2020” PASCAL project, we are interested in the radioisotopes of Cs and I, which are among the most problematic fission products. Compared to the single elements dissolved in LBE, modeling of the chemical equilibrium of the LBE/O/Cs/I system has indicated that Cs and I may show increased evaporation from LBE attributed to the formation of CsI. This thermodynamic prediction needs to be experimentally verified, since it is not evident that the chemical equilibrium required to form CsI will be established quickly. To address this question, on one side, the solubility of the CsI salt in LBE is determined to get a deeper insight into the quaternary system. On the other side, the species evaporated from I-containing LBE samples, Cs-containing LBE samples and Cs-I-containing LBE samples are investigated through thermosublimatography experiments to verify the formation of CsI in LBE.

ID: 1014

APPLICATION OF MSO TECHNOLOGY FOR RADIOACTIVE WASTE RETREATMENT

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MSO (Molten Salt Oxidation) is a technology of flame-less oxidation in molten salts. This technology is mainly used to reduce the volume of hazardous solid and liquid wastes and allows the processing of loose materials, semi-liquid suspensions or liquids over a wide range of viscosity. Combustible wastes are fed through the dosing system into the reactor together with air or oxygen. During the flame-less oxidation, which is deliberately under the level of salt melt, heavy metals and radionuclides are captured within. This paper deals with the process of combustion of an ion exchange resin containing stable Cs, Co and Sr isotopes. The research phase is focused on the dissipation of these isotopes in MSO technology.

ID: 1018

REDUCING UNCERTAINTIES IN THE MANAGEMENT OF HIGHER ACTIVITY NUCLEAR WASTES

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The United Kingdom has one of the largest and most diverse nuclear waste inventories in the world arising from Magnox, AGR, PWR and prototype reactors. In part, this reflects the country’s long history of reactor development and operation, but the issue has been compounded by frequent changes in government policy. Proposals for disposal of low (LLW) and intermediate level (ILW) waste repositories have been advanced and then withdrawn whereas significant research into high level waste (HLW) disposal has re-started only recently after a hiatus of more than 40 years, necessitated by the decision to cease fuel reprocessing. Retrieval and relocation operations for legacy fuels are imminent owing to the deterioration of storage ponds operating beyond their design lifetimes. Policy considerations notwithstanding, there are major technical challenges associated with ongoing waste storage and geological disposal. This paper will discuss attempts to address two of the main sources of uncertainty; the fate of spent nuclear fuel during prolonged storage and the validity of the ‘chemical containment concept’ in which cements perform an essential role within the disposal system by creating a favourable environment that limits the aqueous concentrations of key radionuclide species and their subsequent migration. The cost of managing legacy sites currently exceeds €4 billion per year with a guaranteed route to a final solution still elusive. Enhanced understanding of the evolution of both the waste and its environment will help mitigate the risks during interim storage and final consignment to a geological disposal facility.

ID: 1024

ACTIVATED BIOPOLYMER HYDROGEL COMPOSITES FOR STRONTIUM REMEDIATION - A MECHANISTIC STUDY

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Engineered biopolymers have received a great deal of interest for environmental remediation of radionuclides in recent years. Composites derived from alginate, chitin or activated biocharsexhibit promising uptake capacities for common and problematic radionuclides such as strontium, cesium and uranium due to a wide range of surface functionality, extensive multiscale porosity, and excellent environmental compatibility. They also lend themselves well to a wide range of directed modification and functionalisation via organic chemical modification, metal doping or physical/structural changes. Traditional inorganic zeolites for radionuclide remediation possess a number of drawbacks which can drastically limit their efficacy in many environmentally relevant scenarios, as is highlighted by continuing challenges at the Fukushima Daiichi site. These include reduced performance in seawater, in acidic media and rapid fouling by dissolved organic matter. Synthetic zeolites can be costly and introduce technical handling challenges as fine particulate matter. Inorganic ion-exchange media can be bulky and expensive to dispose of. Biopolymers are well suited to address these deficiencies in remediation technologies and compliment the use of inorganic ion-exchangers. Unlike ion-exchange resins, the carbonaceous nature and large pore volume of biopolymers means that post-processing volume reduction ratios of up to 1000 are possible by compaction or ashing. Keeping final waste inventories to a minimum volume is in line with key decommissioning principles and representing savings in long term storage costs. Synthesis of monolithic composites such as biochar-alginate hydrogel beads are trivial to separate post uptake and do not suffer significant performance issues at low pH. Biopolymers are cheap, sustainable and readily available, and their use in this context is compatible with in-situ methods such as Monitored Natural Attenuation. Lacking however, are in-depth fundamental studies which unravel the binding mechanisms in such novel materials. Without which, optimisation and making a safety case for this class of materials as a remediation technology will be difficult. We aim to address these shortcomings by:

- Developing functionalisation regimes that not only nullify physicochemical variability but also enhance the number of binding sites and binding affinity; judicious choice of activation protocol also allows for tunability of the adsorbent in response to specific remediation requirements or local environmental conditions
- Creation of activated monolithic biopolymer hydrogel composites, making recovery of spent materials trivial compared with fine particulate adsorbents and
- Present mechanistic EXAFS data which highlights the inner sphere-type binding of strontium adsorbed to the biopolymer hydrogel.

ID: 1028

THE INFLUENCE OF BATCH SORPTION METHODOLOGIES ON CESIUM, NICKEL UPTAKE ONTO GRANITIC ROCK

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The batch sorption method of measuring radionuclide distribution between the crushed, sieved solid phase and liquid phase is commonly used for obtaining sorption coefficients Rd/Kd. These are often crucial input parameters used in reactive transport models for estimating a possible future radionuclide release from a radioactive waste repository. Therefore, it is extremely important to standardize a robust methodology for the batch sorption experiments which can provide results (under simulated repository conditions) with non-systematic error margins, so the possible affecting parameters are under control and not allowed to influence the measured Rd values (or possibly Kd in case of the linear sorption isotherm). Seven different methodological features of the batch sorption experiments were investigated using ^{134}Cs and ^{63}Ni as tracers. The solid phase was represented by migmatite gneiss from Bukov URF, using synthetic granitic water SGW2 as the liquid phase. Investigated methodological features were following: contact time (7, 14, 28 days), non-radioactive carrier vs no-carrier experimental setup, continuous shaking vs occasionally shaking, pre-equilibrated vs non-equilibrated solid phase, different construction of linear sorption isotherm (variable concentration vs solid/liquid ratio), anaerobic vs aerobic environment. Continual sample shaking during the batch experiment led to higher Rd values for both radionuclides compared to occasionally shaking. The reason for this difference could be that in the occasionally shaking method, diffusion can be the process controlling the sorption of radionuclides. In the case of Cs, Rd values obtained for pre-equilibrated granite were higher than for non-equilibrated migmatite, probably due to different ionic strength of supernatant after pre-equilibration or by exchange with cations in the rock component biotite during pre-equilibration. However, lower Rd values for Ni on pre-equilibrated granite were observed, which points to a different sorption mechanism for Ni than for Cs. Measured Rd values increased with sorption time for both radionuclides. This observation can be possibly explained by the formation of new sorption sites due to particles mechanical abrasion or rock surface changes. Kd values, obtained from a linear sorption isotherm for the Cs sorption with non-radioactive carrier were lower than for the system where the carrier was not present as a consequence of a lower number of available sorption sites in the case of Cs carrier presence. In the case of linear sorption isotherm obtained using differing V/m or differing radionuclide concentration, both methods gave similar Kd values for both radionuclides. However variable concentration method suffers from the small number of experimental values. Moreover, using batch sorption experiment with variable concentration method produced a larger volume of radioactive waste. Anaerobic conditions might have a significant impact not only on the chemistry of studied radionuclides in the case of Ni. Cs as a weak Lewis acid practically do not form complexes. In this investigation, SGW2 was replaced by a calcium chloride solution with equivalent ionic strength, considering composition changes of SGW2 in anaerobic conditions. Kd values for Ni and Cs were higher under the anaerobic than in the ambient environment.

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ID: 1029

FUNDAMENTAL INVESTIGATIONS OF ACTINIDE IMMOBILIZATION BY INCORPORATION INTO SOLID PHASES RELEVANT FOR FINAL DISPOSAL

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This contribution provides an overview of a current research network funded by the German Federal Ministry of Education and Research (BMBF), entitled "Fundamental investigations of actinide immobilization by incorporation into solid phases relevant for final disposal" – AcE. The AcE project aims at understanding the incorporation and immobilization of actinides (An) in crystalline, repository-relevant solid phases, such as zirconia (ZrO_2) and UO_2, but also in zircon ($ZrSiO_4$), pyrochlores ($Ln_2Zr_2O_7$) and orthophosphates of the monazite type ($LnPO_4$), which may find use as host matrices for the immobilization and safe disposal of high-level waste streams. The main objectives of the AcE project are (i) the development of synthesis strategies for An(IV)-doped solid phases, (ii) understanding their associated structural and physical properties using combined modelling and experimental approaches and (iii) determining their performance after irradiation with particular regard to an assessment of their long-term stability, dissolution behavior, and suitability for An matrix incorporation. Recent results obtained for ZrO_2, the main corrosion product of the Zircaloy cladding material surrounding nuclear fuel rods, will also be discussed. ZrO_2 is monoclinic phase (P_2(1)/c) at ambient conditions, and transforms into tetragonal (P_4(2)/nmc) and cubic phases (Fm$_3$ $\bar{\text{m}}$) at high temperatures of around 1200 °C and 2370 °C, respectively. However, particle size effects, the incorporation of foreign ions such as the actinides, as well as high radiation fields are known to also influence the stability fields of the polymorphs. A short overview of experimental studies conducted by the AcE partners, addressing both the ZrO_2 bulk structure, irradiation-induced changes, as well as the An environment during and after such structural transformations, will be given.

ID: 1036

STABILITY & PHYSICO-CHEMICAL CHARACTERISATION OF RECONDITIONED WASTE FORM RELEVANT TO RADIOACTIVE WASTES

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Nuclear power plants use ion exchange resins to remove radioactive contaminants present in the process waters. At certain intervals, spent ion exchange resins are replaced with fresh resin and the now active, spent resins are typically immobilized and disposed of. VTT, Finland, has developed a process to treat spent resins that significantly reduces the volume of resin to be disposed of and enables more efficient immobilization prior to disposal. Here, a tailored gasification process ashes the used resin and the residue is geopolymerized and prepared for storage. The geopolymerized resin residue will be stored in a Low and Intermediate Level Waste (LILW) disposal in a geological repository ~100 m below the surface in crystalline bedrock. During storage, the space surrounding the solidified waste packages will be backfilled with cement; and any passing waters will therefore be in equilibrium with this resulting in a high pH (~12), Ca rich solution which could act as a leachate potentially mobilizing the geopolymerized radionuclides. In order to better comprehend radionuclide stability in the geopolymer, we conduct laboratory-scale leaching experiments relevant for Finnish LILW disposal. Aqueous analyses assess changes in solution geochemistry and the potential for colloid formation. Here, we follow the geochemical behaviour of key leachant and geopolymer components such as Ca, Al and Si; we also assess the potential for radionuclide mobilization using stable isotopes as analogues for radionuclides of interest (^{59}Ni, ^{60}Co and ^{137}Cs). Following on-going short-term (30 & 90 day) and long-terms (1 & 2 year) leaching experiments we sacrifice samples to characterize waste element speciation within and across the leaching zone of the waste form. As a result of these experiments, we expect to have a comprehensive understanding of this wasteform performance and ability to retain contaminant elements specifically targeting radioactive waste speciation in both short- & long-term behaviour. These data will demonstrate the reliability of the novel geopolymer and may be used to inform the safety case for the long-term storage of LILW.

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ID: 1043

DIFFRACTION AND RAMAN SPECTROSCOPY STUDIES OF ZIRCONIA SOLID PHASES CONTAINING CERIUM

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Zirconia (ZrO_2) doped with lanthanides or actinides has been extensively studied for several tailored applications, such as for the immobilization of actinides present in high-level radioactive waste streams (HLW) or as inert matrix fuel for the incineration of e.g. waste plutonium. Doped zirconia matrices have been reported to have a very high radiation tolerance, however, inconsistencies exist concerning the role of the different structural polymorphs in the high radiation resistance. In addition, the role of oxygen vacancies, which are formed for charge compensation when subvalent dopants are incorporated into the ZrO_2 crystal structure, have not been clarified. The lanthanide cerium is often used as a surrogate for plutonium due to its similar ionic radius and comparable chemical properties in the oxidation states +III and +IV. In this study, different Ce concentrations were incorporated in zirconia via the co-precipitation route, with the goal to stabilize the cubic zirconia polymorph. Due to the extraordinary luminescent properties of Eu, a trace amount of this lanthanide was added together with Ce in the synthesis process. A series of zirconia samples doped with 14 to 70 mol % Ce was prepared. Seven selected compositions are presented here, namely samples with 14, 22, 30, 42, 50, 62, 70 mol % Ce, as they demonstrate a clear phase transformation. The phase composition was evaluated by powder X-Ray diffraction (PXRD), and Raman spectroscopy. In the PXRD diffractograms, the monoclinic phase, characterized by diffraction peaks at 28.2 ° and 31.3 °, is dominant only in the composition with 14 mol % Ce. Above this concentration, a peak around 29.9°, assigned to the tetragonal phase, increases in intensity as a function of increasing Ce concentration in the zirconia matrix up to a concentration of 42 mol%. Beyond this, the cubic phase starts to dominate the phase composition, with the intensive characteristic peak around 29.3°. Additionally, the presence of a tetragonal metastable phase in samples with 30, 42, and 50 mol % Ce, and a trace of the tetragonal phase in the composition with 70 mol % Ce were identified. Generally, the diffraction peaks are shifted to lower 2 angles as a result of the substitution of Zr_4^+ by the larger Ce_4^+ cation which increases the lattice parameters. The Raman results corroborate the assignment of the different phase compositions identified in the PXRD studies. However, a large band at 512 cm^{-1} becomes visible in the samples with 30 mol % Ce and grows in intensity with increasing Ce-doping. This band has been reported to arise from Frenkel-type defects, typically found in samples with oxygen vacancies or/and due to partial reduction of Ce_4^+ to Ce_3^+ occurring in the samples, and causing the formation of oxygen vacancies in the ZrO_2 structure for charge compensation. This, however, has to be verified in future XANES experiments. No solid phase separation was detected in both characterization analyses, PXRD and Raman. Luminescence spectroscopic studies, probing the Eu ion which has been incorporated together with Ce in all ZrO_2 solid phases, will be conducted in future studies. Preliminary results of these investigations will be shown at the RadChem conference.

ID: 1050

INTERACTION OF ^{137}CS WITH SYNTHETIC GEOPOLYMER AND CEMENTIOUS MATERIALS

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When nuclear facilities, all types of radioactive waste are generated, including intermediate and highly active radioactive waste, which cannot be disposed of in current radioactive waste repositories in the Czech Republic. Therefore, it is necessary to find filling materials used in deep repositories that are environmentally safe for a long time. The aim of the ALMARA project is to study and test these materials. The subject of this work is the sorption of cesium in cementitious materials and synthetic geopolymers, the determination of the equilibrium time required for the sorption of ^{137}Cs. The batch experiment was carried out with a solution of ^{137}CsCl in synthetic granite water on a synthetic geopolymer, hardened cement paste, cement mixed with iron nanoparticles (1 % by weight) or bentonite (20 % by weight). These materials were used in two different particle sizes (mesh fractions less than 0.5 mm and mesh fractions between 1 and 2 mm) and two different liquid-solid ratios (5 and 10 ml/g). Partition coefficients were calculated to compare these materials.

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ID: 1051

UPTAKE OF ^{85}Sr ONTO CEMENTIOUS MATERIALS AND SYNTHETIC GEOPOLYMER

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The decommissioning of nuclear facilities will produce all levels of radioactive waste, including intermediate- and high-level waste that is not allowed to be disposed of in low- and intermediate-level radioactive waste repositories. Therefore, it is necessary to find infill materials used in a deep geological repository that will make it safe for the environment for a long period of time. The aim of the ALMARA project is to study those materials and test them. The subject of this work is strontium sorption on cementious materials, finding the equilibrium time needed for ^{85}Sr sorption. Batch type experiment was performed with solution of $^{85}SrCl_2$ in synthetic granitic water on hardened cement paste (CEM), cement mixed with 20 wt% of bentonite (AFM), cement mixed with 1 wt% of nano iron particles (NNM) and synthetic geopolymer (GP). These materials were used in two different grain sizes (mesh fraction less than 0.5 mm and mesh fraction between 1 – 2 mm) and two different liquid-solid ratios (5 and 10 ml/g). To compare those materials, distribution coefficients (R_d) were calculated. The best result was obtained for GP, with the equilibrium time being one week and the Rd value being in the thousands of ml/g. For other materials, the equilibrium time was about two months and the Rd values were significantly smaller as follows: AFM > NNM > CEM.

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ID: 1053

ALTERNATIVE INFILL MATERIALS FOR DISPOSAL OF NPP DECOMMISSIONING RADIOACTIVE WASTE – ALMARA PROJECT INTRODUCTION

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Project introduction The aim of the ALMARA project is the optimization of infill matrixes for disposal of intermediate and high level wastes from nuclear power plant decommissioning, which fulfil requirements that ensure long term safety of deep geological repository over long term period. The project is also focused on radionuclide interaction and migration with/in the matrix materials and corrosion and microbiological studies. Finally, technological application will be evaluated for studied infill matrixes. Project experimental plan and testsFour different infill matrixes were designed at the beginning of the project: CEM – ordinary Portland cement; AFM – alternative filling matrix, which is based on cement and bentonite; NNM – new nano-based matrix, which is based on cement and nanomaterial; GP – geopolymer matrix. Experimental program started by developing of infill matrixes composition and verifying the pro-posed properties (workability, mechanical and structural stability, compressive strength). Composition of matrixes is followed: CEM – cement type CEM I only, using W/C ratio 0.34; AFM – mixture of cement type CEM I and Ca/Mg bentonite (ratio 80/20 wt.%) with addition of commercial plasticizer and W/C ratio 0.5; NNM – cement type CEM I with addition (1 wt.%) of iron nano-powder (using W/C ratio 0.34); GP – based on composite aluminosilicate and alkaline activator, detailed composition is confidential. Experiments studying corrosion processes on stainless and carbon steel (representative materials of NPP decommissioning) are performed in all four types of matrixes. Disc shape specimens of steels were fixed into the tested materials. Two types of samples were casted (cubes with an edge of 5 cm and 10 cm). Testing cubes are placed in synthetic granitic water to simulate the interactions in the repository. Real waste samples of activated steel from NPP witness samples programme and contaminated tita-nium material (filter from NPP operation) are also studied in this project. These samples were immobilised in cement and geopolymer matrix. This part of the project is focused on studies of radionuclide release from real wastes into the matrix or surrounding environment. The degradation of mechanical and chemical properties of matrix materials will be studied after de-fined periods of time (up to 3 years). The chemical composition, mineralogy, structural and mechanical properties and microbial activity will be tested and also interaction and migration of radionuclides on degraded materials will be characterised by sorption and diffusion experiments and compared to unaffected matrix materials. All initial requirements on workability, mechanical and structural stability, compressive strength of matrix materials (CEM, AFM, NNM, GP) fulfilled and materials were characterised by chemical com-position, mineralogy and porosity. Real waste samples of activated steel and titanium filter were characterised for their radiochemical composition and main radioactive contaminants were identified. Sorption and interaction experiments with radionuclide tracers were started.

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ID: 1054

^{14}C LABELLED ORGANIC MOLECULES MIGRATION AND INTERACTION IN/WITH THE CEMENT BASED MATERIALS

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Introduction Cements and concretes have been widely used in intermediate/low level waste (I/LLW) management in Czech Republic, being used namely as solidification material. Moreover, cement materials are con-sidered also for disposal of high level waste (HLW) in deep geology repository (DGR), hereby being considered both for solidification and construction materials. ^{14}C was chosen as a relevant contami-nant and also organic compounds are present in concrete as an additives and plasticizers and their degradation products which may influence the migration and interaction of other contaminants. Sorption and diffusion experiments ^{14}C acetate and ^{14}C formate were applied as tracers in the sorption and diffusion experiments on different cement based materials (cement pastes, mortars and concretes). The acetate and formate were also identified as the representatives of organics degradation product of plasticizers. Sorption experiments with were arranged in the form of batch reactors, where the crushed material is mixed with the liquid phase, pure portlandite water or synthetic granitic water (SGW). Diffusion exper-iments were applying through diffusion method. The tested solid phase materials are disc shape samples (50 mm in diameter, approx. 10 mm thick) which are placed into own designed and made diffusion cells using the portlandite water or synthetic granitic water (SGW) as the liquid phase. Migration and interaction of ^{14}C labelled acetate and formate are also performed to describe the behaviour of organics degradation products in cement environment. Structural, mechanical, chemical and mineralogical characterisation of cementitious materials is also provided to describe the properties of solid phase and their influence in migration properties.

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ID: 1059

RADIATION STABILITY OF EXTRACTION SYSTEMS WITH HYDROPHOBIC CYME4-BTBP AND CYME4-BTPHEN EXTRACTING COMPOUNDS AND HYDROPHILIC $(SO_3H)_2$-BTP, $(SO_3H)_2$-BTBP, AND $(SO_3H)_2$-BTPHEN MASKING AGENTS

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Extraction properties and radiation stability of two systems with hydrophobic extractants (CyMe4-BTBP or CyMe4 BTPhen) and three systems with hydrophilic masking agents ($(SO_3H)_2$-BTP, $(SO_3H)_2$-BTBP or $(SO_3H)_2$ BTPhen) were studied throughout the GENIORS project. The study of extraction systems containing CyMe4-BTBP or CyMe4-BTPhen dissolved in fluorinated BK-1 diluent revealed that the extraction properties of the BK 1-based solvents are promising. However, the radiation stability of those extracting compounds in the diluent BK-1 is insufficient for practical application.[1] Next, a study performed with another fluorinated diluent FS-13 revealed that this system shows much better stability than the system with BK-1. The behaviour of the CHALMEX system after irradiation by accelerated electrons at different temperatures was studied. The study of the irradiation of the solvent in the presence of nitric acid aqueous phase confirmed its earlier observed radioprotective efffect. A comparison of the theoretical values of distribution ratios after irradiation, calculated from the residual concentrations of the ligand, with their experimental values suggests that some of the CyMe4-BTBP degradation products or adducts of CyMe4-BTBP or TBP are probably able to extract the studied metals, too. The i-SANEX extraction system with the masking tetrasulfonated BTPhen ligand ($(SO_3H)_2$-BTPhen) was tested for its behaviour under irradiation. The general trend observed was that up to 200/250 kGy the D values for Am(III) and Eu(III) remained similar, but for higher doses the values of distribution ratios changed dramatically. The trend in SFCm/Am values was similar to the trend of values of SFEu/Am.[2] Furthermore, characterization of $(SO_3H)_2$-BTP or $(SO_3H)_2$-BTBP irradiated at different temperatures was performed. After the irradiation of $(SO_3H)_2$-BTP-containing aqueous phase without contact with organic phase to high absorbed doses, the DAm and DEu exceed the values of blank – some degradation products/adducts formed probably act as efficient non-selective extractants of the mentioned metals. When the aqueous phase was in contact with organic phase during the irradiation, the system was more stable against radiolysis and DAm values increased more slowly. The system with $(SO_3H)_2$-BTBP behaved in a very similar way as the $(SO_3H)_2$-BTP one, however, it was much more radiation stable than the $(SO_3H)_2$-BTP system.

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ID: 1062
SORPTION OF RADIONUCLIDES FROM AQUEOUS SOLUTIONS ONTO ZEOLITES

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Radioactive waste contains large amount of radionuclides. A radioactive isotopes of caesium (^{137}Cs, half-life time 30 years) and strontium (^{90}Sr, half-life time 28 years) belong to the main fission products existing in the radioactive wastes produced in nuclear power. The ion exchange technology is one of the most commonly used methods for safe treatment of radionuclide waste. Zeolites are often used as sorbents of radionuclides. In this study, zeolites A, X and JBW, were synthesized by a hydrothermal method from metakaoline. Sorbents were tested for sorption of ^{134}Cs and ^{85}Sr from various aqueous solutions. The prepared zeolites were characterized by X-ray powder diffraction, Raman and FTIR spectroscopy. Morphology was observed by scanning electron microscopy. The results showed that ^{134}Cs and ^{85}Sr radionuclides were efficiently adsorbed onto zeolites.

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ID: 1073
SORPTION OF EUROPIUM ON CEMENTITIOUS MATERIALS IN THE PRESENCE OF ORGANIC SUBSTANCES

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Cement-based materials are used as engineering barriers for low-level and intermediate-level radwaste and in high-level radwaste disposal as facilities or buffer materials. To evaluate the long-term safety of radioactive waste repositories, it is necessary to describe the behavior of stored radionuclides that will be present, such as europium used as an analog of trivalent actinoids. The migration of radionuclides is affected by interaction with the material of the engineering barriers of the repositories, including cement-based barriers. In this study, the sorption of europium on hardened cement paste (HCP) of type CEM I, CEM III and synthetic cement phase CSH (Calcium-Silicate-Hydrate) with Ca/Si ratio 1.0 was carried out with the addition of different organic substances (adipic acid, phthalic acid, EDTA). Radionuclide sorption is described by the distribution ratio (R_d) between the liquid and solid phases (L/S) or with sorption isotherm.

The research leading to these results has received funding from the European Union’s Horizon 2020 Innovation Programme under grant agreement n° 847593 (EURAD – CORI). The output was created with financial participation of SÚRAO (Czech Radioactive Waste Repository Authority) (SO2020-017). This contribution is also partially a result of the grant of the CTU Student Grant Scheme No. SGS22/187/OHK4/3T/14.

ID: 1075
GEOPOLYMER MATERIALS FOR NUCLEAR INDUSTRY

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Geopolymers are promising materials applicable in nuclear industry. They have been tested for various applications like nuclear waste immobilisation, as a construction material, for concrete cracks reparation, etc. We have developed a synthetic route for production of versatile geopolymer material with application in a passive system of GEN IV reactor safety. Such material contains a geopolymer matrix based on alumino-silicates and a filler. The filler powder can be broadly selected according to the needs (Gd_2O_3 as neutron absorber, Al_2O_3 as refractory oxide, Fe_2O_3 as chemically active material, etc). We have characterised the products by various solid-state techniques (mechanical properties, electron microscopy, X-ray powder diffraction). Additionally, we have tested the radiation stability of the material under gamma radiation of ^{60}Co source mimicking the condition around the reactor pit during the life time of the material.

ID: 1077
STRUCTURAL CHARACTERISATION OF HEAVY LANTHANIDE OXALATES SYNTHESIZED BY HOMOGENEOUS PRECIPITATION

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Metal oxalates are salts of very low solubility allowing an easy precipitation of metal ions from the acidic aqueous solutions into crystalline material. Due to this property, oxalates have an important role in the technology of lanthanides and actinides. It is namely the separation of actinides from the spent nuclear fuel that is its application of interest. 4f-lanthanides are often used as 5f-actinides surrogates when optimizing or modeling the precipitation process. A synthetic route for homogeneous precipitation of oxalates based on the thermal decomposition of oxamic acid was developed, as published recently[1]. Further investigation about the structure and morphology of products of this reaction using six heaviest lanthanides (Dy, Ho, Er, Tm, Yb, Lu) was carried out. The products, developed microcrystals, were studied by means of solid-state analysis (X-ray crystallography, X-ray powder diffraction, TGA). New, previously undescribed structures of $Er_2(C_2O_4)_3.7H_2O$, $Tm_2(C_2O_4)_3.7H_2O$, and $Lu_2(C_2O_4)_3.7H_2O$ were obtained. According to our observations, the synthesized oxalates form 2D framework structures that are stacked in one direction and connected by hydrogen bonds to form a 3D supramolecular structure. Each lanthanide atom is coordinated by three bidentate oxalates, two water molecules and weakly by one water molecule residing in a cavity. The cavities observed in the structure increase in size with decreasing atomic number of lanthanoid and are of great interest for its possible applications.

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ID: 1090
THE SORPTION OF Tc(IV) TO SOME CLAY MINERALS

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^{99}Tc is one of the most important isotopes likely to be disposed of in the proposed UK Geological Disposal Facility for higher-activity radioactive wastes, due to its long half-life, high fission yield and ability to migrate through the geosphere as the pertechnetate ion. However, much of the technetium is likely to be in the lower oxidation state of Tc(IV) due to the low Eh in the near field. Batch sorption experiments across the pH range have been performed on Tc(IV) using ^{95m}Tc as a spike in the presence of some representative clay minerals (bentonite, smectite, kaolininte, montmorillonite and illite). Tc(IV) solutions were used at trace concentrations to avoid precipitation as technetium dioxide. Values for the partition coefficient (Rd) were found to range from 7 to 2e5 ml/g. Rd was heavily dependent on pH in all cases, with the highest values being found in the circumneutral area. These data will inform the performance assessment for the behaviour of technetium in the near-field of the UK's planned higher-activity wastes GDF. Surface complexation modelling of the data has been performed.

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5. Production and Application of Radionuclides

ID: 0882

IRRADIATION OF YTTRIUM MICROSPHERES AT THE IRT-T REACTOR

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National Research Tomsk Polytechnic University (TPU), together with an industrial partner (BEBIG), start produced yttrium microspheres at its research nuclear reactor this fall. This radiopharmaceutical is intended for the treatment of liver cancer in inoperable patients and, unlike analogs, it destroys the tumor in a targeted manner without affecting healthy organs and tissues. Irradiation of yttrium-89 microspheres will be carried out at the IRT-T reactor, power 6 MW, neutron flux 1.7 10^{14} neutron/cm^2. To obtain one dose of the final preparation, a weighed portion of 0.1 g of yttrium microspheres is irradiated. Microspheres are processed using distilled water, alcohol and hydrochloric acid. Yttrium microspheres are injected into the patient's bloodstream, which delivers them directly to the tumor. After delivery, the microspheres block the access of blood with oxygen to the metastases, in parallel acting on them with beta radiation. In the Russian Federation, this method of treatment is not yet massively applied. The industrial production of yttrium microspheres began in the fall of 2020. There have already been test deliveries to Moscow clinics. The drug is registered and has permits for use in clinical practice. April 9, 2021 at A. Tsyb Medical Radiological Research Center for the first time in Russia, clinical trials of the method of radioembolization of tumors with domestic microspheres produced by the Russian company "Bebig" began. Four operations were performed at once on patients with inoperable forms of liver cancer.

ID: 0885

PRODUCTION OF THORIUM-227 EXPERIMENTAL SAMPLES: PROBLEM OF ACTINIUM-227 IMPURITY CONTROL

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Thorium-227 radionuclide is perspective as a part of radiochemicals for therapy of various types of cancer. It forms strong complexes with monoclonal antibodies due to its chemical properties. In case of its full decay, five alpha particles are emitted, though there are some concerns related to the opportunity of its daughter radionuclide radium-223 uncontrolled migration over the patients’ organisms. At present, thorium-227 based radiochemicals are undergoing clinical and preclinical studies. At RIAR thorium-227 was generated from a long-living radionuclide actinium-227, that was produced by irradiating of the targets containing radium-226, in a high-flux SM-3 reactor. ^{227}Th was extracted by anion exchange chromatography from 8 M HNO_3 on BioRad AG-1x8 sorbent. ^{227}Th was eluated with 1 M HCl from chromatography column. For additional purification, ^{227}Th solution was evaporated to dryness, the residual was dissolved in 8 M HNO_3 and then it was purified by anion exchange again. One of the most important characteristics of thorium-227 based radiochemicals is the impurity content of a long-living actinium-227 alpha-emitter. At the same time, direct measurement of its activity by alpha and beta spectrometry does not allow achieving the required detection limits. ^{227}Ac decay is not accompanied by a characteristic gamma-radiation. The peaks in alpha-spectrum corresponding to actinium-227 is on the low-energy slope of alpha peaks of thorium-227 and radium-223. The situation is complicated due to the low yield of alpha-radiation (1,38 %), that gives the detection limit ~1 % at the collected statistics of 1 million pulses in the alpha spectrum. The conversion electrons and beta-radiation of ^{223}Ra daughter decay products namely lead-211 and bismuth-211 interfere with the measurement of ^{227}Ac activity due to beta-radiation. There are two main approaches to determine actinium-227 content in thorium-227. The first approach is to store the chosen radiochemical sampling for a long time for thorium-227 decay and to measure alpha and gamma spectra. The second one is based on the chemical extraction of actinium-227 traces from the radiochemical aliquot and the following measurement of their activity. Both approaches are used in this work. The radiochemical thorium-227 aliquot with activity up to 1 mCi were used for chemical extraction. To prevent ^{227}Ac sorption on the lab glassware surface and to estimate its chemical yield, 1 µg of stable europium in the form of a nitric acid solution of $Eu(NO_3)_3$ containing the radioisotope label europium-152 was added to the aliquot. The chemical extraction was performed in two stages. At the first stage actinium-227 was separated from thorium-227 by anion exchange chromatography on BioRad AG1x8 resin. At the second stage actinium-227 was separated from radium-223 traces by cation exchange chromatography in the presence of ammonium salt EDTA on BioRad AG50x8 resin. Actinium-227 activity was measured by alpha-spectrometry. Actinium-227 detection limit achieved by this method was from ~(7-9)·10^{-5} %. Full characteristics of the obtained experimental samples are represented in the report.

ID: 0895
DESIGNING METAL OXIDE-BASED STATIONARY PHASES FOR THE SEPARATION OF Ac-225 AND BI-213 FOR BIOMEDICAL APPLICATIONS

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Cancer is a disease that still kills nearly 10 million people every year[1] despite the countless existing treatments. The amount of novel therapies is growing every day and targeted alpha therapy is one of them. Herein, the patient is treated with a radiopharmaceutical that binds to the cancer cell and emits ionizing radiation to induce cell damage. Since alpha radiation has a very short range (only a few cell diameters), the healthy cells around the tumor are spared. Amongst a number of radioactive isotopes that can be used for this purpose, ^{213}Bi is a promising candidate because of its suitable half-life and well-known chelation chemistry. Clinical trials with this isotope have already been performed in the past and results are very promising[2]. The biggest hurdle, however, is the ^{213}Bi production, which is now far from sufficient to implement this isotope in targeted alpha therapy on a large scale. The most commonly used generator for the separation of ^{225}Ac and ^{213}Bi is based on AG MP-50. This is a macroporous strong acid cation exchange resin composed of sulphonic acid groups attached to a styrene divinylbenzene copolymer backbone. The AG MP-50 generator has a high yield (>76 % ^{213}Bi), a very low breakthrough of ^{225}Ac (<210-5 %) and the ^{213}Bi elution is very fast (2-3 min). Despite these great advantages, the main disadvantage is its low radiation stability. The organic backbone and especially the link with the functional group is very susceptible to radiation damage, such that in normal operation conditions the performance of the generator will substantially decrease[3]. On the other hand, inorganic materials are known to be more resistant against radiation. Therefore, this contribution presents the study of surface modified inorganic materials and their performance towards ^{225}Ac/^{213}Bi separation. The materials under investigation are titania and zirconia, which are modified with phosphoric acid groups. Different analytical techniques such as zeta potential and ICP-MS are utilized to characterize the surface modification of the inorganic materials. Furthermore, the modified materials are subjected to a so-called ‘cold sorption’ test. Via this method, sorption properties such as separation efficiency, leaching stability and sorption capacity are evaluated by means of non-radioactive solutions of La and Bi. Ultimately, the actual ^{225}Ac/^{213}Bi sorption properties of the phosphoric acid-modified materials will be tested in a radiochemical laboratory. The final goal of this research is to develop an improved direct ^{225}Ac/^{213}Bi generator material with high radiation stability and increased ^{213}Bi yield and purity to make targeted alpha therapy more available and to give cancer patients new hope for a better future.

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ID: 0905
MEASUREMENTS OF CUMULATIVE CROSS SECTIONS

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By definition a cumulative production cross section of a radionuclide includes formation via direct nuclear reactions and formation via radioactive decay of the precursor nuclide (considering isomeric transition too). In literature different definitions of the cumulative cross sections exist. The solution of the coupled differential equation describing the radioactive growth and decay for a mother – daughter pair provides a formula which is used to determine the cumulative cross section by experimental activation technique. As most of the light charged particle induced reaction cross sections are measured using activation technique and gamma-spectrometry this formula is used for the definition of the cumulative cross section. Using this method the cross section deduced from the peak area evaluated from the gamma-spectra measured after the complete decay of the precursor and that is most time defined as cumulative cross section in the literature. The tc cooling time is measured from the end-of-bombardment (EOB), but after EOB decay production of daughter nuclides is still continued. Therefore, defining the cumulative cross section this way is always larger then the sum of the direct cross sections of the precursor and the daughter nuclides since the c cumulative factor is larger than 1. However, using different experimental technique, for example, counting the number of neutrons formed in a (p,n) reaction producing an IT decaying isomeric state (m) and ground state (g) pair of a radionuclide the deduced cumulative cross section is the sum of the two direct cross sections. Due to different definitions of the cumulative cross section data measured by activation method and other experimental techniques should be handled separately and carefully. The c cumulative factor is time dependent. For a precursor – daughter pair, when the half life of the precursor is shorter than the half life of the daughter, applying a very long cooling time c can be approximated as c=Td/(Td-Tp). However, applying short cooling time the value of c can be even less than 1 indicating the partial decay of the precursor. Doing measurement for a cumulative cross section one should avoid partial decay of the precursors, therefore the measurement should be done always after the total decay of the precursor(s). Otherwise, the deduced cross sections would be time dependent because of the time dependence of the c factor. Early measurements would result in scattered and too low cross sections. The time dependent experimental cumulative cross sections cannot be compared to other literature data and therefore they are useless. Analysis of time dependence of the cumulative factor c and its consequences are discussed in the work.

ID: 0907

SEPARATION OF ^{213}Bi VIA AN INVERSE ^{225}Ac/^{213}Bi RADIONUCLIDE GENERATOR BASED ON SULFONATED CARBON MATERIALS

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Bismuth-213 (^{213}Bi) is an alpha-emitting radioisotope with large potential in nuclear medicine for cancer treatment. Several clinical trials of ^{213}Bi-based radiopharmaceuticals have provided evidence for its therapeutic efficacy. ^{213}Bi is produced from the relatively long-lived parent nuclide actinium-225 (^{225}Ac) and then separated in a radionuclide generator. The patient dose of ^{213}Bi is estimated to be 1 mCi/kg body mass, and the optimal ^{225}Ac/^{213}Bi radionuclide generator should separate ^{213}Bi from 100-150 mCi ^{225}Ac. However, previously used sorbent materials (e.g. Dowex 50W-X8, AG MP-50, Actinide Resin, UTEVA Resin, and Termoxid-39) have limitations, including poor radiolytic, chemical and physical stabilities. Therefore, developing alternative materials to overcome those shortcomings is a priority. This study evaluated sulfonated carbon materials for use in inverse ^{225}Ac/^{213}Bi radionuclide generators. The synthesis protocol was optimized with regard to the pyrolysis temperature and sulfonation conditions. The materials were characterized with regard to the pore size distribution, the nature of the functional groups and the zeta potential. The separation of Bi_3^+, La_3^+ (as surrogate for ^{225}Ac) and Ac_3^+ using the sulfonated carbon materials were examined by batch experiments, as a function of different parameters (e.g. pH, salt concentration, solid-to-liquid ratio, and contact time). The sulfonated carbon materials exhibited a high selective uptake of Bi_3^+ in the presence of high salt concentrations at low pH. Inversely, the sorption capacity for La_3^+ and Ac_3^+ onto the sulfonated carbon materials decreased with increasing salt concentrations and decreasing pH. Batch desorption results showed that a high percentage of Bi_3^+ can be eluted from sulfonated carbon materials by HCl, NaI, or NaCl solutions. More importantly, the sulfonated carbon materials showed high levels of resistance against radiolysis, fast sorption kinetics, good durability, and recyclability. The evaluation of the different materials enabled a better understanding of the sorption mechanisms of Bi_3^+ and Ac_3^+ onto these sulfonated carbon materials. Based on the separation performance, the most suitable sorbent was examined using column chromatography. The ^{213}Bi yield from the inverse generator reached 94 % in 1 mL of 1 M HCl with an ^{225}Ac impurity of less than 0.04 % of the eluted ^{213}Bi activity. Experiments also revealed that a guard column with AG MP-50 reduced the impurity of ^{225}Ac without affecting the ^{213}Bi yield. These findings indicate that the sulfonated carbon materials are promising adsorbents in inverse ^{225}Ac/^{213}Bi radionuclide generators for the production of high-purity ^{213}Bi for medical applications.

ID: 0959

SHINE PHASE II OF IV: A FOCUS ON Lu-177

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As part of a long-term vision to deploy fusion technology in a scalable path to practical fusion energy, SHINE Technologies has ventured to produce lutetium-177. While a one-of-kind ^{99}Mo production plant is being constructed, SHINE's Therapeutics division is currently manufacturing small quantities of $^{177}LuCl_3$ to support clinical studies for cancer radioligand therapy. SHINE expects a combination of technologies, including in-house target enrichment/recycling and non-reactor irradiation to be integrated into SHINE's commercial ^{177}Lu production, which could help to mitigate the supply chain shortages and position SHINE to scale the process to hundreds of patient doses per batch. This presentation will introduce you to SHINE Technologies and provide an overview of the clinical ^{177}Lu process.

ID: 0967
UPDATING THE NUCLEAR DATABASES: RE-MEASUREMENT OF THE HALF-LIFE OF Sm-146, Gd-148, AND Dy-154

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During the last years, an increased effort was put into the measurement of decay data with high precision, with the ultimate goal of confirming the existent nuclear values and/or lowering their uncertainties. Precise nuclear data is extremely important in the description of galactic events, in the nuclear dating of samples, as well in the evaluation of the toxicity of nuclear waste. Surprisingly, the re-evaluated half-life data of a considerable number of radionuclides (e.g., ^{60}Fe[1], ^{79}Sm[2,3] ^{146}Sm[4]) showed a substantial disagreement with previous values. The repercussions are significant: For example, the shorter measured half-life for ^{146}Sm implies a higher abundance of this radionuclide in the early Solar System, and thus, planetary events dated with the ^{146}Sm/^{142}Nd chronometer converge now to a shorter time span than previously estimated. Similarly to ^{146}Sm, the currently available half-life data of other radio-lanthanides, such as ^{148}Gd and ^{154}Dy, are inconsistent or affected by uncertainties even up to 50 %[5-7]. Through their pure alpha-decay chain, ^{148}Gd and ^{154}Dy directly influence both the abundance of ^{146}Sm, as well as the natural isotopic composition of stable Nd, contributing thus to the ^{146}Sm/^{142}Nd chronometer. Therefore, an exact knowledge on the half-life of both ^{148}Gd and ^{154}Dy is urgently required. It has to be mentioned, that reasons for these imprecise nuclear data lie in the difficulty of obtaining samples of the isotopes of interest in sufficient amounts and purity, together with inherent problematics in performing such demanding measurements. In this work, that belongs to the initiative “ERAWAST - Exotic Radionuclides from Accelerator Waste for Science and Technology”[8], we obtained sufficient amounts of ^{146}Sm, ^{148}Gd, and ^{154}Dy by reprocessing irradiated Ta materials available at the PSI accelerator-facilities. The re-determination of the half-lives of the above-mentioned radio-lanthanides proceeded by successively applying the “direct” method, which consists in the determination of the number of radioactive atoms in a specific sample, combined with the measurement of its radioactivity. Here, preliminary results on the measured half-lives of ^{148}Gd and ^{154}Dy, together with the first steps towards determining the decay constant of ^{146}Sm, will be presented.

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ID: 0983
METAL-ORGANIC FRAMEWORKS AS ADSORBENT FOR THE ^{99}Mo/^{99M}Tc GENERATOR

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This work explored the potential of several metal-organic frameworks (MOFs) as adsorbents for the development of ^{99}Mo/^{99m}Tc generator using ^{99}Mo produced by neutron capture of ^{98}Mo which is hampered by its low specific activity. The molybdenum adsorption performance of MOFs, including adsorption kinetics and isotherms, were determined and adsorption mechanism was studied by several analytical techniques such as X-ray photoelectron spectroscopy, Raman spectroscopy, zeta potential and density functional theory calculation. The results showed that the maximum adsorption capacity of UiO-66 (Ce) can reach up to 475 mg/g at pH 3. The high surface area and defects appear to increase adsorption sites, enhancing the affinity between the UiO-66 and the molybdenum ions by Zr-O-Mo coordination, anion-π as well as hydrogen bonds. To achieve a clinical application, the performance of ^{99}Mo/^{99m}Tc generator fabricated with the most promising MOFs was evaluated. The results demonstrated that 90 % of ^{99m}Tc can be eluted with acceptable cerium breakthrough. The obtained excellent separation performance demonstrates that MOFs are good candidates as adsorbents for ^{99}Mo/^{99m}Tc generator and should be explored more extensively in the future.

ID: 0994
NUCLEAR FISSION PRODUCTS FROM THE SINQ GAS-JET FACILITY

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Neutron-induced fission of ^{235}U leads to a variety of radionuclides. With such targets being hosted in the inner shielding shell of the Swiss Spallation Neutron Source SINQ at the Paul Scherrer Institute, the produced radionuclides can be readily transported to the radiochemical laboratory by the SINQ gas-jet installation. The transport of the resulting fission products occurs either in pure (volatile fraction of fission products) or aerosol-particle loaded (non-volatile fraction of fission products; e.g., carbon or KCl) carrier gases. Subsequently, the entire radionuclide mixture can be used for radiochemical experiments in the liquid or in the gas phase. In this way, the upgraded SINQ gas-jet facility has been successfully used to demonstrate the applicability of a whey-based filter material for the treatment of low- to medium-level radioactive wastewater. Further, experiments are currently in preparation. Here, we present the results of the re-commissioning experiments at the SINQ gas-jet facility, revealing its versatility towards radiochemical applications.

ID: 0999
PREPARATION THIN FILM SOURCES OF RADIOLANTHANIDES FOR THE MEASUREMENT OF AUGER ELECTRON ENERGIES AND BRANCHING RATIOS

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With recent advances in the use of novel radiolanthanides for targeted Auger electron therapy, the request for precise and controlled dosimetry has increased in importance. So far, no experimental data for the energy and branching ratios of Auger electrons have been reported for Auger-electron-emitting lanthanides. One of the main challenges related to such measurements is the production of a uniform thin film in order to avoid energy loss of the emitted Auger electrons through self-absorption. ^{161}Ho is considered a promising radiolanthanide for internal radiotherapy, thanks to its appropriate half-life (2.48 hours) and its chemistry being very similar to the one of other well-consolidated therapeutic lanthanides. In this study, holmium is deposited as a thin film via molecular plating. In particular, the effect of the solvent's vapor pressure on the deposition morphology is thoroughly investigated by means of autoradiography and scanning electron microscopy coupled with energy dispersive X-ray spectroscopy.

ID: 1002
PRODUCTION OF HIGH SPECIFIC ACTIVITY ^{51}Cr BY CHROMIUM-BASED METAL-ORGANIC FRAMEWORKS AND THE SZILARD-CHALMERS EFFECT

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Chromium-51 (^{51}Cr) is an attractive radionuclide in the clinical application for labelling of red blood cells, diagnosis of gastrointestinal bleeding and assessing life span of red cells. This work reports that synthesis of two chromium based metal-organic frameworks (MOFs) as radiation targets to produce high specific activity ^{51}Cr production utilizing the Szilard-Chalmers effect. First, the radiation stability of two MOFs under high gamma doses was determined. The results showed that MIL-100 (Cr) and MIL-101 (Cr) exhibited excellent radiation stability after exposure to gamma radiation dose of 4 MGy. However, MIL-101 (Cr) started decomposing with increasing gamma dose, while MIL-100 (Cr) still kept a stable crystal structure at even higher radiation dose. Subsequently the Cr-MOFs were irradiated in the HOR reactor of the Reactor Institute Delft. The ^{51}Cr yield and specific activity was determined at different irradiation time, extracting agents, extraction time and temperature. The most optimal results show that high enrichment factor (>500) and yield (>40 %) can be achieved, appearing a promising ^{51}Cr production routes for nuclear medicine.

ID: 1007
MOLYBDENUM NANOPARTICLES AS TARGET FOR THE PRODUCTION OF MOLBYDENUM-99

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The main objective is to develop a new production route for high specific activity molybdenum-99 based on the irradiation of molybdenum nanoparticles. In order to achieve this, molybdenum nanoparticles have been produced by spark ablation. The spark ablation technology provides an easy method to produce nanoparticles with controlled size. It consists of a gas phase physical process in which nanoparticles are produced by inducing spark discharges between electrodes, made of molybdenum in our case. Once produced, the nanoparticles will be irradiated by the accelerators in the JRC facilities in Geel and in the nuclear reactor HOR in TU Delft with high-energy photons or neutrons. The produced ^{99}Mo will experience a recoil effect that will expel the radionuclide out of the nanoparticle. By this atom displacement, the produced ^{99}Mo may be separated from the other isotopes of molybdenum that constitute the nanoparticles, leading to a high specific activity product. After the irradiation, the targets will be eluted to separate the produced ^{99}Mo from the nanoparticles. Depending on the type of target, different separation methods will be applied, including specific extracting agents, filtration and centrifugation. The eluate will be analysed by gamma spectroscopy and elemental analysis characterization techniques to determine the extraction yield and the specific activity. In a first step, molybdenum nanoparticles have been successfully generated by spark ablation using natural molybdenum electrodes and argon as carrier gas. We observed that the particle size mainly depended on the inert gas flow. At a argon flow of 1 L/min big agglomerates of more than 200 nm, meanwhile at 2 L/min the size of the agglomerates is decreased by a half. At 5 L/min the agglomerates, of smaller size, are mixed with small particles. When the gas flow is increased to 10 L/min now agglomerates are visible and only particles are produced.

ID: 1079

PRODUCTION OF ^{212}Pb TRACER FROM ^{232}Th AND ITS APPLICATION TO INVESTIGATE LEAD CHEMISTRY IN ACIDIC NITRO-PHOSPHATE SOLUTIONS

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Abstract Lead occurs in small amounts in the nature mainly as lead sulfide, however the main exposure and contamination are largely results of increased anthropogenic use (e.g. mining, refining, lead-acid batteries, paints, gasoline). Lead is a cumulative toxicant, and extensive use causes environmental contamination and health problems. The recommended limit for lead in drinking water is 10 ppb (WHO). Therefore, removal and recovery of Pb from industrial solutions is very important. In this study, the extraction chemistry of Pb from high acidic and high saline solutions are being studied. To achieve this, one goal is to produce ^{212}Pb tracer to determine the speciation of Pb in high acidic saline solutions. For this purpose, a lead generator system is developed based on the principle of mother-daughter relationship. In the first part, the thorium is separated from its daughters by using D2EHPA in benzene. After appropriate time, ^{224}Ra and its daughters are extracted from the organic phase with 0.1 M HNO_3 and loaded on an AG 50W-8X cation exchange column. The ^{212}Pb can subsequently be eluted quantitatively from the column with 1 M HNO_3.

6. Radiation Chemistry

ID: 0900

STUDY OF THE RADIATION INFLUENCE ON THE CHARACTERISTICS OF SPECIALIZED CONCRETES USED IN THE CONSTRUCTION OF NUCLEAR POWER PLANTS

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In the design and construction of nuclear power plants, a number of specialized concretes are used, such as TsKS, TsKS-M, OKA and OKA-M. However, concrete properties and project strength characteristics may change during nuclear power plant operation (design value is 60 years) under the radiation influence. Therefore, the aim of this work is the study of characteristics of specialized concrete under the radiation influence. The maintaining of the properties and characteristics of concrete specified in the design of nuclear power plant directly affect the justification of NPP safety and, accordingly, the NPP accident (severe and beyond design basis accidents) propagation paths. The test samples of concrete were produced and the drying modes were worked through as a result of the experimental studies. Samples were irradiated in the IRT-T research reactor. The properties of concretes are determined: - compressive resistance; - the amount of free and combined water in concrete; - density, porosity, water absorption; - thermal conductivity compressive resistance after thermal influence. The studies of concrete samples with OS-51-03 coating were singled out as a separate series of experiments. Organosilicate coating OS-51-03 is widely used in the design and construction of nuclear power plants in order to protect against radiation. Coating with an organic composition (OS-51-03) of concrete samples, and their subsequent heat treatment leads to a significant increase in compressive strength for all concrete compositions. The increase in strength in the low-temperature range from 200 to 600 °C is associated with the sealing of the sample surface during the drying of the organic composition and the creation under the impermeable film of conditions close to the conditions of heat and moisture treatment (steaming), which significantly intensifies the synthesis of the main phases of cements, providing an increase in strength. Further, with raise of temperature, complete burnout of organics from the organic composition occurs. Thus, can be said about the prolonged maintaining of the concrete properties (tensile strength) when exposed to temperatures up to 600 °C.

ID: 0908

POLY(HYDROXYETHYL METHACRYLATE) AS A HOST MATRIX FOR THE RADIOLYTIC SYNTHESIS OF HYDROGEL – AU(0) NANOCOMPOSITES

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As a part of a project aiming at the post-fabrication modification of 3D-printed objects by radiation processing, we are currently investigating the formation of gold nanoparticles by irradiating poly(hydroxyethyl methacrylate) (PHEMA)-based hydrogels, swollen with aqueous Au(III) solutions. The radiolytic behavior of PHEMA has apparently not been deeply investigated nor reported in the literature, in spite of its quite common uses in many biomedical applications. We have therefore examined in some detail the radiolytic behavior of commercial linear PHEMA samples or of covalent networks (PHEMA-networks) prepared by photopolymerization of HEMA in presence of (ethyleneglycol)dimethacrylate (EGDMA). Various spectroscopic and analytic methods were used to monitor the effects of electron beam radiation to assess PHEMA's reactivity during irradiation in the solid amorphous state for doses ranging from 5 to 100 kGy. Intrinsic viscosity measurements and molecular weight (MW) determination using size exclusion chromatography (SEC) confirmed the propensity to undergo chain scission, a common trend observed with poly(methacrylates) submitted to irradiation. The radiolytic yields for cross-linking and scission were assessed. Spectroscopic data allow to propose a reasonable mechanism accounting for these observations. EB-irradiation (10 – 100 kGy) of poly(HEMA) networks with various initial cross-link densities was shown to have a moderate impact on the swelling characteristics and thermo-mechanical properties of the materials treated in the dry or swollen state. However, gravimetric measurements revealed that irradiation induces the formation of extractables. Different approaches were explored to synthesize the nanocomposites with inclusions of gold nanoparticles: (i) direct Au(III) reduction within the matrix at various doses and (ii) pre-irradiation of dry or water-swollen disks followed by soaking in Au(III) solutions. The efficiency of Au(III) reduction was evaluated by UV-vis spectroscopy. These results confirmed that in spite of trend for main chain scission, direct Au(III) reduction by exposing the swollen PHEMA-based matrices could be achieved at doses where network degradation is kept at a very low level. The efficiency of Au(III) reduction was confirmed and quantified by UV-vis spectroscopy. The effect of network mesh size which is dependent on the content in cross-linking agent (0.5, 1, 2 and 5 wt-% of EGDMA) was shown to modify the UV-vis. spectra of the nanocomposites, presumably by controlling the nanoparticle size via the diffusion rate of Au(III) ions, of reducing species and/or of nanoparticles formed upon radiolytic synthesis. Sub-millimeter 2D PHEMA-based patterns were successfully converted into nanocomposites using the direct reduction method within the hydrogel matrix.

ID: 0937
IMPACT OF ELECTRON DENSITY ON REACTIONS AND STABILITY OF ONE-ELECTRON OXIDIZED PHENYL SULFONATES IN AQUEOUS SOLUTION

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Presently transportation is responsible for nearly a third of greenhouse gas emissions worldwide and is one of the major contributing factors to global warming.[1] Renewable energy sources in combination with electrochemical devices for energy storage have partially been able to mitigate these problems. It is conceivable to replace traditional combustion engines with environmentally friendly fuel cell-based systems that directly convert the chemical energy stored in various fuels into electricity with negligible emissions. Sulfonated aromatic hydrocarbon-based ionomers are typical constituents of proton exchange membrane fuel cells (PEMFC). Widespread application is currently limited due to their susceptibility to oxidative degradation initiated by HO•, produced in the membrane and electrodes during operation.[2] Degradation involves the formation of highly reactive aromatic cation radicals.[3] These intermediates are Lewis acids that can undergo protolysis equilibria with water and form hydroxycyclohexadienyl radicals and protons. For a given pH, the position of this equilibrium depends on the electron density of the aromatic ring. An equilibrium on the cyclohexadienyl side will increase the degradation rate of the PEM under normal operating conditions. Reduction of the cation radical may lead to repair whereas reduction of the cyclohexadienyl radical will lead to irreversible damage. In our study, we chose simple alkylated phenyl sulfonates as representatives of PEMs. We were interested in how electron density influences the rates and mechanism of degradation and whether repair via one electron reduction is feasible. Therefore, aqueous solutions of model compounds were one-electron oxidized by hydroxyl or sulfate radicals and the reactions were followed by time-resolved UV-Vis spectrometry. These highly oxidizing radicals were produced via pulse radiolysis. Additionally, continuous irradiation in water with a ^{60}Co source provided radicals at a constant low concentration and allowed us to compare the oxidative stability of the model compounds at different pH values. The degree of degradation was quantified by UPLC-UV/MS. We also studied whether addition of a suitable reductant affected the yield of degradation. Our findings can be applied to the development of next generation PEMs.

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ID: 0985
RADIOLYTIC PRODUCT DISTRIBUTION IN SELF-IRRADIATED AND GAMMA IRRADIATION SOLID STATE $Ca^{14}CO_3$: NEW DATA

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Both auto-radiolysis and gamma irradiation induce transformations in solid powdered $Ca^{14}CO_3$. Liquid chromatography followed by liquid scintillation counting were used to identify eight compounds after aqueous dissolution of the irradiated samples. The distributions of the radiolytic products depend on the gamma dose or the auto-radiolysis time. Formic acid is the principal product, both originating from the CO_2^- ion radical. Secondary reactions produce other compounds such as glycolic, glyoxylic, acetic and malonic acids, formaldehyde and methanol, with the yields depending on the total dose.

ID: 1021
ACTIVATION OF NOVEL PRODRUGS BY X-RAYS AND GAMMA RAYS - TOWARDS COMBINED CHEMO- AND RADIOTHERAPY WITH LITTLE SIDE EFFECTS

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Combined external beam radiotherapy and chemotherapy are known to have much more favourable disease outcome due to synergistic working, e.g., DNA repair inhibition by drugs making ionizing radiation more potent. However, anti-tumour drugs are known to cause severe adverse effects since they attack healthy fast dividing cells besides cancer cells. To minimize the systemic toxicity of such chemotherapeutics, we designed a prodrug (also called caged drug), which becomes active only upon removal of a protecting group, triggered by external ionizing radiation. Using such a strategy, the drug toxicity to healthy tissue is greatly reduce, while still achieving high killing efficiency to tumour cells. To proof this concept, a fluorescent probe, 4-methyl-7-hydroxycoumarin, was used as the reporter instead of the drug, to enable easy evaluation of the working mechanism. The hydroxy group of the probe was protected by an aryl boronate ester based self-immolative linker through a carbonate bond, and thus the fluorescence is largely quenched because of the electron withdrawing property of the latter. The aryl boronate ester can be oxidised by hydrogen peroxide which is generated by the radiolysis of water, leading to the release of the reporter (the fluorescent probe). When this compound was irradiated by X-ray or gamma radiation, a significant increase of the emission intensity was observed, which demonstrates that the protecting group was successfully removed. More importantly, an increase of fluorescence emission intensity is already detected when the probe solution is exposed to just 2 Gy of radiation, which is typically used in external beam therapy. After confirming the release of the reporter triggered by ionizing radiation, we replace the fluorescent probe by a widely used anti-tumour drug, doxorubicin. The toxicity of this prodrug appeared to be 10 times less than free drug, and the release of doxorubicin after irradiation was successfully detected by liquid chromatography – mass spectrometry (LC-MS). Currently we are conducting cell experiments to test the killing efficiency of this prodrug when exposed to ionizing radiation and determine its potential in combined chemo- and radiotherapy.

ID: 1022

RADIATION-CHEMICAL CONVERSION OF CELLULOSE

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There was studied the conversion of cellulose into liquid and gaseous products in the high-temperature range under the influence of ionizing radiation. There was found that at high temperatures, the yield of gaseous products prevails at the values of the absorbed dose D≤10 kGy, and at higher doses, the yield of liquid and solid phase products prevails. This is due to the processes of dimerization or trimerization of high molecular weight radicals formed in the environment. A comparison of thermal and radiation-thermal processes shows that the output under the influence of ionizing radiation is higher, increasing the rate of conversion by up to 60 %.

ID: 1023

TRANSFORMATION OF N-HEXANE ON THE SURFACE OF INITIALLY MODIFIED NANO-AL_2O_3 SURFACE OF THE CATALYST IN AN OXYGEN MEDIUM UNDER THE INFLUENCE OF GAMMA RAYS.

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Initially, the catalyst was modified in an oxygen medium under the influence of surface radiation. The conversion processes were compared on the surface of compacted and unmodified catalysts, in the range ΔT = 380 ÷ 420 °C, the yield of gaseous and liquid products from the conversion of hexane in a mini-flow reactor was studied. The output of the products was monitored with IR and UV spectrometers and gas-liquid chromatographic devices. There was found that as the surface temperature of the catalyst increases, the yield in liquid products decreases by 14 %, and the yield of gaseous products increases by 20 %. Due to the additional anion centers (O-ads) formed on the surface of the oxygen-modified Al_2O_3 catalyst under the influence of radiation, the volume yield of oxidation products is higher and has a lower activation energy (ΔEin = 11.27 kCol / mol.).

ID: 1035

CHEMICAL EFFECTS OF IONIZING RADIATION ON MOLTEN SALT SYSTEMS

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Ionizing radiation in the molten salt reactor environment will drive chemical changes in the composition of the salt and induce additional pathways for corrosive degradation of reactor components. Although the products of radiolysis are largely expected to recombine in the high-temperature molten salts, even a small mechanistic leakage out of the recombination cycle will result in substantial formation of permanent products, such as halogen gas (F_2 or Cl_2 depending on the salt) or metal particles that may deposit on surfaces in the reactor circuit. Thus, it is important to understand the initial products of molten salt radiolysis and their reaction kinetics with solutes including fission products. MSR fuel and corroded infrastructure materials are full of potential scavengers for these initial products. The goal of the US Department of Energy Office of Basic Energy Sciences Energy Frontier Research Center for Molten Salts in Extreme Environments is to provide fundamental understanding, based in atomistic level descriptions, of molten salt bulk and interfacial chemistry including the effects of solutes, impurities, and intense ionizing radiation fields. State-of-the-art steady-state and pulse radiolysis techniques have been deployed to understand the radiation-induced chemistry of molten chloride salt systems and to develop a predictive multi-scale radiation chemical kinetics model. A brief summary of the experimental and modelling and simulation results will be presented.

ID: 1040

ONE-POT SYNTHESIS OF POLYMERIC HYDROGEL WITH ALBUMIN NANOPARTICLES

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Applications of polymeric hydrogels in the biomedical field include contact lenses, artificial corneas, wound dressing, coating for sutures, catheters, and electrode sensors. Polymeric hydrogels have the property of absorbing and retaining different amounts of water due to their three-dimensional networks constitution. These hydrogels can be produced by chemical polymerization or by utilization of ionizing radiation technique which the main advantage is the absence of any chemical initiator Hydrogel dressings have shown very interesting properties as temporary skin cover for wound and burn healing, pain relief, moist environmental maintenance, a barrier to microorganism contamination, oxygen access in the injured area. Polymeric hydrogels are composed of hydrophilic polymers networks and water. They are very viscous aqueous systems or even solid systems. They find multiple applications in medicine among other fields. Wound dressings based on hydrogels have been prepared by a technology based on Rosiak's process (one-pot crosslinking and sterilization). Our group, however, develop an original process to synthesize metallic nanoparticles in a simultaneous way for the crosslinking and sterilization of hydrogels. The radiation-induced crosslinking of Albumin and Papain by intramolecular and intermolecular crosslinking was also developed to improve its biological stability aiming for its use on dressings and radiopharmacy carriers. This review will discuss aspects of radiation-induced synthesis and its application in the medical field.

ID: 1045

INVESTIGATING THE MECHANISM BEHIND THE RADIATION-INDUCED PHOTOCHEMICAL PROPERTIES OF PHOTO-ACTIVE SUBSTANCES

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Photodynamic therapy (PDT) is a clinical treatment that uses light to activate a photosensitizer which produces reactive oxygen species (ROS) able to destroy tumor cells. Photosensitizers are nontoxic in dark until they are activated by a specific wavelength of light and then generate different ROS, such as singlet oxygen $1O_2$, superoxide anion ($\cdot O_2^-$) and hydroxyl radicals($\cdot$OH), which are quite cytotoxic to tumor cells. However, PDT has several drawbacks such as low yield of ROS and short tissue penetration depth of light which reduces the treatment effectivity. Similar to photosensitizers, photocatalysts such as TiO_2 and ZnO are also able to generate ROS when exposed to light. It has been recently suggested that such photocatalysts can also be activated by ionizing radiation and exert anti-tumour effect which is very promising because this implies that there is much less penetration issues allowing treatment of deeper situated tumours. In this study, we attempted to determine the photochemical mechanism of TiO_2 and ZnO NPs when exposed to ionizing radiation. First, we systematically studied the interaction between ionizing radiation, such as X-rays and gamma-rays and photocatalysts like TiO_2 and ZnO in aqueous suspensions. Two fluorescence probes SOSG (specific for detection of O_2) and APF (specific for detection of $\cdot$OH) were applied to evaluate the ROS generation after radiation exposure. According to our results, ionizing radiation can strongly activate the photocatalytic properties of both TiO_2 and ZnO since dramatic increase could be seen in the yields of ROS compared to water alone. However, TiO_2 and ZnO nanoparticles exhibited quite different photochemical response upon radiation exposure. TiO_2 could mainly generate O_2 either under X-ray or gamma-ray radiation while ZnO nanoparticles mainly generated $\cdot$OH. Further attempts were made to figure out the dominant species which are responsible for the increased generation of ROS. Different scavengers were applied to quench different species contributing to the generation of ROS. Research so far demonstrated that O_2 played the most important role in generating O_2. More efforts will be devoted to further clarify the generation process of interesting ROS and to determine the photocatalytic effect of different radiation types.

ID: 1046

RADIOLYTIC ALTERATION OF ORGANIC MATTER IN URANIFEROUS ENVIRONMENTS STUDIED BY ARTIFICIAL IRRADIATIONS

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The natural organic matter present in uranium deposits (coal, amber-like fossil resins, bitumen) plays an important role in the transport, accumulation and mineralization of uranium in sediments, and it is also a sensitive marker allowing characterization of the depositional and diagenetic history of sedimentary formations. The energy transferred by ionizing radiation to organic matter can induce a complex series of reactions generating and altering organic matter. The radiolytic alteration is usually interpreted as a process spatially localized within reach of the emitted alpha particles, observed as, e.g., typical halos around uranium mineral inclusions. Structural changes observed in the bulk organic matter beyond the range of the alpha particles may be ascribed to fine dispersion of uranium and its daughters within the organic matrix, or by radon diffusion. Despite lower ionizing efficiency of beta and gamma radiation produced in the uranium decay chain, their radiation effects caused by interaction with natural organic matter may participate in mild structural alterations observed beyond the reach of alpha particles. To simulate and study natural irradiation of organic matter associated with uranium mineralization, artificial irradiation of model samples was carried out with 5-10 MeV electrons accelerated at the MT-25 microtron, and with 4.8 MeV He^+ ions accelerated at Tandetron 3140 MC accelerator. The effect of increasing cumulative dose was studied. Irradiation was followed by FTIR analysis of original and irradiated samples. For electron-irradiated samples, GC/MS analyses of sample extracts and gases released from samples upon irradiation were carried out. The observed chemical and structural changes were compared with alteration observed in the natural organic matter with uranium mineral inclusions.

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ID: 1063

STUDY OF CELL PROTECTIVE EFFECTS OF ALCOHOLS AGAINST UV-C RADIATION

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In previous study of the effects of irradiation on Escherichia coli DBM 272 bacteria, we found that simple alcohols such as methanol or ethanol decrease the radiation sensitivity of cells towards ionizing radiation, probably by scavenging OH radicals in the irradiated system. A similar effect of decrease in radiation sensitivity occurred when the system was irradiated with UV-C (254 nm) radiation. However, OH radicals are unlikely to form in sufficient amounts in aqueous solutions under UV-C radiation and the mechanism of radical scavenging should not play a significant role in affecting radiation sensitivity. Therefore, the protective effect of alcohols was studied in connection with singlet oxygen being produced in the UV-C irradiated system with Rose Bengal photosensitizer added. A singlet oxygen production was monitored using a Singlet oxygen sensor green chemical probe; reaction of chemical probe with singlet oxygen produces a fluorescent endoperoxide. Adding ethanol to the irradiated system resulted in decrease of the fluorescence signal, which indicates a decrease in concentration of singlet oxygen formed under UV-C irradiation. Thus, ethanol is likely to quench singlet oxygen in a system under study. This quenching does not occur with the use of methanol. When irradiating E. coli cells in the presence of ethanol and Rose bengal for higher singlet oxygen production, there was a greater reduction in the radiation sensitivity of the cells compared to the system without Rose bengal. Higher concentration of ethanol caused greater protection of cells. Thus, it is likely that ethanol can scavenge singlet oxygen and thus increase the protection of bacteria from the effects of UV-C radiation. However, the comparison of protective effect of various alcohols in the field of ionizing and non-ionizing radiations requires furthermore detailed and systematic study.

ID: 1066

DOSIMETRIC CHARACTERISATION OF SCIOX BEAM, AN X-RAY CABINET FOR EXPERIMENTAL RADIATION CHEMISTRY

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Cabinet X-ray instruments are used in various fields. They are primarily used for security screening or industrial quality control. Security applications comprise airport baggage security screening, cargo inspection of trucks crossing international borders and food inspection to check for foreign objects; industrial applications often involve circuit board inspection to find defects or tire inspection to identify manufacturing defects. Many X-ray cabinets are also used for medical applications or research. X-ray cabinet SCIOX Beam is a custom-made research device newly installed at the Department of Nuclear Chemistry at the Faculty of Nuclear Sciences and Physical Engineering of the Czech Technical University in Prague. This device is equipped with two wide-angle tungsten X-ray tubes with variable voltage and current settings, allowing irradiation by variable X-ray spectra. The low-energy X-ray tube works in a high voltage range of 4 – 60 kV with an output current of up to 3 mA (output power 0,4 – 50 W). The high-energy X-ray tube works in a high voltage range of 50 - 350 kV with an output current of up to 30 mA (output power 5 – 4200 W). The lead-shielded irradiation chamber has rather large dimensions of 750 x 750 x 720 mm and includes a movable shelf controlled by the machine software. In addition, there is a switchable light source, a camera, a temperature sensor and a computer-controlled ventilating fan in the chamber. A scintillating plate can be inserted into the chamber to check the width and extent of the X-ray beam on the current shelf position. The main goal of the presented work is the dosimetric characterisation of beams generated by the X-ray cabinet SCIOX Beam. It involves a determination of the dose rate at different sample positions in the cabinet and its relation to operational parameters of the X-ray tube(s) – voltage and current. The dose rate was determined by a Fricke dosimeter for different source-to-surface distances (SSD). The linearity of dose to irradiation time and repeatability was also evaluated. All such parameters are of utmost importance for any radiation-chemical evaluations using this new cabinet X-ray apparatus.

7. Radionuclides in the Environment, Radioecology

ID: 0880
LASER SPECTROSCOPY, INSTRUMENTAL NEUTRON ACTIVATION, AND MASS SPECTROMETRY TRACE ANALYSIS

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Among the modern analytical methods, laser spectroscopy, Instrumental Neutron Activation (INAA), and mass spectrometry analysis are the leading techniques for the detection of trace amounts of different isotopes in complex matrices providing the breadth of information about the elemental and isotope composition[1-10]. We report on chemiluminescence of plutonium, uranium, and samarium in solutions excited by laser radiation. The details of multi-step excitation of species and time-resolved detection[1-4,8] of resulting luminescence (Time Resolved Laser Induced Fluorescence – TRLIF) and chemiluminescence (Time Resolved Laser Induced Chemiluminescence – TRLIC) are considered. In the next step, we combine the atomic laser spectroscopy with mass spectrometry detection (Resonance Ionization Mass Spectrometry – RIMS). The high sensitivity has been demonstrated for krypton isotopes (including ^{81}Kr) of radiogenic (nuclear power plants) and cosmogenic (meteorites and other extraterrestrial material) origin[5,6]. Several multi-step RIMS approaches have been extended to uranium and other radioisotopes from solid and liquid samples[7,8]. The development of a suitable excitation/ionisation schemes for both TRLIF/TRLIC and RIMS is of a high priority allowing more complex sample characterisation. We have applied both INAA and Inductively Coupled Plasma Mass Spectrometry (ICP-MS) methods and analysed the elemental composition (64 elements) of bones of dinosaurs, South mammoths, prehistoric bear and archanthropus as well as the samples of surrounding soils; everything collected in different parts of Uzbekistan[9,10]. A high concentration of uranium we detected in the bones of dinosaurs (122 mg/kg), South mammoth (220 mg/kg), prehistoric bear (24 mg/kg) and archanthropus (1.5 mg/kg) compared to surrounding soils (3.7-7.8 mg/kg) and standard bones (<0.01 mg/kg) is a bit of a puzzle[10].

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ID: 0881
AIR RADIOLOGICAL MONITORING IN NORTHERN ALGERIA: SPATIAL DISTRIBUTIONS, METEOROLOGICAL INFLUENCE AND AIR MASSE’S ORIGIN

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AbstractBy its geographical position, Algeria in general and its Northern part in particular is not far from any radioactive contamination originating from nearby countries, especially industrial ones. The activity concentrations of natural radionuclides (namely ^{210}Pb and ^{7}Be) were measured by direct counting gamma spectrometry of particulate matter filters collected at two different altitude sampling sites in Northern Algeria. In particular, the two sampling sites were Algiers low altitude site , where the samples were collected over a four-year period (June 2014 - June 2018), and Chrea high altitude site , with sampling over a four month period (Nov. 2017-Feb. 2018). The activity of ^{7}Be shows high values in summer and low values in winter, while the seasonal ^{210}Pb activities increase during summer and decrease during winter and spring. Correlations between activity concentrations of both radionuclides, meteorological parameters and sunspots number (SSN) in Algiers were studied by simple regression and principal component analysis using Varimax rotation. These methods identified a negative correlation between ^{7}Be and SSN, while both radionuclides are negatively correlated with the amount of precipitations, as linked to the effect of wet deposition over the carrier-aerosol. ^{210}Pb and ^{7}Be levels observed at the two different altitude sites were compared during the period of contemporary sampling. Finally, the air masses sources and pathways responsible for high concentrations of ^{210}Pb and

^{7}Be at both sites were investigated through cluster analysis of HYSPLIT-4 back-trajectories.

ID: 0886

SELF-PREPARED STANDARDIZED NATURAL URANIUM ISOTOPES SOLUTION AND ITS USE FOR THE SORPTION EXPERIMENTS - PRACTICAL APPROACH

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Diversification of energy resources in order to tackle climate change and increase energy efficiency triggers a need to deploy more nuclear reactors worldwide, as a stable source of both electricity and heat. The leaching of radioactive elements to the surface waters and groundwaters due to the extraction of uranium as a key component of the nuclear fuel cycle from natural deposits causes an immense threat to the integrity of the environment and human health. Therefore a need to effectively tackle radiopollution in aqueous media by sorption processes seems desirable. Here we present a procedure to chemically prepare the standardized natural uranium isotopes solution from the pure, depleted uranium nitrate salt - $UO_2(NO_3)_2$ extracted from the Jachymov deposit (former Czecho-Slovakia). As prepared standard solution was implemented as a source of natural uranium isotopes for the sorption experiments onto synthetic zeolites. Alpha spectrometry measurements allow to precisely determine the activity of ^{238}U and ^{234}U isotopes before and after sorption experiments. The full agreement between the theoretically calculated radiochemical activities and practically measured ones resembles an affordable way to prepare natural uranium isotopes solution simultaneously allowing to omit formalities connected with cross-border and transcontinental trading and transportation of radioactive materials.

ID: 0887

ASSESSING LONG-TERM MICROBIAL IMPACT ON MINERALOGICAL TRAPPING OF URANIUM FROM DEEP GROUNDWATER AT ÄSPÖ HARD ROCK LABORATORY, SWEDEN

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Natural uranium (U) in deep groundwater has been extensively studied in connection to the search for suitable locations for final disposal of spent nuclear fuel (SNF). The U removal process depends on environmental and geochemical conditions and is often associated with fractionation of the main ‘stable’ isotopes, ^{238}U and ^{235}U (denoted as δ^{238}U), during reduction of hexavalent (U(VI)) to tetravalent (U(IV)) species. Thus, δ^{238}U serves as an important tracer for redox specific scenarios, in local to global temporal and spatial scales. In this contribution, one specific borehole setting built at 415 m depth into Paleoproterozoic granitoid rock at the Äspö Hard Rock Laboratory (HRL), Sweden, is investigated after 17-year experiment period (1995-2012). The HRL is constructed and operated by the Swedish Nuclear Fuel and Waste Management Co. and serves as a full-scale tunnel as a test-facility for the actual SNF repository to be built at Forsmark. We show how various analytical techniques, modelling, and isotope methods can be utilized to reveal U speciation and removal pathways, associated redox changes and related U isotope fractionations in the deep aquifer and during U mineralogical trapping. Synchrotron-based spectroscopic techniques reveal that calcite and Fe(II) sulfides precipitated on the borehole equipment contains intermittent highly elevated U, occurring as U(IV), and hence serves as a mineralogical sink for U. Thermodynamic modelling shows that aqueous Fe(II) is the main driving force for the reduction of U(VI) in the borehole water, alongside sulfides formed from bacterial sulfate reduction. The bacteria-driven degradation of technical polymer constituents present in the borehole equipment is central to processes forming the carbonates and sulfides that facilitate reduction of U(VI) and subsequent immobilisation of U(IV) into the calcite and Fe sulfides. We use δ^{238}U to show that U(VI) undergoes redox transformations in granitic rock aquifers, involving mineralogical and microbial pathways. The recorded δ^{238}U provides evidence for reductive removal of U(VI) from fracture water, presumably taking place along redox fronts in the fracture network. The obtained data on removal of U by calcite and Fe sulfides provide important insights for the

assessment of the geochemical behaviour of U and other redox-sensitive species in deep anoxic aquifers, that is relevant for trace metal mobility and long-term storage of SNF and nuclear waste.

ID: 0896

MULTI-METHOD INVESTIGATION OF EUROPIUM(III) COMPLEXATION WITH THE DECORPORATION AGENT HEDP OVER A WIDE PH RANGE

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In case radionuclides (RN) enter the food chain and are incorporated by humans, they pose a possible health risk due to their radio- and chemotoxicity. In case of such incorporation, HEDP (1-Hydroxyethylidene-1,1-diphosphonic acid; etidronic acid) and DTPA (diethylenetriaminepentaacetic acid; pentetic acid) are common decorporation agents for uranium and transuranium RN, respectively. Since HEDP also binds trivalent actinides, An(III), we investigated the complexation of HEDP with Eu(III) as a luminescent non-radioactive analog of An(III) at I = 0.1 M (NaCl) from pH 1 - 12 using a variety of spectroscopic and analytical methods. The pH-dependent ligand behavior was studied with infrared spectroscopy with attenuated total reflection (ATR-FT-IR) combined with density functional theory (DFT) and pKa values were determined by nuclear magnetic resonance spectroscopy (NMR). The Eu(III)-ligand system was, then, investigated by time-resolved laser-induced fluorescence spectroscopy (TRLFS), ATR-FT-IR, solution and solid-state NMR as well as mass spectrometry with inductive coupled plasma (ICP-MS) and with electron spray ionization (ESI-MS). Depending on both the pH and the metal to ligand ratio, several Eu(III)-HEDP complex species were observed and characterized within this study. Over a wide pH range and especially at physiological values, the complexation of Eu(III) and HEDP leads to the precipitation of hardly soluble species.

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ID: 0898

ANALYSIS OF THE ROLLED COTTON CLOTH FIXED ON THE OUTER SURFACE OF THE INTERNATIONAL SPACE STATION USING NEUTRON ACTIVATION ANALYSIS AND COMPLEMENTARY TECHNIQUES

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As part of the space experiment “Test”, a roll of cotton cloth fixed on the outer surface of the International Space Station for more than 10 years was delivered to the Earth in September 2019. The elemental composition of two fragments of the cloth, contaminated and clean, was determined using instrumental neutron activation analysis at the IBR-2 reactor. Along with 19 elements (Mg, Al, Cl, Ca, Ti, Cr, Mn, Fe, Co, Ni, Zn, Rb, Sr, Ag, Sb, Ba, Sm) determined on both fragments of the cloth, additional 20 elements (Na, Si, Sc, Cu, As, Se, Br, Mo, Zr, Cd, I, La, Ce, Eu, Ta, W, Re, Ir, Th, and U) were determined only on the contaminated fragment. The morphology of the rolled cotton cloth was characterized using Scanning Electron Microscopy. Gama and liquid beta spectroscopy were applied to measure the radioactivity of the cloth fragments. Possible sources of the determined chemical elements deposited on the cloth were discussed.

ID: 0902

SPECIATION AND TRANSFER OF RADIONUCLIDES IN THE HUMAN ORGANISM ESPECIALLY TAKING INTO ACCOUNT DECORPORATION AGENTS (RADEKOR) – A JOINT PROJECT

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In case radionuclides (RN) enter the food chain and are incorporated by humans, they pose a possible health risk due to their radio- and chemotoxicity. To precisely assess the health risk after oral incorporation of RN with food and beverages and to apply effective decontamination methods, it is mandatory to understand the processes of RN biokinetics on both cellular and molecular scale. Within the joint research project “Speciation and transfer of radionuclides in the human organism especially taking into account decorporation agents (RADEKOR)”, quantitative excretion analysis and biokinetic modeling of orally incorporated RN are performed. Additionally, these macroscopic investigations are combined with molecular speciation studies of RN in artificial fluids of the alimentary tract of humans and cytotoxicity studies with respective human and rat cell lines both in the absence and presence of decorporation agents. Aim of the project is to expand the knowledge of processes underlying RN interactions within the human alimentary tract on a cellular and molecular scale to establish a precise biokinetic model as well as to contribute to the development and improvement of nuclide specific decontamination methods.

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ID: 0910

UPTAKE OF Np(V) BY ZIRCONIA: A COMBINED BATCH, SPECTROSCOPIC, AND SURFACE COMPLEXATION MODELING STUDY

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The interactions of long-lived actinides, such as the transuranium element neptunium, with corrosion products in the near-field of a repository are important processes that have to be considered when assessing the safety of a nuclear waste repository. As a main corrosion product of the zircaloy cladding material of spent nuclear fuel rods, zirconia (ZrO_2) constitutes a first possible barrier against the release of radionuclides. To gain a detailed understanding of the surface processes in the Np(V)−zirconia system, a comprehensive, multi-method approach was applied. The Np(V)−ZrO_2 system has been studied on the macroscopic level by conducting pH-dependent batch sorption experiments under varying conditions (ionic strength, Np(V) concentration, and solid-to-liquid ratio (m/V)). In addition, a Np(V) sorption isotherm at pH 6 was collected. The results revealed that Np(V) sorption onto ZrO_2 was affected by pH, Np(V) concentration, and solid-to-liquid ratio. Uptake of Np(V) increased with pH, starting around pH 3 with maximum sorption reached from pH 6. The shift of the sorption edge towards lower pH with increase of the m/V ratio points to the presence of different kinds of sorption sites. This is supported by the Np(V) sorption isotherm results, where the shape suggests strong and weak binding sites. Furthermore, Np(V) uptake was found to be independent of ionic strength and zeta potential measurements revealed a shift towards higher pH values of the isoelectric point of the neat ZrO_2 in the presence of Np(V). Hence, the formation of Np(V) inner-sphere surface complexes is indicated. Molecular information about the surface species were obtained by Extended X-ray Absorption Fine Structure Spectroscopy (EXAFS) and in situ Infrared (IR) Spectroscopy, revealing the predominant formation of inner-sphere Np(V) surface complexes. A short Np−Zr distance derived from EXAFS suggests the presence of Np(V) bidentate inner-sphere complexes on the ZrO_2 surface. These information obtained on a macroscopic and a molecular level can be used to restrict the number of surface species as well as their denticity in a surface complexation model. The thermodynamic surface complexation parameters of the Np(V)−zirconia system derived in this study will help to make more reliable predictions about the fate of Np(V) in the environment.

ID: 0914

MULTI-TECHNIQUE STUDY OF THE COMPLEXATION OF Np(V)/Pa(V) WITH MELLITATE

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Activities related to nuclear energy are at the origin of a dissemination of actinides in the environment. The migration of actinides in the environment is strongly dependent on their speciation and consequently on the pH, the ionic strength, the redox potential but also on the presence of ligands which can promote their mobility. The An(V) ions are, in general, highly soluble leading to a high mobility in waters and are for some actinides (Pu, Np and Pa) the most stable oxidation state in natural waters. The knowledge of fundamental data, both thermodynamic and structural, on actinide complexes formed with ligands of interest is essential to improve models of actinide migration in the geosphere and biosphere. Among the pentavalent actinides, Pa(V) is the unique An(V) without actinyl moiety, and data on the protactinium chemistry remain incomplete. Thus, this study focuses on the behavior of Np(V) and Pa(V) in aqueous solution in the presence of mellitic acid, derived from minerals having organic origins. Mellitic acid is also used as a model ligand of natural organic matters. This is an aromatic acid with six carboxyl groups, that displays a high solubility in aqueous phase. The complexation of Np(V) was studied using ultraviolet visible near infrared spectroscopy (UV-VIS-NIR) and attenuated total reflection Fourier transform infrared (ATR FT-IR) at mM scale and pH =4 and

6. Because of the high tendency of Pa(V) to hydrolyze and polymerize at high concentration, its complexation with mellitic acid was carried out combining partition methods at ultra-trace level (liquid-liquid extraction) with γ-spectrometry measurements (^{233}Pa(V) total=10-10M, I=1.0M $NaClO_4$, pH=1). In the extraction experiment, the distribution coefficient D was determined in the system $NaClO_4$/$HClO_4$/ligand||TBP/Toluene, according to the ligand concentration. Two different behaviors have been observed for each An(V). By varying the mellitic concentration, a stoichiometry of 1:1 has been observed in presence of Np(V), whereas, for Pa(V), complexes of stoichiometry 1:1 and 1:2 were observed. The structure of the complexes of Np(V) and Pa(V) was investigated using x-rays absorption spectroscopy and ATR-FTIR spectroscopy. The coordination with carboxylate can be monodentate with one O atom of one COO-group, bidentate with both O atoms of the same one COO^- group (end-on), or monodentate with two O-atom of two neighboring COO^- group form chelate rings (side-on). For Np(V), the spectroscopic techniques seem to indicate that the coordination of the carboxylate groups from the mellitic ligand is in a bidentate fashion.

ID: 0923

EFFICIENT PHOTOREDUCTION STRATEGY FOR URANIUM IMMOBILIZATION BASED ON GRAPHITE CARBON NITRIDE HETEROJUNCTION NANOCOMPOSITES

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The conversion of soluble U(VI) to insoluble U(IV) is an economic strategy to efficiently remove U(VI) from radioactive wastewater by photoreduction. However, high photogenerated electron-hole recombination rate and low sunlight utilization are still huge challenges. Here, we designed a carbon nitride-based heterojunction composite material for photocatalytic reduction of U(VI) from aqueous solutions and real seawater systems under simulated sunlight/visible light. The construction mechanism of the heterojunction structure was further clarified through theoretical calculations, and the reduction fixation mechanism was explained with the help of a series of spectral evidence. This strategy of constructing a heterojunction structure effectively inhibits the recombination of photogenerated electrons and holes, and greatly prolongs the lifetime of carriers in the heterojunction system. Therefore, the g-C_3N_4/$LaFeO_3$ heterojunction exhibits high removal ability in a wide range of U(VI) concentration (460 mg/g). Although the construction of g-C_3N_4/$LaFeO_3$ heterostructure effectively alleviates the problem of rapid recombination of photogenerated electron-hole pairs, the visible light response is still not ideal. Therefore, to enhance the visible light response of the catalyst, we developed a carbon nitride/ceria (CN-CeO_2-x) heterojunction with a type II band structure rich in oxygen vacancies based on the heterojunction-defect synergistic modification strategy, used for photoreduction of U(VI) under visible light. CN-CeO_2-x heterojunction has excellent photoreduction ability of U(VI) (96.1 %), and it can maintain excellent synergistic removal efficiency even in the presence of organic matter that is not conducive to U(VI) removal. The catalytic efficiency of the two catalysts remained at a relatively high level after being used repeatedly for 5 times, indicating that the two photocatalysts have good cost-effectiveness and applicability. In short, g-C_3N_4/$LaFeO_3$ and CN-CeO_2-x heterojunction provide a promising strategy for using inexhaustible solar energy to recover uranium resources.

ID: 0924

ROOM-TEMPERATURE RN-222 ADSORPTION USING SILVER EXCHANGED ZEOLITES

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Radon-222 is a radioactive noble gas omnipresent in the environment. Being part of the ^{238}U/^{226}Ra decay chain, it has been found to considerably contribute to the radiation dose to the public. Exposure to ^{222}Rn at home and workplace has been extensively monitored and specific guidelines exist to mitigate exposure. ^{222}Rn has also gained considerable attention in particle physics research, where it is considered as unwanted contaminant in low-background environments. There has been extensive research on Rn removal since the discovery of its adsorption on activated charcoal by Rutherford in 1906 and numerous scientific publications exist on the adsorption of ^{222}Rn on carbon and on other materials. However, there is little development in the field of radon capture at room temperature and cryogenically cooled charcoal remains the material of choice as of today. We report here on the remarkable property of silver exchanged zeolites to efficiently adsorb radon at room temperature. We have investigated three commercially available zeolite materials and compared their adsorption performance to commonly available activated carbons by measuring ^{222}Rn breakthrough curves under different experimental conditions. Depending on the type of carrier gas and gas velocity, dynamic adsorption factors k were found to be at least two orders of magnitude higher for zeolite materials as compared to activated carbons. Moisture control was found to be crucial for effective ^{222}Rn adsorption since water has shown to exhibit a strong competitive effect. This contribution will present experimental results obtained from systematic investigations involving the practical usage of these materials. It is shown that more compact and economic room-temperature ^{222}Rn adsorption systems can be designed using silver exchanged zeolites considerably reducing the impact of ^{222}Rn originating from ^{226}Ra emanations in environmental, scientific and industrial applications.

ID: 0925

DEFINITION AND OPTIMIZATION OF A PROCEDURE FOR OBTAINING CROSSLINKED PLASTIC SCINTILLATION MICROSPHERES

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The measurement of alpha and beta radioactivity is something common in several fields (i.e. environmental monitoring, medicine, research…) although there is still a necessity for more environmentally respectful methods with a low generation of residues and new strategies for complex scenarios, like continuous monitoring or fast methods for the analysis of difficult to measure radionuclides (DTM). Following these aims, plastic scintillators in form of microspheres (PSm) has been proposed in the last years as an alternative to liquid scintillation. The use of plastic scintillators avoids the generation of mixed wastes, allow high selectivity in the determination of DTM radionuclides when these are combined with extracting agents (PSresins) as well as provide high viability for applications in continuous detection systems. Previous experience of the authors on PSm based on linear polystyrene and prepared by evaporation/extraction method has shown that PSm provide a high detection efficiency in the determination of alpha and medium or high energetic beta emitting radionuclides. However, these can be used in a framework of limited conditions since the polymer is soluble in organic solvents in which the PSm are dissolved or the fluorescent solutes leached. This situation can be solved by the use of microspheres including a crosslinking agent, divinylbenzene, which is copolymerized with styrene creating a crosslinked scintillating microsphere (CPSm). These CPSm allow, according to their structure of chains of polymer linked one to another in different points, to be used in aggressive conditions, such as organic matrices, opening the possibility to measure organic samples or treat it with organic solvents to modify its structure (addition of pores) or its surface (adding functional groups on its surface) creating advanced PS systems. The objective of the present study was to define and optimize a methodology to obtain CPSm by polymerisation of styrene and DVB, and to evaluate the morphological, resistance and scintillation properties of CPSm. The polymerization method used is a free radical polymerisation in a dispersant medium with the presence of a surfactant (PVA). Initially, the effect of the styrene-DVB ratio was verified. It was proved that a higher proportion of DVB produces a reduction in the medium size of the CPSm without significantly altering the radiometric properties. Values of detection efficiency are comparable to those PSm prepared by the evaporation/extraction method which demonstrate the low effect of crosslinking with DVB in the scintillation mechanism. In terms of resistance properties, melting point and extraction of fluorescent solutes encapsulated with different organic solvents was studied. Results shown that temperature resistance increases with the increase on the DVB proportions. Moreover, no morphological and scintillation properties changes were produced when the CPSm were treated with several organic solvents, highlighting the effect of the crosslinker agent. In contrast, it was determined that a high fraction of solutes encapsulated in CPSm were extracted by dichloromethane, acetone and toluene whereas in methanol and hexane the extraction was lower. As a conclusion, a procedure for the preparation CPSm has been developed. The CPSm present comparable scintillation properties to that of lineal polystyrene PSm but with higher resistivity to temperature and organic solvents which increases the ranges of application and enables further structural modifications.

ID: 0926

INVESTIGATING PO-210 AND PB-210 UPTAKE FROM COAL ASH AND SLAG BY EARTHWORM EISENIA FETIDA

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Natural radionuclides can be concentrated in coal ash and slag. When such a waste is deposited in the environment, it is normally inhabited by different animals and plants. Earthworms are one of the most exposed as they live in close contact with such a waste throughout their entire life. ^{210}Po and ^{210}Pb represent one of the most radiotoxic radionuclides, especially when ingested. Within this study we wanted to assess the uptake of ^{210}Po and ^{210}Pb by earthworm Eisenia fetida transplanted on the coal ash and slag sampled from the former chemical factory in Kaštela Bay (Croatia). Uptake was compared with cultivation on non-contaminated and reference soil. After cultivation period of 28 days, earthworm samples were cleaned. From one set of samples, which was used also for cytotoxic and genotoxic investigations, coelomic fluid was removed and from other not. They were dried at 65 °C. Then the samples were digested and Po was spontaneously deposited on Ag disc and ^{210}Po activity concentration was determined by alpha-particle spectrometry. ^{210}Pb was separated using Sr Resin column and measured by gas-flow proportional counter. Results show that ^{210}Po activity concentrations in earthworms cultivated on coal ash and slag were in the range of 43-93 Bq/kg, whereas on non-contaminated and reference soil values were in the range of 17-39 Bq/kg. Therefore, ^{210}Po activity concentrations found in earthworms cultivated on coal ash and slag are 2-3 times higher compared to control group. Similar observation was found for ^{210}Pb only for samples without coelomic fluid, where samples with coelomic fluid did not show any significant difference between those cultivated on coal ash and slag and control ones. This indicates different distribution of ^{210}Pb and ^{210}Po between coelomic fluid and other earthworm's body compartments.

ID: 0929

SPECTROSCOPIC INVESTIGATION OF THE SPECIATION OF URANIUM(VI) IN THE BIOFLUIDS OF THE HUMAN DIGESTIVE SYSTEM

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In case radionuclides (RN) enter the food chain and are incorporated by humans, they pose a possible health risk due to their radio- and chemotoxicity. Therefore, it is necessary to know the biokinetic processes as well as the speciation of the RN after ingestion in order to develop and improve specific methods for their decorporation. When RN are ingested orally, they first come into contact and interact with the biofluids of the digestive tract. However, for most actinides, little is known to date about their speciation and biochemical behavior in the gastrointestinal tract. Therefore, the aim of this study was to investigate the interactions of hexavalent uranium in the gastrointestinal compartments and respective biofluids in the stomach (saliva and gastric juice) and small intestine (pancreatic juice and bile fluid). The combination of these two segments was also considered. Biofluids and digestive steps were artificially simulated based on human physiology. The chemical speciation of uranium was investigated using cryogenic time-resolved laser-induced fluorescence spectroscopy (cryo-TRLFS) at 153 K. The results were compared with thermodynamic modeling. The TRLFS experiments show that uranium is predominantly complexed with inorganic ligands, mainly carbonate and to a smaller extent phosphate, as confirmed by thermodynamic modeling. For the organic ligands, only some proteins, e.g. prancreatin, are involved in the speciation to a small extent. Based on this knowledge, specific decorporation agents can be developed and their influence on uranium speciation can be observed using cryo-TRLFS.

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ID: 0931

GROSS-ALPHA PARAMETER DETERMINATION WITH A METHANDIPHOSPHONIC ACID BASED PLASTIC SCINTILLATION RESIN

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The assessment of the sample radioactivity content through global or screening parameters is of interest since it permits to obtain valuable information about the presence of radionuclides in samples. This strategy has the advantage that avoids the use of laborious selective procedures for each radionuclide which are usually long and complex, leading to the generation of more information in less time and with lesser costs. One of those parameters is the gross alpha. This parameter is measured for example in the characterization of radioactivity in water intended for human consumption. If the value of the gross alpha is under 0.1 Bq L^{-1} the water is considered drinkable and the specific analysis of several alpha emitters can be avoided. In another situation, in a nuclear dismantling process, screening parameters could be used to improve waste management as it permits to check easily if the sample can be treated as a conventional or as radioactive waste. Although there are several approaches to the measurement of the gross alpha parameter, there is still a need to simplify the determination by the use of faster methods which could also be capable to reduce the uncertainty associated to such determination. This work presents a new method to determine gross alpha by using plastic scintillation resin (PSresins) packed in a solid-phase extraction cartridge. One of the advantages of PSresin regarding other methodologies is that allows separation and measurement using the same material simplifying the analytical procedure. This can even be improved if the sample treatment is simply reduced to a valence state adjustment and the pass of the sample though the PSresin. In this work, a new selective PSresin for all actinides, radium and polonium has been developed using bis-(3-trimethylsilyl-1-propyl)-methanediphosphonic acid as an extractant. This new PSresin allows the determination of alpha emitting radionuclides with a detection efficiency above 95 %. All alpha emitters studied (^{241}Am, ^{238}Pu, ^{230}Th, ^{236}U, ^{210}Po and ^{226}Ra) presented quantitative retention in moderated nitric acid media (pH 2) and up to 400 mL of sample can be passed though the PSresin without retention being affected. Moreover, applying alpha/beta discrimination adequate separation from the beta emitters retained could be achieved (i.e. misclassification error around 15 % at PSA level 135). The analysis procedure proposed consisted just on passing directly though the PSresin cartridge 100 mL of sample treated previously during 30' at 50 ºC with 1 % of hydrogen peroxide as a valence adjustment agent. Sample were spiked with iron (III) or europium (III), working as tracers, for the posterior confirmation of the total retention of the alpha emitting radionuclides by a colorimetric visual check using thiocyanate or arsenazo (III) complexes respectively for iron (III) and europium (III). After that, the cartridge is directly measured in the scintillation detector for 2 hours with no further treatment. In this conditions the alpha parameter has been determined in several water samples in less than 4 hours from the sample reception with quantification errors lower than the ones obtained with the current used methods.

ID: 0935

IMPACT OF Eu(III) AND U(VI) AS WELL AS HEDP AND DTPA ON HUMAN AND RAT KIDNEY CELLS IN VITRO

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In case radionuclides (RN) enter the food chain and are incorporated by humans, they pose a possible health risk due to their radio- and chemotoxicity. When RN incorporation occurs, decorporation agents (DA) play a critical role in health maintenance and reduction of toxicological damage. Since excretory organs are highly exposed to incorporated RN, we performed in vitro cell culture experiments with renal cells from human (HEK-293) and rat (NRK-52E) and investigated the effect of U(VI) and Eu(III) as non-radioactive analog of trivalent actinides onto these cells. Furthermore, also the effect of the common DA HEDP (1-Hydroxyethylidene-1,1-diphosphonic acid; etidronic acid) and DTPA (diethylenetriamine-pentaacetic acid; penteteic acid) was studied. In exposure experiments, cells were incubated with Eu(III), U(VI) or DA for 8h, 24h and 48h, respectively. Applied Eu(III) and U(VI) concentrations ranged from environmentally relevant trace concentrations up to millimolar solutions near the solubility limit. HEDP and DTPA were applied in the same concentration range, including recommended therapeutic dosages. The cell viability was measured using the XTT assay and dose response curves were determined for each exposure time and cell line. Microscopic investigations of cell morphology were performed to study possible alterations of exposed cells. In addition, ICP-MS analyses were conducted to determine Eu(III) and U(VI) solubility in the cell culture medium. Cell viability studies reveal a concentration- and time-dependent effect on both cell lines for Eu(III) and U(VI) as well as for HEDP and DTPA. This enables the calculation of EC_{50} values from the dose response curves and comparison with literature values for other heavy metals and DA. Microscopic investigations reveal morphological alterations of the cells upon exposure. ICP-MS results indicate high solubility and, thus, high bioavailability of Eu(III) and U(VI) in cell culture medium.

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ID: 0946

THE DETERMINATION OF RADIOSTRONTIUM, Cs-ISOTOPES AND Pu-ISOTOPES IN TYROLEAN FOODSTUFF AND ENVIRONMENTAL SAMPLES

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In case of emergency situations a specific sampling plan exists in Austria. During regular emergency exercises, samples are taken according to this sampling plan. Thus, during an emergency exercise in Tyrol, various foodstuffs and the environment were sampled. Therefore, these samples were taken into account for the determination of $^{239/240}$Pu, ^{90}Sr, ^{134}Cs and ^{137}Cs. Surface water, raw milk, radish, leeks, kohlrabi and feedstuff were part of the analyzed samples. These samples can be used to give additional data to the Austrian monitoring program conducted according to the commission recommendation 2000/473/Euratom. The analytical method used in this project was a combined approach for the simultaneous determination of ^{90}Sr and Pu-isotopes. It was based on the selective separation by ion exchange chromatography (DOWEX®) and extraction chromatography (Eichrom Sr-Resin) followed by liquid scintillation counting for ^{90}Sr and alpha spectrometry for the Pu-isotopes. The counting sources for the alpha spectrometric measurements were prepared by coprecipitation using Nd as carrier. In order to monitor the chemical recovery of the analytes ^{242}Pu and natural $^{86/88}$Sr were added as tracers. Whereas the ^{242}Pu tracer was measured in the end by alpha spectrometry, the content of the Sr-tracer was monitored by ICP-MS. Using this separation method good chemical yields for Sr of about 90 % and for Pu above 70 % were observed. Further results of this project will be presented at the conference.

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ID: 0947
DETERMINATION OF RADIOCARBON ACTIVITY IN MIXED DIET VIA INDIRECT MEASUREMENTS

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This contribution describes an alternative mean how to perform monitoring of ^{14}C activity in mix diet via indirect measurements, i.e. via analyses of other sample kinds with carbon isotopic composition linked to the radiocarbon composition of the individual diet. This research was launched due the European Commission Recommendation 2000/473/Euratom, requesting monitoring of ^{14}C activity in a sparse network in a mixed diet. This kind of monitoring is not being performed yet in the Czech Republic. Aim of the research was to find a simple and effective procedure, how to perform determination ^{14}C activity in the mixed diet. Monitoring of ^{14}C activity in mixed diets by sampling and analysing the mixed diet itself appears impractical due to inhomogeneity if such kind of samples. There are different types of samples, various kinds of human skin derivatives, that reliably represent the carbon isotope composition of the human mixed diet. Such samples, allowing uncomplicated sampling, are naturally robust against inhomogeneity and easy to analyse. Human hair were chosen as an ideal type of sample for this purpose. A preliminary monitoring of ^{14}C activity in mixed diet was performed simultaneously by both direct and indirect measurements. Our contribution will contain summarization of yielded results, evaluation of the relation between the composition of mixed diet and human hair, and proposals, how to exploit reached knowledge for practical ^{14}C in mixed diet monitoring.

ID: 0948
RHIZOSPHERE INTERACTIONS IN RADIONUCLIDE SPECIATION, TRANSFER AND PLANT-UPTAKE – IMPACTS ON SPENT NUCLEAR FUEL DISPOSAL AND MINING AREAS

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Radionuclides may have damaging consequences for ecosystems and may present variable threats for biota, if released from repository or mining areas. In soils, bioavailable radionuclides may affect adversely to microbiological activity and vitality, cause changes in community structure and inhibit the growth of microorganisms. On the other hand, soil microbiota can modify soil chemical environment and thus alter the mobility and uptake rates of radionuclides into plant tissues. These modifications may occur through various mechanisms including biosorption and –accumulation, bioprecipitation, oxidation and reduction reactions as well as through complexation on excreted ligands. Moreover, soil microorganisms may also prompt mobilization of radionuclides through acidic extracellular metabolites thus potentially increasing their uptake and transfer to plants. Considering the effects of soil microorganisms on the behaviour of radionuclides originating from spent nuclear fuel in acidic bog environments, we have observed that the highly diverse microbial communities inhabiting the deep anoxic soil layers endure in high metal concentrations and may concurrently contribute to the total retention of radionuclides[1]. Microbiota dwelling in this habitat contribute to radionuclide mobility through several distinctive mechanisms including redox reactions (Se(IV))[2], direct biosorption (Ni(II), Ag(I), Cs(I))[1,3-5] and extracellular enzyme activity (I(-I))[6]. Similarly, in a former uranium mine area (Paukkajanvaara) we also observed several bacterial groups adapted to the elevated radiation levels[7]. We also observed certain bacterial strains (Pseudomonas sp.) to take up radium and to concurrently increase sulfate concentrations in the leached solution. Rhizospheric root-microbe interactions are of key importance to the terrestrial ecosystems. Certain soil bacteria (e.g. Pseudomonas sp. T5-6-I) can influence radionuclide plant-uptake (e.g. transfer of ^{75}Se to Brassica oleracea and Arabidopsis thaliana), and to produce changes in root morphology as well as in protein expression[8]. Although generally Se(IV)-->Se(0) reducing, we have shown Pseudomonas sp. T5-6-I to promote Se(IV) transfer to plants by a yet unknown mechanism[8]. However, Se(IV) taken up by the plant appears to end up in organic C-Se-H and C-Se-C bonds irrespective of the presence of bacteria[8]. Regardless of the evident importance of rhizospheric interactions, they are often overlooked due to the difficulty to image the root-microorganism-system. However, the emergence of new imaging techniques, such as state-of-the-art high-resolution synchrotron methods, have enabled more detailed imaging and better understanding of these interactions[9].

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ID: 0951

METHODS FOR THE DETERMINATION OF BIOMASS CONTENT IN SOLID RECOVERED FUELS

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The European Union has a long-term objective to reduce greenhouse gas emissions causing global warming. In 2005, the so-called Kyoto Protocol entered into force, which introduces a system of emission allowance (EUA) that creates gradually increasing financial pressure on companies emitting CO_2. This agreement distinguishes between carbon, which comes mainly from the combustion of fossil fuels (fossil carbon), and so-called biomass carbon, which is defined as carbon occurring in all material of biological origin. If the company proves that it has combusted biomass carbon, then the portion of CO_2 produced in this way is removed from the EUA. Methods for carbon characterization are described in the standard ČSN EN ISO 21644: 2021 Solid recovered fuels – Methods for the determination of biomass content, which came into force in August 2021. There are reported three options for determination of biomass content: Selective dissolution method (SDM), Manual sorting method (Msort), and method based on determination of ^{14}C. SDM and Msort have quite a lot of limitations for example higher content of rubber or plastics or small particle size. On the other hand, ^{14}C method is the most universal, however, it has requirements for high-tech measuring equipment. Central analytical laboratory - testing laboratory (CAL-TL) in ÚJV Řež a.s. has been performing an accredited determination of biomass carbon content since 2009. The method is based on the determination of ^{14}C content in CO_2 from combusted solid recovered fuel (SRF) sample using liquid scintillation counter (LSC). The tube furnace consists of the 1st zone where 2 g of sample is placed and the 2nd zone where the CuO catalyst is located, which ensures complete oxidation of the flue gases. The temperature in the area with the catalyst is constant at 800 °C, while in the 1st zone the temperature gradually increases up to 1000 °C. A mixture of O_2 and N_2 gases flow through the whole system. The O_2 content raises with increasing temperature up to 100 % oxygen atmosphere. The flue gases are then purged in several bubblers and the CO_2 is trapped into ice-cooled amines-based trapping solution. The trapping solution is then weighted into a measuring plastic vial and mixed with scintillation cocktail in ratio 1:1. A coal sample is combusted together with each set of samples, which represents a pure fossil source and therefore zero value of biomass carbon. On the other hand, 100 % biomass carbon content is represented by the oxalic acid standard, which is also combusted with each set of samples. The samples prepared in this way are then measured by low background LSC for 24 hours each. Demand for the determination of biomass content will undoubtedly increase as rising EAU costs and energy companies switch from traditional fossil to alternative sources.

ID: 0954

THE INTERACTION OF ILLITE COLLOID AND Eu(III) UNDER ENVIRONMENTAL CONDITIONS: ROLE OF CHARGE HETEROGENEITY

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Radionuclides with high radiotoxicity and long half-life derived from nuclear industry activities would inevitably be introduced into the soil or groundwater. In order to propose a safe and efficient method to control the migration of radionuclides, it is of great significance to understand the environmental behavior of nuclides. The laboratory experiments and field studies have proven that colloids play a very important role for the transport of radionuclides in aquifer environment. Therefore, in this work, the typical environmental clay mineral colloid-illite colloid, was used to study the interaction between colloid and Eu(III) under environmental conditions. The transport behaviors of illite colloid in saturated quartz sand column is completely dependent on the stability of the colloid. Illite colloid could complete breakthrough the quartz sand due to the high stability at high pH, low ionic strength and presence of HA, and the colloid could be blocked at low pH and high ionic strength; The transport behavior of Eu(III) is completely simultaneous with the colloids during the co-transport, that is, essentially depends on the stability of the colloid. Illite colloid faciliated the transport of Eu(III) under the conditions favorable to the stability of colloid, otherwise, the transport of Eu(III) was retained. Nevertheless, the retention of the colloid and Eu(III) was reversible, the blocked colloid and Eu(III) could be released and re-transported when the environmental chemical conditions became be favorable for the dispersion of the colloid. However, the real environmental media is not the same as the ideal quartz sand media. Iron and aluminum oxides usually adhere on the media surface, resulting in nonuniform surface charge. The co-transport results of colloid and Eu(III) in the charge heterogeneous media show that the co-transport behavior of colloid and Eu(III) was consistent, that is, the transport behaviors of Eu(III) relyed on the transport behaviors of colloid. However, unlike pure quartz media, the transport of colloids in heterogeneous media were influenced by not only the dispersion stability of the colloid, but also the electrostatic attraction of colloids and media. Even the transport of stable colloids would be partially blocked, so the facilitation of colloid on Eu(III) transport was weaker than that in pure quartz sand. Moreover, due to the formation of additional colloid-iron oxide or colloid-Eu(III)-iron oxide, the co-transport of colloid and Eu(III) was more sensitive to changes in environmental chemical conditions. Therefore, it could be concluded that the influence of colloid on the transport of radionuclides was highly correlated on the stability of colloid and the interaction between colloid and media based on the above analysis. These findings could improve the understanding on the transport, retention and remobilization of environmental colloids and actinide nuclides, raising concerns about their potential transport risk to subsurface water, and provide theoretical guidance for the proposal of the control the transport of radionuclides in the environment.

ID: 0957
CHARACTERIZATION OF RADIUM-CONTAINING COMPOUNDS IN THE HUMAN DIGESTIVE TRACT FOR DECORPORATION

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Radionuclides ingested with food may accumulate in the human body and pose a potential health risk. Radium is a naturally occurring radionuclide, which may be present in drinking water and in certain foods in larger quantities up to ca. 10 Bq kg^{-11}. Daily consumption of only a few Brazil nuts, which can have activity concentrations of > 200 Bq kg^{-1} caused by radium[2], may lead to an additional committed dose in the mSv range, while consumption of drinking water high in uranium progenies, such as the Disi Aquifer in Jordan, can lead to even higher doses[3]. For risk assessment, precise knowledge of the radionuclides' biokinetics is needed. This is also important for development and application of decontamination procedures after accidental incorporation of very high radium quantities. In this project, artificial biofluids produced according to the UBM protocol (BARGE) are used to investigate the interaction of radium in the alimentary tract of humans. In preliminary experiments, the non-radioactive homologue barium is used to determine compounds being formed in these biofluids. The speciation of radium in biofluids, i.e. in the digestive system, are determined by ESI-MS and gamma-spectroscopy. Subsequently, the decorporation agent is added, and the Ra-containing complexes are analyzed again with ESI-MS, and additionally with NMR and IR. The influence of complexation agents on Ra(II) speciation under physiological conditions is investigated. Potential decorporation strategies will be tested in further work. The experimental determination of the speciation of Ra(II) is corroborated by thermodynamic modelling using appropriate tools (e.g. PHREECC).

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ID: 0973
DETERMINATION OF URANIUM IN CALCAREOUS ROCKS/MINERALS AFTER SEPARATION BY NTA-MEDIATED SAMPLE DISSOLUTION AND CATION-EXCHANGE

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The separation and determination of uranium in calcareous samples (e.g. calcite, phosphogypsum and phosphate rock) has been investigated by means of alpha-spectroscopy after dissolution of the samples by nitrilotriacetic solution (NTA), uranium separation by cation-exchange and electrodeposition on stainless steel planchets. Method recovery as well as uranium analysis and isotopic ratio composition of the samples was performed using a uranium standard tracer solution (^{232}U). Comparison of the data with corresponding data obtained from experiments after EDTA-dissolution have indicated lower selectivity for uranium and coextraction of radium isotopes present in the samples, assuming applicability of the method also for radium separation.

ID: 1006
URANIUM SPECIATION IN AN ALKALINE ENVIRONMENT USING TRLFS METHOD

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The importance of the uranium speciation study, specifically in the alkaline environment of cement-based materials, stems from the conditions that can be expected in low and intermediate level waste repositories and also deep geological repositories for spent nuclear fuel, in their section for the storage of high-level decommissioning waste. In addition to its common construction function, cementitious material is used as an engineering barrier, the purpose of which is to slow down transport processes as much as possible, and thus prevent radionuclides from escaping from the repository. Cement is ordinarily utilized as a filler of barrels and a backfill in low and intermediate level waste repositories and will also serve as a barrier in concrete containers when depositing highly active waste during decommissioning. For reliable modelling of processes taking place within this barrier, it is necessary to know the present species of radionuclides. Time resolved laser induced fluorescence spectroscopy (TRLFS) was used to study the uranium speciation in the pH range 6-12 at room temperature. The measured spectral characteristics were mathematically processed. Based on spectra and time characteristics (lifetimes) depending on the pH, specific species were assigned to individual samples by comparison with speciation diagrams modelled using the database Termochimie.

ID: 1009

THE RADIOLOGICAL PROFILE OF THE NORTH AND THE CENTRAL AEGEAN SEA FOR ^{137}Cs, THE WATER MASSES CARRIERS, AND THEIR SPREAD

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The Black Sea is connecting to the Aegean Sea through the Dardanelles Strait and determine the hydrological structure of the area, providing low salinity surface water at the North which is moved southwards along the western shores of the sea, following a generally cyclonic circulation. The data represented in vertical plots combining oceanographic parameters and the radiotracer's ^{137}Cs activity concentration values, for a better representation and understanding of the mixing and circulation of the water masses. The way that ^{137}Cs distributed through the marine masses in the overall area of the north and the central Aegean Sea is described detailed. This area is a starting point of one of the heaviest loaded carriers of ^{137}Cs, the Black Sea Water (BSW) mass. Heavy load from the deposition area of the Black Sea outflows from the strait of Dardanelles and spread directly, and mixes with the other local water masses. Starting with a concentration at the outflow ≥ 7 Bq/m^3 it spreads in an area, where the load afterward varies from 1.6 to 5.5 Bq/m^3. Cs is mostly present in soluble form, while partly absorbed from the particular matter in a small percentage. Following the path of Cs, and analysing its strong influence on the different stratified layers of water masses in the basins, any concentration ≥ 3 Bq/m^3 is, generally, attributed to the influence of the BSW. BSW mixed with the surface waters of the North Aegean, creates mixtures of greater density, as shown by the characteristics of the surface and intermediate waters, examined in the basins under study, the Black Sea waters branch at their exit in 2 main parts. The one part follows a northern course parallel to the coastline of Northern Greece and later descends to the central Aegean, while the second branch leads south with strong mixing and sharp thickening of its features around the central plateau of the central Aegean. Is evident from our combined types of graphs, Θ/S graphs and those incorporating as a parameter the ^{137}Cs concentrations, how the waters of the Black Sea substantially contribute to the creation of a new marine mass called Transitional Subsurface Aegean Water (TSAW), since the radiological load of the remaining marine masses would not allow the formation of this marine mass. At the same time the substantial contribution of upper marine masses to the final load of ^{137}Cs in the bottom masses, as shown also by other studies i.e., Cretan Deep Basin in the southern part of the Aegean, phenomena of over-accumulation are directly related to the residence time of the water masses. There are specific graphic areas where the points of those water masses can be depicted. The spectrum for each of them becomes clearer as more parameters we add for the determination. When an artificial radioisotope is added as a parameter, the characterization becomes easier, because there are no possible natural sources. Further interactions with the Levantine's water entering from the Aegean through the Karpathos Straits, and extends all the way to the surface along the eastern shores of the Central Aegean, once again highlighted in this research. Adapting more radioisotopes, like ^{236}U, and the development of a standard model for characterisation and identification of the marine masses, incorporating artificial radioisotopes with natural ones, are necessary steps for an integrated environmental radiology analytical method, providing information about radio-isotopic depositories.

ID: 1016

DETERMINATION OF NATURAL RADIONUCLIDES IN GROUNDWATER SAMPLES IN AUSTRIA – ANALYSIS OF SEASONAL VARIATIONS AND CORRELATIONS WITH CHEMICAL PARAMETERS

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The Monitoring of natural radionuclides in groundwater samples has been carried out in various projects in Austria over the last 25 years. A large project monitoring groundwater samples from all over Austria took place in 2008 and 2009. Building on this, a follow-up project was launched in 2021, funded by the Federal Ministry for Climate Action, Environment, Energy, Mobility, Innovation and Technology. Sampling sites were selected to check whether activity concentrations have changed over the years. In addition, samples were taken at these sampling sites each quarter of the year to analyse seasonal variations at these sampling sites. ^{222}Rn, ^{226}Ra, ^{228}Ra, ^{210}Pb and ^{210}Po were analysed by liquid scintillation counting. ^{238}U, ^{235}U and ^{234}U were analysed by ICP-MS. ^{210}Pb and ^{210}Po were measured using the extractive scintillation cocktail Polex™. Radionuclides were separated by sulfide and sulfate precipitation, and only 1 L of water sample was used. ^{222}Rn activity concentrations were between the lower limit of detection (LLD) and 620 Bq/l. The activity concentration of ^{226}Ra and ^{228}Ra was between the LLD and 0.14 Bq/l and 0.11 Bq/l respectively. The activity concentration of ^{210}Pb and ^{210}Po was between the LLD and 0.07 Bq/l and 0,11 Bq/l respectively. The activity concentration of ^{238}U, ^{235}U and ^{234}U was between the LLD and 2.7 Bq/l, 0.13 Bq/l and 2.0 Bq/l respectively. Correlations between these radionuclides and chemical parameters such as chloride, boron, sulfate, oxygen content, hydrogen carbonate, nitrate, sodium, potassium, magnesium were analysed. In the presentation a short description of the method is given and the results of the analysis of the seasonal variations and the correlations are presented.

ID: 1020

RADIOCARBON AS A TRACER FOR CLIMATE CHANGE STUDIES

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Radiocarbon analyzed in isotopic archives such as tree rings and marine sediments have been used for tracing past as well as recent climate changes caused by Sun-Earth orbital parameters, solar activity cycles and by increases of green-house gases in the atmosphere. The most visible past climate cycles were represented by Milankovitch cycles, as documented by observed ^{18}O and ^{10}Be variations in ice cores sampled in Antarctica and Greenland, where in the past 800 kyr the dominant cycle was due to the variations in the eccentricity of the Earth orbit (125 kyr period). The exceptional case was a warm period during the Holocene (last 11 kyr), where radiocarbon records during the last 50 kyr, as possible proxies of climate changes, have been recently investigated. The climate changes during the Holocene were most probably due to orbital variations, which changed the latitudinal and seasonal distribution of solar radiation on the Earth. Although the Holocene was generally a warm epoch, there were cool periods as well, such as a Little Ice Age (14th-19th century), which followed after the Medieval Warm Period (11th – middle of 14th century). The cool period coincided partly with the Maunder minimum, however, physical mechanisms explaining how changes in solar activity could affect Earth’s climate require further investigations. Remarkable increases in surface temperature have been observed during the last century, which could be associated with increases of green-house gases in the atmosphere (mainly CO_2 and CH_4), having also impacts on sea ice cover changes, on sea level rise, etc. Future trends in climate change remain, however, a great challenge for better understanding of anthropogenic and natural processes which could affect Earth’s climate (e.g. solar activity impacts via secondary effects). High-quality radionuclide data (mainly radiocarbon in tree rings and ^{10}Be in ice cores) will be useful source of information on past climate changes.

ID: 1027

SOURCES AND THE DISTRIBUTION OF ^{129}I IN SOILS FROM NORTHEAST CHINA

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Human nuclear activities including nuclear weapons tests and nuclear accidents have released large amounts of radioactive substances to the environment. Due to the high volatility of iodine and high fission yield of radioactive iodine (^{129}I, ^{131}I), the released radioactive iodine has been dispersed and deposited in a large area. The long-lived iodine-129 is therefore can be used as an ideal tracer for investigation on the dispersion of radioactive substances in the atmosphere and the assessment of environmental impact, meanwhile to study environmental process. About 100 surface soil samples (0-5 cm) collected from Northeast China in 2014 were analyzed for ^{129}I and ^{127}I concentrations using accelerator mass spectrometry and ICP-MS after chemical separation of iodine from the sample matrix and other interferences. The concentrates of ^{127}I in the soil samples are 0.13-10.07 µg/g, with an average of 2.67 µg/g, which is consistent with the observed value in a large spatial scale of China. The variation trend of ^{127}I concentrations shows high values in the southeast region, which is attributed to the organic matter content in the soils and the high precipitation rate in this region. The ^{129}I/^{127}I atomic ratios vary in 2.61×10^{-10} -6.49×10^{-8}, which are 2-3 orders of magnitude higher compared to the estimated pre-nuclear level. This level is also consistent with the level observed in other areas in China. Unexpectedly enhanced ^{129}I/^{127}I ratios of more than 1×10^{-8} were observed in the Mid-eastern Inner Mongolia (MIM). The high ^{129}I level in this region indicates that besides the global fallout, the European nuclear reprocessing plants and nuclear weapon testing in Semipalatinsk and Lop Nor are the fraction of anthropogenic ^{129}I sources in MIM. In addition to the long-range atmospheric dispersal of ^{129}I from nuclear fuel reprocessing plants, the dust and airflow carried ^{129}I deposited in the Asian-dust area, which might be another significant ^{129}I input in the MIM. In addition to the abundant ^{129}I source, the special atmospheric circulation and the topographical factors have provided favourable conditions for deposition of radioactive materials, which lead to the dust that carried ^{129}I from the Asian-dust area and atmospheric ^{129}I from long-range transportation would be prone to deposition, compared with the relatively arid area of the west. Finally, the higher vegetation cover and the soil type with higher content of the organic substance in MIM, worked together leading to the effective preservation of the deposited ^{129}I, which is also an important factor for high ^{129}I in MIM. This work systematically investigated the distribution of ^{129}I in Northeast China for the first time and unexpectedly found a high ^{129}I level in the area far away from the point sources. This feature might shed light on the role of the positive deposition and the effective preservation in the distribution of radionuclides. The implication of ^{129}I as a tracer to understand the mechanisms of regional deposition of radionuclides is essential to allow prediction of future distribution of anthropogenic nuclides and for evaluation of its possible effects on human health.

ID: 1037
URANIUM DESORPTION FROM MICROPLASTIC SURFACES

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The desorption of uranium from microplastic surfaces has been investigated as a function of pH in aqueous solutions under ambient conditions. In addition, the effect of complexing ligands (e.g. EDTA, NTA, Citrate) on the desorption from plastic surfaces (including the plastic containment surface) has been studied. The evaluation of the experimental data indicate that the solution pH and the presence of complexing ligand in solution strongly affect the desorption process. These findings suggest that microplastics could act as radionuclide/uranium carriers into environment and living organisms.

ID: 1041
EVALUATION OF THE DEPENDENCE OF THE ACTIVITY CONCENTRATIONS RATIOS $^{234}U/^{238}U$ AND $^{235}U/^{238}U$ IN WATERS ON THE ^{238}U ACTIVITY CONCENTRATION

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In routine radiochemical analyzes of water, especially drinking water, in recent years we have analyzed more than 4,000 water samples for uranium ^{238}U, ^{235}U and ^{234}U isotope contents by ICP/SFMS method in the ALS Scandinavia AB laboratory, Luleå, Sweden and the Central Analytical Laboratory of ÚJV Řež, a.s., Husinec – Řež, in the Czech Republic. By statistical processing of an extensive set of data, we evaluated the interesting patterns that are discussed in this article and which can significantly help in the evaluation and interpretation of measurements of natural radionuclides in drinking water. While the $^{235}U/^{238}U$ activity ratios in natural waters and rocks are more or less constant, the $^{234}U/^{238}U$ activity ratios in natural waters, especially groundwater, but also in some soils, sediments and rocks, usually do not correspond to the calculated theoretical values. The $^{234}U/^{238}U$ activity ratio is 1.0 only in closed systems, e.g. in some minerals that have not been subjected to any chemical effect for a long time. In waters, the activity ratio of $^{234}U/^{238}U$ is often many times higher than 1.0. The reason for this is the fact that ^{234}U in rocks is formed by the decay of its parent nuclide, ^{238}U. After one alpha decay and two beta decays, the daughter ^{234}U is released from its original position in the crystal lattice of the mineral due to "atomic recoil", and upon subsequent chemical attack of the rock by groundwater, ^{234}U atoms enter the aquatic environment much more easily than its parent ^{238}U atoms. Thus, uranium-234 isotope is much more chemically reactive than uranium-238 in the solid sample, and as a result, the $^{234}U/^{238}U$ activity ratio in waters and in the formed secondary minerals and rocks is much higher than 1.0. The fact that 234Th, the short-term alpha decay product of ^{238}U, can be released to a greater extent into the aqueous environment after atomic recoil, where it decays to ^{234}U very quickly, also contributes in part to the increase in ^{234}U activity against ^{238}U. We confirmed these facts by statistical processing of an extensive data set of ^{238}U, ^{235}U and ^{234}U contents in the analyzed water samples. The $^{235}U/^{238}U$ activity ratio is constant within the uncertainties of the determination, no exception was found. In contrast, the activity ratio of $^{234}U/^{238}U$ is significantly higher than 1.0, the overall average of this ratio is 1.9. The value of this ratio depends on the hydro-chemical conditions of the environment, but it can generally be said that the lower the total volume concentration of uranium resp. the volume activity of ^{238}U in groundwater, the higher the value of this ratio. If the volume activity of ^{238}U in groundwater decreases from XX-XXX Bq/L to 0.0XX Bq/L, the 95 % percentile distribution of the $^{234}U/^{238}U$ ratio will increase from about 3 to 8-10. The work also discusses relatively "rare cases" where the activity ratio $^{234}U/^{238}U$ is less than 1.0. The values of these activity ratios were also evaluated for about 500 analyzed solid samples.

ID: 1042

A MECHANISTIC MODEL FOR COLLOID AND RADIONUCLIDE CO-TRANSPORT IN SATURATED POROUS SYSTEMS: BASED ON A TWO-SITE ADSORPTION BLOCK MECHANISM

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The presence of colloids in groundwater can enhance radionuclides transport by loading on colloid to co-transport. The mechanism of co-transport is difficult to be fully revealed based on macroscopic experimental observations because the co-transport process cannot be monitored in situ. Hence a reliable model of colloid and nuclide co-transport is needed. Considering that colloid transport dominates the coupled transport in the process of co-transport, a finite difference model was constructed to describe the co-transport behavior of nuclide and colloid in saturated homogeneous porous media, focusing on colloid transport mechanism. The proposed model is formulated based on the assumption that the colloid transport is not affected by the presence of nuclides by ignoring the decay process of nuclides in a short period of time. The one-dimensional convection-dispersion transport of nuclides in porous media is coupled with the transport of nuclides attached to colloid as the pseudo-colloid. The adsorption and desorption processes (linear kinetic reactions) of nuclides on porous media and on mobile or filtered colloid are considered. The colloid transport model includes the reversible and irreversible adsorption of colloid on the host medium and the adsorption arrest process. With the assistant of the proposed model, the distribution ratio of nuclides as the free solute and colloidal attached states during co-transport was exploded, the related colloid driven transport mechanism was further elucidated, which would be helpful to predict the transport and retention risk of nuclides in porous media. The transmission model is validated by the comparison of calculated results with experimental co-transport results.

ID: 1052

RETROSPECTIVE DETERMINATION OF ^{236}U/^{238}U AND ^{240}PU/^{239}PU ATOM RATIOS IN AEROSOLS AND LUNG ASHES FROM VIENNA, AUSTRIA

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Already in the frame of the first scientific project sponsored by the IAEA with the title “Factors controlling the distribution of fission products in the biosphere” in the early 60's of the last century, air filters and lung autopsy samples from people, who had been living in Vienna, Austria, were collected and investigated with regard to short-lived fission products from the nuclear weapons tests. The organs were ashed and then measured with a well-type NaI(Tl) crystal to quantify ^{141}Ce+^{144}Ce, ^{103}Ru, and ^{95}Zr+^{95}Nb contamination of humans. Later on (after decay of these rather short-lived radionuclides) also ^{137}Cs was determined. From a part of the ash samples Pu was separated and the sum activity of ^{239}Pu and ^{240}Pu was measured by alpha-spectrometry. Corresponding investigations were also performed with air filters. In the last years we investigated some of the remaining air filters and lung ash samples with Accelorator Mass Spectrometry (AMS) with regard to ^{236}U (half-life $2.3 \cdot 10^7$ years) which is mainly produced via the reactions ^{235}U(n,γ) ^{236}U and ^{238}U(n,3n) ^{236}U. Additionally, the Pu isotopes ^{239}Pu and ^{240}Pu were measured to obtain an improved data set with reduced uncertainties compared to the earlier study. The samples were dissolved in half-concentrated nitric acid, a defined amount of ^{233}U was added as a spike and uranium was then separated by anion exchange (UTEVA) and co-precipitated with Fe(OH)$_3$. Pu was separated with DOWEX 1x2, here a ^{242}Pu spike was used. After calcination the samples were analysed by AMS at the VERA laboratory (university of Vienna). The results of our lung measurements will be compared to the corresponding filter results. Possible explanations of the discrepancies will be discussed.

ID: 1067

HIGHLY SENSITIVE METHOD FOR DETERMINATION OF ^{90}Sr IN LARGE-VOLUME GROUNDWATER SAMPLES USING PIXEL DETECTORS

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This work describes a newly developed method for determination the specific activity of ^{90}Sr in large-volume groundwater samples in which a high concentration of sodium, calcium and magnesium can be expected. This method is based on separation of strontium from other cations which increase the thickness of target sample, and radionuclides, especially beta emitters. In the groundwater sample, the concentrations of Ca, Mg and Sr are determined by OES. Depending on the concentration of natural occurring strontium, the sample volume between 30 and 50 L is analysed. If the concentration of naturally occurring strontium is low, the sample volume will be 50 L and stable strontium is added to the sample to give 30 mg of strontium in the whole sample. Stable strontium is used as a carrier and yield monitor. This method consist of two main phases: separation of the strontium and ^{90}Sr activity determination. The separation consists of five steps. The first step is the pre-treatment and mineralization of the insoluble fraction of the sample. The sample is then concentrated by carbonates precipitation, followed by pre-separation on a strongly acidic cation exchanger (Dowex® 50w x8 100–200 mesh) by EDTA solution. The main separation of strontium is performed using Sr resin. The final step is the preparation of the sample for measurement. After 26 days secular equilibrium of the ^{90}Sr and ^{90}Y is achieved and samples are measured using pixel detector in a shielded chamber. The evaluation of the measured data is performed using an algorithm written in python. Using this procedure a detection limit lowered by one order of magnitude (0,1 mBq per sample) in comparison with conventional method used in NRPI can be obtained.

ID: 1069

RADIOCARBON STUDY OF CHARCOALS OF ROMANSEQUE PASSAGE AT PRAGUE CASTLE

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Below ground level at Prague Castle there is several remainders of ancient buildings and structures which have been of interest of intense archaeological research since thirties of the 20th century. One of them, part of the so-called Small Excavations of the Third Courtyard is the Romanesque corridor. As most of these underground structures, it is not accessible to public. The passage was part of the Romanesque bishop's area at Prague Castle, connecting the Basilica of St. Vitus, Wenceslas, Adalbert and the Virgin Mary and the Chapel of St. Bartholomew. The last chapel stood in the centre of the present Third Courtyard. According to current research, the corridor is a building from the 12th century and is believed to disappeared during the 14th century. As an alternative to direct dating of mortars, radiocarbon dating of charcoals collected in the walls of the corridor was performed with MILEA accelerator mass spectrometry system at Řež by Prague. The charcoals are believed to originate from wood used in lime burning. 28 charcoals were found in surface mortar layers and 8 charcoals were extracted from depth probes into the mortars. Out of total 36 charcoals, five was of under minimal sufficient weight, one was discarded as a stone and one as a contaminated with mold, 14 dissolved during chemical pretreatment and the remaining 15 was successfully dated. The results are discussed together with specific contexts of the sample points.

ID: 1082

NATURAL BITUMEN HOSTED URANIUM MINERALIZATION, STABILITY OF THE RADIOGENIC SYSTEM AND MOBILITY OF SELECTED ELEMENTS IN SURFACE CONDITIONS.

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Uranium mineralization hosted in sedimentary rocks is exposed in the roadcut. The sediments are of lacustrine origin with a high proportion of organic matter and enriched in uranium and several other potentially hazardous elements. The most remarkable are two mineralized uranium-rich sulphide-bearing bitumen (thucholite) veins with locally high contents of U (up to the order of wt.%). The exposed rock profile in the length of about 250 m was examined by the method of field gamma spectrometry. Uranium-rich zones were documented and sampled. The exposure of mineralization to the atmosphere, precipitation, infiltration and drainage through disrupted mineralization structures causes local oxidation of sulphides and arsenides to release acidic solutions carrying sulphate, arsenate, uranyl, Co_2^+, Cu_2^+, Ni_2^+, Pb_2^+ and other species. Subsequently, amorphous precipitates and secondary minerals of the mentioned elements can be found on the surface of the crushed loose rocks and the bitumen itself. Specimens with U content and manifestations of S, Cu, Ni, Co, As mineralization were taken in form of secondary minerals. Consequently, optical microscopy, SEM/EDX, XRD, NAA and LA-ICP-MS analyses were carried out to obtain data on the chemical, mineral, and lead isotopic composition of the constituent phases. Microscopy methods revealed structural features which provide evidence of multi-stage evolution of bitumen mineralization accompanied by both ductile and brittle deformations of matrix and embedded uraninite, respectively. Despite complex evolution within a dynamic fault environment and at least one hydrothermal event after the U-mineralization stage, no significant mobilization and migration of uranium occurred. Reaction-diffusion features at the uraninite grain margins did not even exceed the range of alpha particles in bitumen and the only significant uranium migration occurred after exhumation. Newly formed discontinuities in the rigid mature bitumen related to shear stress and volume change (contraction cracks) can serve as pathways for the infiltration of meteoric water-based fluids. Weathering of sulphides locally causes so-called acid mine drainage (AMD) phenomena associated with the production of highly mobile aqueous species of several hazardous elements including uranium. On the other hand, the presence of arsenic and its oxidation to As(V) species effectively inhibits the migration of U(VI) and other heavy metal ions by forming insoluble mineral phases. Ferric hydroxy oxides (FHO) are found to be another scavenger of uranyl and arsenate ions, likely by the sorption mechanism, however, the SEM/EDX investigation reveals that FHO may also serve as a nucleation substrate for U-bearing arsenates. It is assumed that the studied outcrop of uranium-bearing bitumens has the potential as a natural analogue for studies of mobility and environmental behaviour of actinides and other hazardous elements found in the mineral association.

The study has been supported by the Czech Science Foundation project 19-05360S, and by the Ministry of Education, Youth and Sports of the Czech Republic within the project LM 2015056 (CANAM infrastructure).

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8. Radiopharmaceutical Chemistry, Labelled Compounds

ID: 0883

DESIGN OF A NOVEL CARRIER FOR DY-166/HO-166 IN VIVO GENERATOR

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Radionuclide therapy is a promising therapeutic modality for cancer treatment. Radionuclides are usually bound to conventional chelators such as DOTA and conjugated to a tumor-targeting agent to selectively kill tumor cells by locally delivering ionizing radiation while sparing healthy tissue. Holmium-166 (^{166}Ho, half-life=26.8 h) has been recently applied in radioembolization to treat hepatocellular carcinoma or liver metastases due to its emission of high-energy beta particles. Its mother nuclide, dysprosium-166 (^{166}Dy), has been considered as an alternative for ^{166}Ho in cancer treatment, i.e. the ^{166}Dy/^{166}Ho in vivo generator. Due to the longer half-life time of ^{166}Dy (half-life 81.5 h), approximately three times higher dose can be delivered to a tumor with the same amount of administrated activity as ^{166}Ho. However, it has been reported that 72 % of the ^{166}Ho disassociates when bound to DOTA or similar chelators due to the increase of charge number of ^{166}Ho ions after the internal conversion. The resulting ^{166}Ho ions with high charge tend to extract electrons from surrounding atoms and thus cause the rupture of the Ho-DOTA complex. The released ^{166}Ho might induce severe side effects to the healthy tissues. Therefore, a carrier that can prevent the loss of ^{166}Ho due to internal conversion has to be developed. In this work, we prepared ^{166}Dy labelled gold nanoparticles via a seed-mediated growth method. First, ^{166}Dy was co-reduced with a gold precursor to form a Dy-Au nanoparticle seed. Then an extra gold layer was grown on top of the seed nanoparticles to form a core-shell structured nanoparticle, i.e. DyAu@Au nanoparticle. The final product had a diameter of 5 nm and a ^{166}Dy labelling efficiency of 60 %. The ^{166}Ho retention tests showed that more than 95 % of ^{166}Ho was retained for at least 72 hours at 37 °C in water. To the best of our knowledge, this is the first study to retain radionuclides freed due to internal conversion with gold nanoparticles. Overall, this study presents a simple, quick, and chelator-free radiolabelling method for ^{166}Dy with minimum loss of internally converted ^{166}Ho.

ID: 1017

PHYSICAL AND BIOLOGICAL EXAMINATION OF GEL Y_2O_3 MICROSPHERES FOR RADIOEMBOLIZATION THERAPY

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Radioembolization is a specific type of internal radiotherapy used to treat primary or metastatic hepatic tumors. The basis of this therapy is the intra-arterial insertion of microspheres containing beta radioactive yttrium in the vicinity of the tumor tissue. The aim of the work was carried out physical and biological experiments performed to determine radiometric parameters and to define the possible medical usefulness of the newly developed gel $^{90}Y_2O_3$ microspheres. Materials and Methods: Manufacturing based on the sol-gel method allowed to obtain spherical, yttrium trioxide grains of fully polycrystalline structure with diameters between 20 µm and 100 µm (62.1 % of the total batch). NAA analysis confirmed a high concentration of radioactive ^{90}Y in the sample (>99.99 %). The developed procedure for determining the specific activity of a single microsphere showed that at the time of administration, the activity can be specified at the level of 2600 – 3200 Bq per microsphere. The reduction in colorectal cancer cell proliferation in vitro confirms the significant influence of beta radiation from yttrium-90 trioxide microspheres. Histopathological examination of the distribution of microspheres in the animal model confirmed the proper location of yttrium trioxide microspheres inside blood vessels in a porcine model. Gel Y_2O_3 microspheres manufactured using the sol-gel method showed relevant properties, indicating the possible use of microspheres for further biological and preclinical studies. Physical investigations, cancer cell proliferation, histopathological studies, and their results created a basis for future activities toward to clinical experiments.

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ID: 1032

IMMUNOMODULATORY NANORADIOPHARMACEUTICALS AS A NEW PARADIGM IN CANCER THERAPY—GREEN NANOTECHNOLOGY TOWARD THE DEVELOPMENT OF MGF-^{198}AuNPS NANOMEDICINE AGENT

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The interrelationship of NF-κB and related signaling pathways involving NF-κB-related effector genes in adaptive radioresistance has attracted significant current interest toward the overall quest of developing more effective radiotherapeutic agents. Compelling evidence suggests that radiotherapy triggers several signaling pathways including NF-κB and related signaling vectors causing repopulation with radioresistant cancer stem cells—thus contributing to tumor radioresistance. Numerous pre-clinical and clinical data suggest the potentiation of radiation-induced therapy resistance is mediated solely by activation of NF-κB signaling pathway. Activated NF-κB in tumor cells is a key mechanism through which tumors acquire radioresistance and this tumor biological process has been linked to increased recurrence and failure of radiation therapy in cancer patients. It is, therefore, imperative to develop radiotherapeutic agents with dual immunomodulation while delivering radiotherapy site specifically to tumor sites. New radiotherapeutic agents must address targeting NF-κB transcription factors for cross talk between cell signaling and macrophage reeducation within the tumor microenvironment. Radiotherapeutic agents with capabilities to deliver effective radiation dose to tumor sites while suppressing NF-κB will provide the best means to treat myriad of human cancers effectively. This lecture focuses on the development of Mangiferin functionalized ^{198}Au nanoparticles (MGF-^{198}AuNPs) as an innovative dual, NF-κB suppressing, radiation therapy with immunomodulatory effects. MGF-^{198}AuNPs provides a desirable beta energy emission and half-life that destroys tumor cells/tumor tissue (βmax = 0.96 MeV; half-life of 2.7 days) while the presence of mangiferin suppresses pro tumor NF-κB signaling pathways. The penetration range of ^{198}Au (up to 4 mm in tissue or up to 1100 cell diameters) is sufficiently long to provide cross-fire effects to destroy tumor cells/tissue, but short enough to minimize radiation exposure to adjacent tissues. One particularly attractive feature of radioactive gold nanoparticles is that it does not have to be incorporated into every tumor cell to have a therapeutic effect. The path length of the emitted radiation is sufficient to allow effective therapy following uptake into a subpopulation of tumor cells. This lecture will provide:

- Scope and prospects of beta emitting radioisotopes in nanomedicine through the design and development of immunomodulatory radiotherapeutic agent: MGF-^{198}AuNPs
- Full in vivo investigations on therapeutic properties of MGF-^{198}AuNP agent in treating prostate tumors;
- Immunomodulatory effects of MGF-^{198}AuNPs through targeting the tumor microenvironments and thereby suppressing NF-κB transcription factors while promoting antitumor M-2 macrophages; and
- Estimation of the dose distribution delivered by radioactive gold nanoparticles (MGF-^{198}AuNPs) to the tumor inside the human prostate as well as to the normal tissues surrounding the tumor using Monte-Carlo N-Particle code (MCNP-6.1.1 code).

This lecture will highlight details on the importance of interaction of various types of cancer cells with immunotherapeutic nanoparticles for stopping the infiltrating aggressive tumor proliferating M2 type macrophages to achieve effective tumor therapy. The overall implications of Green Nanotechnology of MGF-^{198}AuNPs as a therapeutic beta emitting nanomedicine immunomodulatory agent in oncology will be discussed.

ID: 1055

DEVELOPMENT OF ^{211}At-RADIOPHARMACEUTICALS FOR PRETARGETED RADIOIMMUNOTHERAPY OF DISSEMINATED CANCER

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To enhance the therapeutic efficacy of radioimmunotherapy of cancer, several pretargeting strategies have been developed. In pretargeted radioimmunotherapy, the tumour is pretargeted with a modified monoclonal antibody that has affinity for both, tumour antigen and radiolabeled carrier. A big challenge in cancer treatment is the elimination of occult disseminated tumour cells, in this context the alpha emitter ^{211}At has drawn attention due to its physicochemical characteristics. This project aims to design new molecules for pretargeting applications based on the Diels Alder click chemistry system (Tetrazine (Tz)/Trans-cyclooctene (Tco)) using ^{211}At as radionuclide, and to evaluate these substances to optimize the pharmacokinetics. The effector molecule was synthesized attaching N-Succinimidyl-3-(trimethylstannyl)-benzoate, Tetrazine-NHS ester, and Succinic Anhydride to a poly(L)lysine (PL) scaffold (HTzPL and MeTzPL). A dry astatine residue was activated with N-iodosuccinimide followed by electrophilic substitution of the trimethyl-tin group on the TzPL, resulting in an astatinated product. Purification was performed eluting the product in PBS on illustra NAP-5 column. Radiochemical purity of the product was determined with radio-TLC using 100 % ethanol as mobile phase and iTLC-SG as stationary phase. Finally, Tco-functionalized magnetic beads (Tco-mBeads) were used to assess the Tetrazine/Tco binding after radiolabeling. Results: Astatinated TzPL resulted in > 80 % of radiochemical yield and > 98 % of radiochemical purity. Preliminary studies on Tco-magnetic beads have shown a binding > 90 % already after the first minute to reach a value > 97 % after one hour. A protocol for the synthesis and radiolabeling of the PL based tetrazine-effector molecule has been developed. High radiochemical yield and radiochemical purity can be obtained in the polymer astatination, resulting in a good binding to Tco-mBeads. H-Tetrazine is reported to react faster than the Me-Tetrazine with Tco, which would be a great advantage when administered in vivo, but it is also less stable. Comparative studies are thus being performed to assess the characteristics of each compound. Additional in vitro and in vivo studies will be carried out for the full evaluation and optimization.

ID: 1078

IN VITRO STUDIES OF RADIUM-223 AND ACTINIUM-225 LABELLED A-ZIRCONIUM PHOSPHATE AS POTENTIAL CARRIER FOR ALPHA TARGETED THERAPY

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Targeted alpha particle therapy (TAT), which uses radionuclides emitting alpha particles is one of the promising possibilities for the treatment of a broad range of malignancies. A short range of alpha particles in soft tissues (approx. 50-100 µm) and high linear energy transfer (LET) allows to destroy tumor cells effectively. Precise targeting by appropriate carrier should ensure efficient and accurate destruction of target cancer tissue while keeping healthy tissues intact. Currently, there are only few such radionuclides that are suitable for use in nuclear medicine. Two widely employed radionuclides, the ^{223}Ra and the ^{225}Ac, are among the most potential radionuclides suitable for targeted alpha therapy[1]. Nanoparticles can serve as suitable radionuclide carriers for TAT, particularly considering their ability to at least partially stop the progeny recoils spread and keep the daughter atoms immobilised in their structure. At the same time these nanoparticles can be surface-modified, which may improve their targeting, stability and other biological and chemical properties[2,3]. Nanoparticles of α-zirconium phosphate (α-ZrP) were selected and tested as potential carriers of ^{223}Ra and ^{225}Ac in this study. Nanoparticles of α-ZrP were prepared by the reaction of zirconium oxychloride octahydrate aqueous solution with sodium dihydrogen phosphate solution in hydrochloric acid under reflux. The prepared particles were washed by deionised water and then redispersed in ultrapure water. The size of the particles was determined using dynamic light scattering method and the Z-potentials were also measured. Consequently, samples were labelled in aqueous solution with ^{223}Ra and ^{225}Ac. The labelling yield exceeded 98 % in most cases both for ^{223}Ra and ^{225}Ac. Subsequently, in vitro stability studies were carried out in four biological matrices: blood serum, blood plasma, saline and 5 % albumin solution during 48 h period. Measurements of released activities revealed that samples exhibit the highest stability in saline. Released activity of ^{223}Ra, ^{225}Ac and their daughter radionuclides was around 0,5 %. On the other hand, the lowest stability was shown in blood plasma for ^{223}Ra, where released activity was above 15 % and 18 % for ^{211}Pb and ^{211}Bi after 48 h. In blood serum was measured highest release activity for ^{225}Ac (close to 18 %) and its daughter radionuclides ^{221}Fr (more than 20 %) and ^{213}Bi (above 32 %).

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ID: 1083

SEPARATION OF TERBIUM-161 FROM GADOLINIUM TARGET IRRADIATED IN NUCLEAR REACTOR

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Terbium-161 is one of the perspective radionuclides with a potential use in nuclear medicine thanks to its ideal energy of beta radiation (E = 593 keV) and half-life (6,9 d). In addition, terbium-161 emits a significant amount of conversion and Auger electrons, which increases its potential therapeutic efficacy. Terbium-161 can be prepared as no carrier added by neutron irradiation of highly enriched gadolinium-160 target in a nuclear reactor, through indirect production route ^{160}Gd (n,γ) ^{161}Gd → ^{161}Tb. After irradiation, it is necessary to separate terbium from the target. For this purpose, the cation exchange chromatography is commonly used. In this work, several irradiations of highly enriched ^{160}Gd target in the form of oxide or nitrate were performed in the nuclear reactor LVR 15 (CV Řež). For separation of ^{161}Tb from target, cation exchange chromatography (Dowex 50W×8 (H+), 100-200 mesh) with variously concentrated α-hydroxyisobutyric acid as eluent was used. The presence of ^{161}Tb and possible radionuclide impurities was verified by gamma spectrometry on an HPGe detector. The fractions containing ^{161}Tb were purified from α-hydroxyisobutyric acid also on the cation exchange resin, using hydrochloric acid as eluent. The presence of stable impurities was verified by ICP-MS method.

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ID: 1089

LABELLING OF PSMA-617 WITH 161-TB

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PSMA (Prostate-Specific Membrane Antigen) is a transmembrane protein which is overexpressed in most cases of prostate cancer. Expression level of this protein has a strong correlation to the stage of the disease, which makes PSMA a very attractive target for radionuclide therapy of metastasized castration-resistant prostate cancer (mCRPC)[1]. One of the most promising radioligands for the treatment of mCRPC is a small-molecule-based PSMA-617 equipped with a DOTA chelator. PSMA-617 labelled with ^{177}Lu has been successfully applied in clinics. Recently, there has been a lot of investigation put into the search for a more powerful and accessible therapeutical radiometal. A recently-introduced ^{161}Tb has similar chemical and nuclear characteristics to ^{177}Lu, but also emits substantial amount of low-energy conversion and Auger electrons, which is believed to enhance therapeutic efficacy of the radiopharmaceutical[2]. In this work, we decided to optimize the reaction conditions for the labelling PSMA-617 with ^{161}Tb and investigate the radiolytic stability of the labelled compound. Radiolabelling was achieved by adding [^{161}Tb]TbCl$_3$ (100-200 MBq) to the 1 mM water solution of PSMA-617 in acetate buffer (pH 4.5). Reaction mixture was stirred for 15-30 min at 95 °C. Radiochemical yield (98 %), as well as radiolytic stability, was studied by HPLC with radiometric detection. Reaction samples were measured every 24 hours. No stabilizing additives were used in this experiment. Promising ligand PSMA-617 was successfully labelled with ^{161}Tb. Radiolytic decomposition of the radiolabelled compound was apparent from the radiochromatograms. Further improvement of reaction conditions is warranted in order to suppress radiolytic processes.

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ID: 1004

FAST, EFFICIENT AND SIMPLE METHOD TO RADIOLABEL POLYMERIC MICELLES WITH RADIONUCLIDES

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A variety of different polymer micelles are applied in the clinic as nano-carriers for chemotherapy. For a safe and effective application, it is imperative to know how they behave in vivo. Here, we present a chelator-free method for radiolabeling of polymer micelles to enable in vivo biodistribution studies. The radiolabeling method is very simple and is achieved by just adding the radioisotope ions, i.e. ^{111}In(III), to the micelle solution and the removal of unencapsulated radionuclides. We tested different polymers and we show that micelles composed of poly(ε-caprolactone-b-ethylene oxide) reach high ^{111}In(III) radiolabeling efficiency (>80 %) and exhibit radiolabeling stability (>90 %). The results indicate that the radiolabeling is driven by two factors: the properties of the core forming block copolymer and the speciation of the radiometal salts. The formation of metal hydroxides and their precipitation in the hydrophobic core seems to be essential for achieving high radiolabeling efficiency and stability. This method was further applied to radiolabel the micelles with ^{177}Lu(III) and in the presence of chemotherapeutic drugs such as paclitaxel (PTX). A SPECT/CT pharmacokinetic study was then applied which revealed that that the radiolabeled samples were stable in vivo. The proposed radiolabeling mechanism appears to be widely applicable and is expected to play a role any fields where tracers are desired.

ID: 1010

PRODUCTION OF CYCLOTRON-BASED ^{68}Ga RADIOISOTOPE AND RELATED RADIOPHARMACEUTICALS

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Radioisotope ^{68}Ga is used for radiopharmaceuticals synthesis word wide and its application is increasing every year. ^{68}Ge/^{68}Ga generator are the main source of ^{68}Ga for radiolabeled radiopharmaceuticals products such as ^{68}Ga-DOTATOC and ^{68}Ga-PSMA. The price of the generator is quite expensive and due to decaying of ^{68}Ge it is necessary to buy a new one nearly every half year. Another limitation of the all available on market generator is its maximum produced activity up to 50 mCi (1.85 GBq). Half-life of ^{68}Ge is 271 days it means that after half year is as little as 1.15 GBq. The yield of ^{68}Ga elution and radiolabeling could be between 80-50 % and for PET examination is necessary about 150-300 MBq radioactivity. Production of ^{68}Ga using cyclotron from enriched ^{68}Zn using ^{68}Zn(p,n)^{68}Ga reaction can be easily prepare with activity as much as 10-100 GBq suitable for radiolabeling. There are two possible way of production ^{68}Ga from point of targetry – from solid target, where the layer of ^{68}Zn could be prepared by pressing of Zink powder or by electrodeposition. The second possibility is liquid target, where the ^{68}Zn is dissolved in nitric acid. Anyway, many technical details have to be solved. The choice of proper proton energy, solid target preparation with suitable amount of zinc used to get proper thickness for proton absorption, dissolution of solid target, optimal concentration of ^{68}Zn in nitric acid solution, separation chemical procedure for zinc removal with high efficiency (Zn decreasing concentration more than several thousand times). Besides of chemical separation processes in which specific sorbents are used and high concentration of hydrochloric acidic solutions. On the other side there is a problem with high acidic solutions for radiolabeling. The main cycle for ^{68}Ga preparation, target preparation, proton irradiation of enriched ^{68}Zn, dissolution, separation and labeling procedure for following radiopharmaceuticals – ^{68}Ga-DOTATOC, ^{68}Ga-DOTANOC and ^{68}Ga-PSMA11 will be presented.

9. Separation Methods, Speciation

ID: 0890

SELECTIVE ACTINIDE(III) SEPARATION USING 2,6-BIS[1-(PROPAN-1-OL)-1,2,3-TRIAZOL-4-YL]PYRIDINE (PYTRI-DIOL) IN THE INNOVATIVE-SANEX PROCESS: LABORATORY SCALE COUNTER CURRENT CENTRIFUGAL CONTACTOR DEMONSTRATION

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An innovative-SANEX process for the selective separation of the trivalent actinides americium and curium from a simulated PUREX raffinate solution was successfully demonstrated on the laboratory scale using a 16-stage 1 cm annular centrifugal contactor setup. The solvent was composed of 0.2 mol L^{-1} N,N,N',N'-tetra-n-octyl-diglycolamide (TODGA) and 5 % v/v 1-octanol in a kerosene diluent. Zr(IV) and Pd(II) co-extraction was prevented using trans-1,2-diaminocyclohexane-N,N,N',N'-tetraacetic acid (CDTA) as a masking agent in the feed. The actinide(III) selective back-extraction was achieved using 2,6-bis[1-(propan-1-ol)-1,2,3-triazol-4-yl]pyridine (PyTri-Diol) in 0.45 mol L^{-1} HNO_3 as a CHON alternative to the sulfur-containing stripping agent used in a previous version of the innovative-SANEX process. The new process described in this paper showed excellent performance for the recovery of An(III). An An(III) product with a quasi-quantitative recovery of americium and curium (≥99.9 %) and very good separation from fission and activation products was obtained (decontamination factors ≥4,000). Only a slight contamination with Zr and Ru was observed. This test demonstrates the successful use of molecules containing only carbon, hydrogen, oxygen, and nitrogen atoms (so-called CHON molecules) for the selective separation of An(III) from a simulated PUREX raffinate solution. By avoiding sulfur- or phosphorous-containing molecules, the generation of secondary radioactive waste during process operation can be reduced drastically. The results of the demonstration test will be presented and discussed.

ID: 0894

INHIBITION THE FORMATION OF CRUDS IN EXTRACTION SYSTEMS

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In radiochemistry, liquid extraction is widely used to concentration and separation of elements. Often, upon contact of two immiscible liquids, interfacial formations (cruds) are formed in the area of the interface, which reduce the rate of extraction, worsen the separation of the emulsion, and complicate the process. Sometimes a precipitate is formed that disrupts the operation of the extractors, reduces their productivity and the duration of uninterrupted operation. The high heat release from radioactive precipitation causes local overheating and may be accompanied by emissions. The search for ways to inhibition the formation of cruds is actual. This report presents the results of investigation on the effect of mechanical vibrations on the formation of interfacial formations during the extraction of rare earth elements with solutions of di-(2-ethylhexyl)phosphoric acid. Local energy supply to the interface of the heterogeneous system was carried out using a vibration element placed in the interfacial layer and driven by an electrodynamic head. Vibration are inhibition structure formation in the interface of the extraction system. Under the action of vibration, the emerging temporary structure is destroyed. In the presence of vibration, the effective viscosity in the interfacial layer does not change in the first 15 min, then it slightly increases. The supply of additional energy to the interfacial layer of the extraction system changes the hydrodynamic situation. Under the influence of mechanical vibrations, particles of different masses move at different speeds, which negatively affects the formation of the temporal structure. Molecules of di-(2-ethylhexyl)lanthanide phosphate are adsorbed on the active lyophobic areas of the surface of emerging particles, creating a structural-mechanical barrier that prevents inertial coagulation of particles, excluding the possibility of strong coagulation contacts, and hence strong high-viscosity dispersion structures. Thus, the vibration in the interfacial layer, placed on the phase interface by a vibrating element, prevents the formation of structure and provides a higher aggregative stability of the system.

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ID: 0901
MICROFLUIDIC SOLVENT EXTRACTION FOR AN AUTOMATED ^{68}Ga CYCLOTRON PRODUCTION LOOP

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Since the introduction of the first commercial ^{68}Ge/^{68}Ga generator about 20 years ago, interest in ^{68}Ga-labelled radiopharmaceuticals is steadily increasing. Currently, over 400 clinical studies including ^{68}Ga are being executed. Additionally, several PET imaging agents were approved for clinical use by the FDA, the latest being a new prostate-specific imaging agent, potentially leading to a significant increase in ^{68}Ga demand in the future. To date ^{68}Ga is still mostly produced by ^{68}Ge/^{68}Ga generators, but these generators show several drawbacks, such as a high amount of radioactive waste after the shelf-life of a generator is reached, the decreasing resources to produce ^{68}Ge and a limited supply of ^{68}Ga per day. To accommodate the increasing demand, alternative production routes are being investigated. Cyclotron-based production of ^{68}Ga, through proton irradiation of $^{68}Zn(NO_3)_2$ liquid targets, is therefore of increasing interest as an alternative. After irradiation, ^{68}Ga needs to be separated from the liquid target solutions, but lengthy, multi-step procedures involving ion exchange chromatography can lead to a significant loss of activity. Furthermore, the use of costly, enriched ^{68}Zn target material necessitates target solution recycling. In the present study, we developed a new, two-step solvent extraction process to recover ^{68}Ga from zinc nitrate solutions in under 15 minutes. Solution compositions can range from 1 - 5 M $natZn(NO_3)_2$ in 0.01 - 1 M HNO_3 and the organic extracting phase consisted of 200 mM N-benzoyl-N-phenylhydroxylamine dissolved in chloroform. In conventional batch extraction, efficiencies of up to 99.6 % ± 0.3 % were achieved within 10 minutes. Back-extraction into different HCl solutions resulted in efficiencies of up to 94.5 % ± 0.6 % within 1 minute. Additionally, microfluidic solvent extraction was investigated, due to its ability to be integrated in an automated set-up, opposed to conventional batch extraction. By microfluidic solvent extraction using a membrane separator, comparable results of 99.2 % ± 0.3 % for extraction and 95.8 % ± 0.8 % for back-extraction could be achieved. Zinc contamination in the final HCl solution after back-extraction, was found to be below 3 ppm and is thus within hospital quality standards. Because all zinc nitrate solutions were used without any modification, this method should allow for direct recycling of the target solutions after the extraction process making it a suitable method to create a fast, efficient cyclotron irradiation loop, that enables a continuous supply of ^{68}Ga.

ID: 0912
COMPLEXATION OF Eu(III) IN ARTIFICIAL DIGESTIVE MEDIA BY AMINOPOLYCARBOXYLIC ACID EGTA

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Radionuclides released from nuclear accidents can be widely distributed and even enter the human food chain. If ingested, they interact with the fluids from the digestive system and can cause necrosis or carcinogenesis of human cells. To remove these radionuclides, decorporation agents are used. Clinical approved decorporation agents like ethylenediaminetetraacetic acid (EDTA) or diethylenetriaminepentaacetic acid (DTPA) that are used against heavy metal poisoning show low oral activity.[1] Therefore, the focus of this work is set on EDTA and DTPA related compound ethylene glycol-bis(β-aminoethyl ether)-N,N,N′,N′-tetraacetic acid (EGTA). Europium(III) is used as a non-radioactive analogue for trivalent actinides like americium and curium. The overall goal of the study is to expand the knowledge of processes underlying the interactions of radionuclides in the human digestive system in the presence of decorporation agents on a molecular level. To derive thermodynamic parameters, the Eu-EGTA-system is investigated at different physiological relevant pH values and molar ratios, using time-resolved laser-induced fluorescence spectroscopy (TRLFS), NMR spectroscopy, electrospray ionization mass spectrometry (ESI-MS) and isothermal titration calorimetry (ITC). Between pH 3 and 9 a very stable complex with a Eu(III)-EGTA-ratio of 1:1 is observed and comprehensively characterised from the ligands and metals perspective. Our knowledge on the speciation of Eu(III) in artificial biofluids of the human digestive system is now extended by the impact of EGTA on the speciation to measure the suitability of EGTA as possible decorporation agent for usage in radiation protection.[2]

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ID: 0916
CHARACTERIZATION OF F-ELEMENT COMPLEXES WITH SOFT-DONOR LIGANDS FOR SELECTIVE AMERICIUM SEPARATION

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In recent years, much effort has been invested in recycling of minor actinides from PUREX raffinate solutions. Hydrophilic N-donor ligands such as tetrasodium-3,3',3'',3'''-([2,2'-bipyridine]-6,6'-diylbis(1,2,4-triazine-3,5,6-triyl))tetrabenzenesulfonate (SO_3-Ph-BTBP) are used for selective separation of Am(III) from Cm(III) and other trivalent fission lanthanides by smart combination with diglycolamide ligands as extractants such as N,N,N',N'-tetra-n-octyl diglycolamide (TODGA). In this work, the extraction behavior of SO_3-Ph-BTBP in combination with TODGA and its mono- and dimethylated derivates Me-TODGA and Me_2-TODGA was investigated. All systems show a high selectivity of An(III) over Ln(III), and Am(III) over Cm(III). Separation factors decrease in the order TODGA > Me-TODGA > Me_2-TODGA. Moreover, a significant drop in distribution ratios of heavier lanthanides (Tb-Lu) was observed in all three extraction systems. This effect was further elucidated by speciation studies of the formed complexes with several lanthanides using UV/Vis spectroscopy. The formation of 1:2 complexes (metal-to-ligand ratio) with SO_3-Ph-BTBP was confirmed for the studied lanthanides and stability constants log β of 7.7 ± 0.8 for Nd(III) at 10^{-3} mol L^{-1} HNO_3 and 6.1 ± 0.4 for Ho(III) at 3 mol L^{-1} HNO_3 were determined. No complexation was observed for Nd(III) at 3 mol L^{-1} HNO_3. Therefore, the formation of complexes with a protonated form of SO_3-Ph-BTBP seems to be possible with Ho(III), but not with Nd(III). The results of the extraction experiments and speciation studies will be presented and discussed in the poster.

ID: 0941
INSIGHTS IN THE BEHAVIOR OF AN EMERGING LIPOPHILIC PYTRI LIGAND FOR SANEX-LIKE PROCESSES

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Continuous efforts in research and development are resulting in new spent nuclear fuel (SNF) management strategies and technologies, that might be the pathway towards the sustainability and social acceptance of nuclear energy. Multi recycling of actinides is one of these paths, reducing the long-term heat load and radiotoxicity of SNF. In the last decades, many solvent extraction systems have been proposed for the hydrometallurgical separation of actinides. Owing to the outstanding properties of the hydrophilic counterparts, the lipophilic PyTri Ethyl-Hexyl (PTEH) ligand has been recently developed for the selective extraction of Am(III) and Cm(III) in SANEX (Selective ActiNide EXtraction) like processes[1,2]. Endowed with a satisfactory solubility in kerosene/1-octanol mixtures, PTEH exhibits a pronounced selectivity (SFAm/Eu ~ 100, SFAm/Ln ~ 60-340) and an extraction kinetics faster than the reference compound CyMe4-BTBP for SANEX-like processes. The extracting performance of the PTEH-based organic solvent was fully studied, also in presence of macro-concentrations of stable lanthanides (about 2.4 g/L) and in the temperature range of interest (20-50 °C) for the industrial application. In addition, qualitative and quantitative investigations on PTEH solutions irradiated up to 300 kGy by means of a ^{60}Co source or aged for several weeks demonstrated the remarkable radiolytic stability of the organic solvent. Furthermore, being crucial for the development of such class of ligands, the complexation and extraction mechanisms were investigated by different techniques. The first insight into the metal/ligand stoichiometry in the species formed upon complexation with PTEH was obtained by electrospray ionization mass spectrometry. Then, ligand stability constants for Cm(III) and Eu(III) were derived by Time Resolved Laser Fluorescence Spectroscopy, confirming the observed An(III)/Ln(III) selectivity. Finally, relevant insights into the binding of Lu(III) and Am(III) ions with PTEH were obtained by NMR investigations, indicative of a different bond nature. The achieved results highlight the noteworthy properties of PTEH-based solvents, confirming their potential to be used in SANEX-like processes and enable to acquire relevant information for further developing PyTri ligands.

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ID: 0953

FIELD EXTRACTION OF RADON FROM THE SPRING WATER INTO OLIVE OIL FOR HEALING PURPOSES

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Carcinogenity of residential radon (^{222}Rn) is already scientifically discredited. Positive correlation between radon and lung cancer is based on LNT (Linear-no-threshold-theory) only. No correlation here, when assumption-free statistics applied. On the other hand, radon is successfully used in balneology due to its strong hormetic effects. In radon balneotherapy, in some cases, the natural source activity for a stronger treatment may be inadequate low. Therefore, a field experiment was made to produce a highly active preparation, by extracting radon from water into olive oil, on a technical scale. The spring of Bretislav (2016) was used as a source of radon water (http://www.estudanky.eu/11000-radonka-pramen-bretislav). Its actual parameters were: Activity 12123 Bq/L of ^{222}Rn, flow rate 2.8 L/min, temperature 8.0 °C. Extraction was carried out in a “2 L” glass separating funnel. 2 liters of fresh water were always used. The starting amount was 120 mL of food-grade, non-virgin olive oil. The 165 mL bubble was left in the funnel to facilitate the mixing of the phases. A time snap was also taken from the all off extraction steps. After shaking (1 min), the phases were separated (2-4 min) and the water was poured, the gamma activity of the oil in the funnel was measured by sensitive scintilometer and a 1 mL sample was taken for later LSC determination of Rn on a field basis. Gradually, 9 subsequent extractions were performed over a total time of 1 h 27 min. a 76 mL of oil with an activity of 130 kBq/L of ^{222}Rn was obtained finally. Gama activity grew gradually, with an equilibrium level of 84 % after the ninth extraction. On the other hand, oil is saturated about 100 % already in the third extraction; in a total time of 23 min. Radon partition coefficient water / olive oil of 10.7 was found, although upto 45 reported in the broad literature. Radon oil was used by the author’s team for healing purposes in the evening.

ID: 0972

Th(IV) REMOVAL FROM AQUEOUS SOLUTIONS BY OXIDIZED BIOCHAR PREPARED FROM PALM TREE FIBERS

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The removal of Th(IV) from aqueous solutions by oxidized biochar fibres derived from palm tree fibers (OBF) has been investigated at pH 3 and under ambient conditions by batch type experiments and FTIR spectroscopy. The experimental data have shown that the Th(IV) adsorption by OBF is well fitted by the Langmuir isotherm model (qmax= 0.18 mol/kg or 42 g/kg), is an entropy-driven process and follows the 2nd order kinetics. Furthermore, FTIR spectroscopic data have indicated that the sorption occurs via formation of inner-sphere complexes between Th(VI) and the carboxylic surface moieties.

ID: 0974

POLONIUM REMOVAL FROM WATERS BY SILVER-COATED LUFFA CYLINDRICA BIOCHAR FIBERS

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The sorption of ultra-trace levels of polonium by Luffa Cylindrica biochar fibers has been investigated in de-ionized water and seawater samples prior and after surface modification (Ag-coating) of the adsorbent. The effect of pH on the sorption efficiency (Kd values) indicated that pH, which governs the solution chemistry of Po(IV), affects to a large degree the adsorption efficiency and that Ag-coating of the surface results in significantly higher Kd values. The modified adsorbent presents enhanced removal efficiency for ^{209}Po even from seawater samples indicating the usefulness of the modified biochar fibers for the treatment of polonium contaminated waters.

ID: 0978

ELECTROCHEMICAL OXIDATION OF TERBIUM(III) IN AQUEOUS MEDIA TOWARDS PURIFICATION OF MEDICAL TB-161

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Radiolanthanides are very promising for use in nuclear medicine. Due to their similar chemical properties, one and the same type of coordination compounds can find various diagnostic and therapeutic applications. Terbium has four radioisotopes with different decay properties, energies and half-lives and these can be used for noninvasive theranostic purposes. Carrier-free terbium-161 is produced via neutron irradiation of highly enriched gadolinium-160 targets in the Belgian Reactor 2 (BR2) at SCK CEN. Terbium-161 emits low-energy beta particles accompanied by gamma photons and shows similar properties to lutetium-177. Furthermore, the co-emission of Auger electrons make terbium-161 more attractive towards to a combined β^-/Auger electron therapy. Apart from the omnipresent trivalent oxidation state, several lanthanides can occur in the divalent or tetravalent oxidation states as well. A change in valence state alters the chemical properties and therefore facilitates intragroup lanthanide separations. The hydrated terbium(III) ion has a highly positive reduction potential (E^0 = + 3.1 V vs. SHE), but it can be oxidized to its tetravalent state via electrolysis and stabilized in highly concentrated carbonate solutions. In this study, Tb(III) is electrochemically oxidized to terbium(IV) in aqueous carbonate, nitrate and periodate media. Spectroscopic and elemental analyses were done to characterize Tb(IV) and to gain insight in the stability of these complexes in the aqueous electrolytes. A detailed parameter study on pH, terbium concentration, salt concentration and applied electrical potentials was performed. Nitrate and periodate media allow for oxidation at lower pH values than carbonate medium. The least positive applied potential value to oxidize Tb(III) was achieved in periodate medium at + 0.9 V vs. Ag/AgCl. However, nitrate and periodate media were poorly stabilizing Tb(IV), whereas the most efficient stabilization of Tb(IV) was observed in carbonate medium. Therefore, aqueous carbonate medium seem to be the most promising medium for developing a separation process based on the change in oxidation state of terbium. Oxidation of Tb(III) to Tb(IV) and the stability of Tb(IV) in carbonate media were confirmed and quantified by UV-Vis spectroscopy and XANES. Finally, Tb(IV) was separated from carbonate medium by solvent extraction and column chromatography.

ID: 0979

UNDERSTANDING THE PERTECHNETATE COMPLEXATION ON ENGINEERED BIOCHAR FOR ITS DETERMINATION BY AMS

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^{99}Tc ($T_{1/2}$ = 2.1·10^5 y) is an abundant product of the nuclear fuel cycle which is lying at the low-mass peak in the ^{235}U fission fragment spectrum. ^{99}Tc accounts for 6.1 % of the fission product mass in (^{235}U-enriched) nuclear fuels following burn-up in a reactor. In the environment, Tc also occurs due to nuclear bombs atmospheric testing and nuclear medicine. Tc is taken up by several plant species. Lichens and mosses retain technetium from airborne emissions, algae and seaweeds accumulate Tc from sea waters. TcO_4^- is the most common and the most environmentally mobile chemical species of Tc. Concentrations of Tc in the environment may vary depending on various factors (pH, microbial activity, amount of precipitations etc.). Many measurements of ^{99}Tc in the environment have been published which provide points of reference. These methods include decay counting, radiochemical neutron activation analysis, inductively coupled plasma mass spectrometry, thermal ionization mass spectrometry, laser resonance ionization mass spectrometry, and accelerator mass spectrometry (AMS). The most sensitive method is the AMS. However, the detection of ^{99}Tc represents some challenges. The most significant is the lack of stable isotope. Re as a chemical analog of Tc could be added in the sample preparation to act as a chemical carrier. A second difficulty is the stable isobar ^{99}Ru. A solid sample must be prepared for AMS measurement. The Tc obtained in solution is then chemically converted to a solid phase, the target matrix. There are currently several general approaches for TcO_4^- immobilization from waste such as evaporation, extraction-chromatographic methods, sorption onto carbon-based materials, and their modified forms, i.e. biochar, etc. A sorption is an effective approach to TcO_4^- sequestration because it is a simple, convenient, and economical in its own process. In this study we focused on the description of the interactions of pertechnetate and biochar using potentiometric titrations. The data were fitted with the surface-complexation model which includes multi-dimensional non-linear regression procedure. As the results show the main sorption mechanism of TcO_4^- on biochar surface is the ion exchange and chemical bonding.

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ID: 0981

APPLICATION OF ULTRA/NANO FILTRATION MEMBRANE ON MINING URANIUM FROM SEAWATERS

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Nuclear energy is a green and renewable energy. Uranium, as an important resource for nuclear energy, exists in seawater at a concentration of 3.3 μg/L, forming highly stable $Ca\text{-}UO_2\text{-}CO_3$ and $Mg\text{-}UO_2\text{-}CO_3$ complex. Though dilute, this amounts to an estimated 4.5 billion tons of uranium, which is approximately 1000 times more than that is available from conventional sources such as terrestrial ores. Uranium in seawater will be a near-limitless resource for nuclear fuel in the future, and its recovery will avoid the deleterious effects of terrestrial mining on the environment. Although there are many difficulties to overcome, mining of U from seawater is the most promising. The objective of this work is to test the possibility to separate Uranium from other salts in natural and recomposed water systems by ultra/nano filtration process realized with inorganic membranes, and to understand the rejection mechanism, the interaction of membrane with solution species. For this goal, various physical-chemical parameters, such as MWCO, pH value of solutions, temperature, applied pressure, ionic strength/salinity, solution composition, are evaluated for their influence on metal rejections. Electric repulsion and steric effect are two mechanisms deducing the interaction of solution species and membrane surface thus the rejections. Speciation distribution of each solution is studied with phreeqC software. Mass conservation law is applied to estimate the error. Concentrations of retentate and permeate are measured by ICPOES and ICPMS for determining the rejections. ConclusionElectric repulsion and attraction is the main mechanism of interaction between membranes and solutions species. pH is the principle factor which influences the rejection of every specie. There is a selectively big reject of U(VI) to Na at pH 3 and 8.25 in solutions of U and $NaHCO_3$. MWCO does not influence the rejections of U or Ca except that the IEP of different membranes can be slightly different. The salinity influences a lot the rejection of U in solutions presenting $NaHCO_3$ and NaCl (1-35g/L). U rejection decreases with the increase of salinity, but is still considerably rejected until salinity 10 g/L. With the $CaCl_2$ solutions of increasing concentration at pH 3 and the same for $MgCl_2$ at pH 3, the salt concentration influences a lot the rejection of both Ca and Mg. Both these experiments prove that high salinity shields the electric repulsion, by consequent decreases the rejection. On the other hand, U can be rejected from sodium in solutions contained only U and Na from $NaHCO_3$, NaCl or Na_2SO_4 at pH 8. U can be separated from sodium salts and be concentrated in a concentration experiment, so as Ca and Mg in the solutions respectively of $CaCl_2$ at pH 3 and of $MgCl_2$ at pH 3. Finally, filtration experiments with complicate natural and recomposed solutions including seawaters, Rhone River and theses solutions doped with U are conducted. In all the natural or recomposed seawater solutions, none of the species (U, Na, Mg, Ca, K) is rejected. However, it is proved that with the inorganic membranes, the presence of $CaCl_2$ or $MgCl_2$ hinders the rejection of every specie in solutions including U.

ID: 0982

RAPID SEPARATION OF ACTINIDES FROM WATER SAMPLES ON TEVA/TK221 STACKED COLUMNS

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Extraction chromatography is widely used in radioanalytical methods for the separation of actinides. The most commonly applied extractants for the separation of tetravalent actinides are quaternary amines (e.g., in TEVA resin), tetra- and hexavalent actinides are often separated with trialkyl phosphonates (e.g., in UTEVA resin), and tri-, tetra- and hexavalent ones can be separated using carbamoylmethylphosphine oxides (CMPO, e.g., in TRU resin), and tetraalkyldiglycolamides (DGA, e.g., in DGA resin). Recently, a mixed resin, the Triskem TK221 Resin that contains i.e. DGA and CMPO on a PS-DVB based inert support became available. Distribution coefficients (k') of actinides on this new resin were determined, k' values for actinides (Th, U, Pu, Am) were higher than 1000 from 3M HNO_3. This is a unique property of the new material offering excellent retention of all actinides, including trivalent ones such as Am(III). A novel method has been developed, tested and applied for the simultaneous determination of actinides from water samples using TEVA/TK221 stacked columns. Water samples (tap and sea water) were spiked with actinide tracers (Th, U, Np, Pu, Am). Calcium phosphate pre-concentration procedure was followed by dissolution in 3M nitric acid/1M aluminum nitrate and redox state adjustment was performed with sulfamic acid, ascorbic acid and sodium nitrite. Np(IV), Th(IV) and Pu(IV) were retained on TEVA resin, U(VI) and Am(III) were retained on TK221 resin. The stacked columns were separated and the actinides were sequentially eluted. Elution conditions were optimized. Alpha sources were prepared by NdF_3 micro-coprecipitation. Chemical recoveries were acceptable high (61-90 % for Th, 59-100 % for U, 92-93 % for Np, 87-100 % for Pu and 89-92 % for Am). No contamination in the actinide sources was detected. These promising results encourage experiments with more difficult matrices, such as soil and sediment samples.

ID: 0989

ESTABLISHMENT OF EXPERIMENTAL POSSIBILITIES FOR SEPARATION EXPERIMENTS ON GASEOUS H-ISOTOPE MIXTURES

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Hydrogen isotopes are studied in many research areas to clarify fundamental and applied aspects of their physicochemical behavior. Besides deuterium, the isotope tritium has become the focus of current investigations. One important application is the separation of hydrogen isotopes. Currently available methods have low separation efficiency and high energy consumption. Therefore, approaches to increase efficiency are presently being explored, with a focus on gaseous deuterium. In our work we will investigate two types of materials, on the one hand membrane materials (graphene and Nafion) and on the other hand nanoporous materials (metal-organic frameworks) with respect to tritium separation. For these studies, we needed to establish an analytical routine for working with gaseous tritium, from access to quantification methods. For this purpose, we employed a tritium manifold that uses heat to release gaseous tritium from a reservoir in a defined manner. The tritium gas under defined pressure and, in the future, mixtures of hydrogen isotopes will be used for separation experiments in flow cells. The gas will eventually be converted to HTO, which will be analyzed by liquid scintillation counting (LSC), where the measured activity directly corresponds to the concentration of tritium in the analyzed sample. To assess the quality of the proposed analytical method, the data obtained from the LSC measurements were compared with the calculated values and with the pressures applied during tritium dosing from the manifold. The measured values correspond directly to the applied pressures and agree well with the calculated data. These results indicate the proficiency of the established analytical approach, which allows to explore the proposed separation methods with high precision.

ID: 1011

OPTIMIZATION OF A CHEMICAL SEPARATION STRATEGY FOR TRIVALENT ACTINIDES FROM RARE-EARTH RICH DEEP-SEA ARCHIVES

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The understanding of the formation of the elements has been an intriguing topic within the last decades. It is now approved that the heaviest naturally occurring elements on earth, the actinides, are produced in the astrophysical r-process. However, the exact site of this process is still disputed. Recently, the amount of interstellar ^{244}Pu ($T_{1/2}$ = 80.6 Ma) in various geological archives like deep-sea ferromanganese crusts and sediments has been investigated by applying highly sensitive accelerator mass spectrometry measurements (AMS).[1,2] Correlation of the influx of supernova-produced ^{60}Fe ($T_{1/2}$ = 2.6 Ma) and ^{244}Pu could point to a possible origin of the r-process in the universe. To further prove this hypothesis, recent investigations focus on the determination of another long-lived radionuclide which is also produced in the r process, ^{247}Cm ($T_{1/2}$ = 15.6 Ma), by AMS. However, the separation of the expected ultra-trace amounts of actinides (a few 100 atoms per gram) from huge amounts of matrix and interfering elements represents a major analytical challenge. Thus, this contribution aims to compare existing chemical separation strategies for trivalent actinides (Am, Cm) from deep-sea reservoirs, like ferromanganese crusts or nodules based on extraction chromatography. Our investigations show that procedures based on trivalent actinide separation by TRU™ resin[3] fail to extract trivalent actinides from matrices with high concentrations of rare-earth elements. Thus, an alternative separation method based on anion exchange (DOWEX 1x8 for Pu separation) and solvent extraction (DGA™ resin for An/Ln separation and TEVA™ resin for the separation of Am/Cm from rare-earths) has been adapted in our studies.[4] ^{241}Am and ^{244}Cm in kBq quantities were used as tracers to determine the yield of the full separation procedure by γ-counting and α spectrometry. The effective separation of trivalent actinides from major matrix elements, like iron and manganese, as well as various rare-earth elements allow for processing multi-gram amounts of deep-sea ferromanganese crusts. This could finally lead to the detection of live ^{247}Cm in geological archives. Furthermore, this adapted method can be used for the analysis of environmental samples regarding their content and isotopic ratio of anthropogenically produced Pu, Am and Cm which holds potential for nuclear safeguards and nuclear forensics studies.

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ID: 1038

SORPTION OF ACTINIDES USING CHEMICALLY AND THERMALLY MODIFIED BIOSORBENTS

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AbstractThe removal of thorium and europium (as homologue for trivalent actinides) from aqueous solutions was explored using winery by-products in batch-type systems and at variable actinide initial concentrations. The modification of the winery by-products included chemical treatment with NaOH, Na_2CO_3 and NaCl, and biomass carbonisation. The investigations were performed by means of gamma-spectrometry using radioactive tracers and optical spectroscopy (UV-Vis). The results showed significant sorption capacity of the tested materials demonstrating their applicability in soil remediation and (waste)water treatment technologies.

ID: 1044

TREATMENT OF SPENT DECONTAMINATION SOLUTIONS BASED ON CITRIC ACID WITH OXIDIC NANOPARTICLES SORBENTS

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Composite sorbents based on nanoparticles of NiO, NiO-TiO_2 and ZrO_2-TiO_2 were chosen for treatment of spent decontamination solutions based on citric acid. Active oxidic materials were prepared by photoinduced synthesis (NiO, NiO-TiO_2) or hydrolytic method (ZrO_2-TiO_2). Weight distribution ratios for radionuclides ^{241}Am, ^{60}Co, ^{137}Cs a ^{90}Sr/^{90}Y and others were investigated in the course of the experiments performed in pH range 2–12. High sorption capacities up to 9 mmol·g^{-1} for ^{137}Cs were found by sorption isotherm experiments. Dynamic experiments were performed with simulated spent decontamination solutions based on citric acid. Elution of radionuclides was tested with mineral acids (HCl, HNO_3, $HClO_4$) or complexing agents (citric acid, Na_3NTA, HEDP). Under suitable conditions, 85 % of radionuclide was eluted in 6-times lower volume compared to treated spent solution. Bleeding of Ni ions from active components was detected during half-scale test due to ICP-MS analysis.

ID: 1049

TRACER-SCALE MO AND W EXTRACTION IN THE CYANEX 600/NITRIC ACID SYSTEM

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The work deals with a complex topic of liquid-liquid extraction of Mo and W from the Sg point of view, which puts emphasis on extraction rate and efficiency. The topic can be divided into three main areas of focus: exploring the possibility of employing industrial grade extraction agent Cyanex 600 for extraction of group 6 elements from nitric acid solutions, suggesting the extraction mechanism of both Mo and W in the Cyanex 600/HNO_3 system, and describing the system’s behaviour in sub-minute continuous extraction process using microfluidic techniques. The Cyanex 600/HNO_3 system characterization revealed similarity with extraction mechanism of organophosphorus acids. The mechanism of Mo extraction with Cyanex 600 was established and apparent extraction constants of three pH-dependent extraction sub-processes were calculated. Although it was not proved with absolute certainty that the mechanisms for W and Mo are the same, results for W extraction were determined identically. In addition, the data analysis provided value of apparent dimerization constant of Cyanex 600 in kerosene. Based on its value, and the mechanism itself, it was revealed that main component of Cyanex 600 might not be Cyanex 272 as anticipated, but rather its dithio-derivate Cyanex 301. Microfluidic system for fast extraction was successfully employed, and yielded aqueous-to-organic overall volumetric mass transfer coefficients that quantify kinetic performance of the system under given conditions. The region between 0.1 and 0.01 M HNO_3 was identified as the most promising for potential Sg application for its fastest kinetics and extraction efficiency for both Mo and W. Furthermore, two microfluidic mixing techniques were compared: extraction in a capillary (inner diameter of 250 µm) and in a micromixer chip. Both were shown to have almost identical kinetic performance.

ID: 1056
LIQUID-LIQUID EXTRACTION OF RADIOSTRONTIUM INTO IONIC LIQUIDS USING CROWN ETHERS

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Chemical decontamination of nuclear facilities generates large volumes of low-level radioactive waste. Following worldwide efforts to reduce spent chemical reagents and radioactive waste volumes, new methods of regeneration and reuse of spent decontamination solutions are studied. This work focuses on the usage of ionic liquids (ILs) as an organic phase in solvent extraction of strontium from solutions containing complexing agents. ^{85}Sr or ^{89}Sr were used as radioactive tracers and the extraction efficiency was evaluated by distribution ratio calculated from count rates of the respective phases. Extractions from HNO_3 solutions into $[C_4mim][NTf_2]$ using various extraction agents were performed. The best results were achieved with dicyclohexano-18-crown-6 ($DCH_{18}C_6$), which has then been used as an extractant in the detailed study of strontium extraction into ILs. High dependence of the extraction efficiency of strontium on the presence of other cations was found. The highest decrease of distribution coefficient was observed in the presence of K^+ ions. The kinetics of the extraction and the dependence of the distribution ratio on the time of extraction was studied. Experiments carried out from solutions containing complexing agents indicate that $DCH_{18}C_6$ could be used to effectively extract strontium from these solutions.

ID: 1061
EXTRACTION OF MOLYBDENUM FROM THE NUCLEAR CYCLE WASTE STREAMS USING IONIC LIQUIDS

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Isotopes of molybdenum, the fission products arising from ^{235}U fission, represent one of the contaminants in liquid radioactive waste generated in the nuclear fuel cycle. With regard to the half-life of ^{93}Mo, this isotope becomes important for safety assessment for disposal sites in long term of deposition of radioactive wastes. Conventional processes for its separation are mostly based on liquid-liquid extraction using common organic solvents such as chloroform. In this work, ionic liquids were used for the extraction studies of molybdenum. Due to their unique properties, ionic liquids provide number of advantages, including the possibility of radionuclides re-extraction and ionic liquid reusing. Radionuclide ^{99}Mo for extraction studies was obtained from the ^{99}Mo-^{99m}Tc radionuclide generator system. Liquid-liquid extraction using 8-hydroxyquinolin in $[C_4mim][NTf2]$ as the organic phase was performed to study the dependence on the equilibrium pH of aqueous phase. Various concentrations of HNO_3, NaOH and phosphate or borate buffers were used for the pH adjustment. Solution with oxalic or citric acid or their mixtures were used to test the possibility of extraction from the organic complexing agents environment. For the comparison with traditional organic solvent, chloroform was used as the organic phase under the same conditions. The samples were measured using the NaI(Tl) scintillation counter.

ID: 1070
PREPARATION AND CHARACTERIZATION OF A-ZIRCONIUM PHOSPHATE AS A PERSPECTIVE MATERIAL FOR SEPARATION OF MEDICINAL RADIONUCLIDES

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Zirconium phosphate (ZrP) has been the subject of research for many years. The interest for this material is based on its outstanding physicochemical properties, both in its amorphous and crystalline phases. It is necessary to unequivocally highlight its extremly high ion-exchange capacity, very good biocompatibility and stability, and good thermal and radiation stability. At the same time, it should be noted that the synthesis of ZrP and its subsequent functionalization is very simple. All of this makes ZrP perspective candidate for a wide range of applications in chemistry, health and nuclear industry, but especially in nuclear medicine area for example as a potentional drug delivery system, ion-exchanger for sources of high purity radionuclides or photovoltaics[1]. Zirconium phosphate, specifically its alpha alotropic modification (α-ZrP), was prepared by refluxing an aqueous solution of oxychloride octahydrate and sodium dihydrogenphosphate monohydrate solution in hydrochloric acid[2]. The prepared α-ZrP was completely characterized using infra red and Raman spectroscopy, X-ray powder difraction, DTA and TG analysis, FEGSEM/SE, TEM/BF 2D-SAED and TEM/EDX analysis. Finally, experiments studying mechanism of sorption on a surface of α-ZrP and characterizing the surface of α-ZrP by its active soprtion sites and functional groups using potentiometric titrations in pH range 2-11 was carried out. The experimental data was analyzed using software FAMULUS. To sum up, it can be said that prepared α-ZrP was completely characterized and sorption mechamism on the surface of this material and its surface characteristics were studied. The titration curve of ZrP is placed in an area with a predominant negative surface charge. ZrP also has high concentration of functional groups for sorption of cations, indicating a potentially high sorption capacity. From this point of view, ZrP appears as promising and interesting for next studies with various purposes like drug delivery systems or ion-exchangers for separations of medicinaly significant radionuclides such as radioactive pair ^{225}Ac and ^{213}Bi.

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ID: 1071

SEPARATION OF Fr-221 FROM Ac-225 USING DIGLYCOLAMIDE SOLID EXTRACTANTS

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Diglycolamides are frequently used as extraction agents in many fields of nuclear and separation chemistry including nuclear waste management, nuclear fuel reprocessing, radioanalytical chemistry, radioecology or nuclear medicine. They are tridentate ligands with extraction abilities to extract trivalent ions, mainly lanthanides and actinides[1]. This ability can be beneficial in separation of radionuclides for nuclear medicine. In practise, diglycolamides are used for example in separation of clinical ^{223}Ra from ^{227}Ac/^{227}Th/^{223}Ra mixtures[2]. Another important medicinal radionuclide of recent years, ^{255}Ac, is separated from its mother radionuclide ^{225}Ra using diglycolamides as well[3]. ^{225}Ac is a radionuclide of great potential for targeted alpha particle therapy. Its decay initiates the cascade of 5 alpha decays in total. The second of its decay products, ^{213}Bi, is another radionuclide in the focus of nuclear medicine research at the moment. The separation of ^{213}Bi cannot be sufficiently performed using diglycolamides because of its chemical properties. However, the first decay product of ^{225}Ac transmutation is ^{221}Fr with a half-life of 5 min decaying to ^{213}Bi. As a monovalent ion it could be easily separable from ^{225}Ac. In this work two most common diglycolamides, TODGA and isoTODGA, were tested for separation of ^{221}Fr from ^{225}Ac in preliminary studies. Diglycolamides were immobilised on polyacrylonitrile beads to form solid extractants TODGA-PAN and isoTODGA-PAN. The loading of such solid extractants was 40 %. Batch sorption experiments with both solid extractants were performed in both hydrochloric and nitric acid media in the concentration range of 0.001-8M. The weight distribution coefficients of ^{225}Ac were determined. As expected, especially high weight distribution coefficients were achieved for both solid extractants in higher concentrations of nitric acid. Subsequently, the most promising conditions of ^{225}Ac sorption were applied in column experiments in order to evaluate ^{221}Fr elution. The initial activity of ^{225}Ac was 4.5 MBq and column bed volume was 0.6 ml. The yield of ^{221}Fr elution was kept over 65 % in 2.5 ml of eluate for the whole period of the long-term study. The contamination by ^{225}Ac was kept under 1.0 % and getting lower during the time of study. To sum up, the proof of concept has been given in this preliminary study. However, in order to minimize ^{225}Ac contamination, the parameters of proposed separation system should be optimized.

This work was supported by Technology Agency of the Czech Republic (TJ04000129) and Czech Technical University in Prague (SGS22/188/OHK4/3T/14).

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10. Chemistry of Actinide and Trans-actinide Elements

ID: 0879

SYNTHESIS, CHARACTERIZATION AND STABILITY OF TWO AMERICIUM VANADATES, $AmVO_3$ AND $AmVO_4$

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The americium isotope ^{241}Am is formed during the storage of plutonium via beta-decay of ^{241}Pu with a half-life of 14.33 years. Due to its accumulation in existing stocks of civil separated plutonium in Europe, and its relatively high specific power of 0.114 W/g, ^{241}Am has been proposed for use in radioisotope power systems (RPS) and is under consideration by the European Space Agency (ESA) as an energy source for future European space missions. The requirements for a stable solid form are very diverse with respect to storage on earth, operation in space and safety performance in case of accidents and post-accident scenarios. Our group studied several ceramic forms containing significant specific Am-amounts (such as uranium-stabilized dioxide, phosphate, or aluminate). In this context, $AmVO_3$ and $AmVO_4$ were prepared by a solid-state reaction and their room-temperature crystal structures were solved by powder X-ray diffraction combined with Rietveld refinement. Spectroscopic (Raman) and microscopic (Scanning Electron Microscopy) studies were carried out in order to check the purity of the samples. The oxidation states of americium were confirmed by High-Resolution X-ray Absorption Near-Edge Structure (HR-XANES) technique. The stability of $AmVO_3$ and $AmVO_4$ under self-irradiation and heat treatment in inert and oxidizing atmosphere was tested and discussed relative to the available literature data and to other americium-containing ceramics.

ID: 0892

SPECTROSCOPIC INVESTIGATION OF THE COVALENCE IN An(III) COMPLEXES WITH PICOLINDIAMIDES

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N-donor and O-donor ligands form strong, ninefold coordinated complexes with trivalent actinide and lanthanide ions. These excellent coordination properties make them suitable for the application as extractants and are interesting systems to study from a fundamental scientific perspective. Extraction studies have shown that N-donor ligands (e.g. nPr-BTP) are suitable for the selective separation of An(III) from Ln(III). The selectivity stems from a different metal-ligand interaction and thus leads to significant differences in complex stability between the Ln(III) and An(III) complexes. In O-donor complexes (e.g. TODGA), stability constants and extraction properties are quite similar. In conclusion, the An(III)-O and Ln(III)-O binding properties are expected to be equivalent. Considering this, the question arises how the metal-ligand interaction changes if structural features of N- and O-donor ligands are combined. For that reason, we present a spectroscopic study of the complexation of Ln(III) and Am(III) with the N,O-donor ligand N,N,N',N'-tetraethyl-2,6-carboxamidopyridine (Et- Pic). By using ^{13}C and ^{15}N NMR spectroscopy, we have found slight differences in the Ln(III)-N and Am(III)-N interaction. In contrast, the Ln(III)-O and Am(III)-O bond in the $[M(Et\text{-}Pic)_3]^{3+}$ (M = Ln, Am) complex shows similar binding properties. In addition, we have observed significant differences in complex stability constants by using time-resolved laser fluorescence spectroscopy (TRLFS). TRLFS results show that Cm(III) forms a stronger 1:3 complex by one order of magnitude compared to the respective Eu(III) complex. Therefore, Et-Pic shows a minor selectivity towards An(III) ions. The data suggests that the observed selectivity descends solely from an increased partial covalent interaction in the An(III)-N bond.

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ID: 0893

TRIVALENT ACTINIDE IONS SHOWING TENFOLD COORDINATION IN SOLUTION

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Trivalent actinides generally exhibit ninefold coordination in solution. nPr-BTP, a tridentate nitrogen donor ligand, is known to form ninefold coordinated 1:3 complexes, $[An(nPr\text{-}BTP)_3]^{3+}$ (An = U, Pu, Am, Cm) in solution. We report the first Cm(III) complex with tenfold coordination in solution $[Cm(nPr\text{-}BTP)_3(NO_3)]^{2+}$. This species was identified using time-resolved laser fluorescence spectroscopy (TRLFS), vibronic side band spectroscopy (VSBS), X-ray photoelectron spectroscopy (XPS) and density functional theory (DFT). Adding nitrate to a solution of the $[Cm(nPr\text{-}BTP)_3]^{3+}$ complex in 2-propanol shifts the Cm(III) emission band from 613.1 nm to 617.3 nm. This bathochromic shift is due to a higher coordination number of the Cm(III) ion in solution, in agreement with the formation of the $[Cm(nPr\text{-}BTP)_3(NO_3)]^{2+}$ complex. No such shift is observed when adding other anions (NO_2^-, CN^- and OTf^-). The formation of the $[Cm(nPr\text{-}BTP)_3(NO_3)]^{2+}$ complex was studied as a function of NO_3^- and nPr-BTP concentrations. Slope analyses confirm the addition of one nitrate anion to the $[Cm(nPr\text{-}BTP)3]^{3+}$ complex. Experiments with varied nPr-BTP concentration show that $[Cm(nPr\text{-}BTP)_3(NO_3)]^{2+}$ only forms at nPr-BTP concentrations below 10^{-4} mol/L. For concentrations greater than 10^{-4} mol/L only $[Cm(nPr\text{-}BTP)_3]^{3+}$ is observed. VSBS allows observing vibrations of coordinated functional groups. The vibronic side band of the $[Cm(nPr\text{-}BTP)_3(NO_3)]^{2+}$ species exhibits a nitrate stretching mode not observed in the $[Cm(nPr\text{-}BTP)_3]^{3+}$ complex. Moreover, XPS on $[Am(nPr\text{-}BTP)_3(NO_3)]^{2+}$ yields signals from both non-coordinated and coordinated nitrate. DFT calculations reveal that the energetically most favoured structure is obtained if the nitrate is positioned on the C_2 axis of the D_3 symmetrical $[Cm(nPr\text{-}BTP)_3]^{3+}$ complex with a bond distance of 413 pm. Combining results from TRLFS, VSBS, XPS and DFT provides sound evidence for a unique tenfold coordinated Cm(III) complex in solution - a novelty in An(III) solution chemistry.

ID: 0903
5F ELECTRON POLARIZABILITY IN A FULL EARLY ACTINIDE (TH-PU) PYREN COMPLEX SERIES

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In contrast to the strongly shielded 4 f electrons of the lanthanides, 5f electrons of particularly the early actinides are found to participate in bonding, e.g. to organic ligands. Reactivity and complexation strength of such bonds are the most influenced by donor properties of the ligand and the electronic situation of the actinide metal center. Furthermore, coinciding properties of ligand and actinide ion regarding Pearson's principle of hard and soft acids and bases (HSAB) can even drive the development of selective ligands, e.g. for extraction processes. Here, soft N-donor ligands were found to interact stronger with trivalent actinides in comparison to their harder lanthanide analogues.[1] To evaluate how these HSAB properties can be extended to a series of tetravalent actinides and their interactions with N-donor ligands, we have studied the complexation of tetravalent Th, Pa, U, Np, and Pu with N, N'-ethylene-bis((pyrrole-2-yl)methanimine (pyren) in comparison to its structural N, O-analogue, the salen ligand.[2] Complex syntheses using one equivalent of $AnCl_4(dme)_x$ (An = Th, U, Np, Pu; x = 0 for U, x = 2 for Th, Np, Pu) and two equivalents of pyren led to isostructural 2:1 complexes, which were analyzed in the solid state by SC-XRD and IR, as well as in solution via NMR spectroscopy. SC-XRD results and quantum chemical calculations revealed differences in An^{IV}–ligand bond length and strength within pyren (N_{imine} vs. $N_{pyrrolide}$ donors) or salen (N_{imine} vs. $O_{phenolate}$). Interestingly, the overall bond strength of the N-donor vs. N, O-donor to An(IV), however, is almost equal for both, $[An(pyren)_2]$ and $[An(salen)_2]$ (An = Th-Pu). Delocalization indices even confirmed slightly more covalent interactions between the N, O-donor salen and Th, U, Np, and Pu in comparison with pyren. For Pa, on the other hand, this trend is reversed. QTAIM analysis could prove particularly strong interactions with the pure N-donor ligand pyren. This extraordinarily good electron sharing between pyren and Pa can be explained by the 5 f^1 configuration of Pa(IV), being particularly well polarizable and thus well suited for an effective backbonding to the soft N-donors of the pyren ligand.

ID: 0915
SYNTHESIS AND COMPLEXATION OF NITROGEN DONOR LIGANDS WITH TETRAVALENT URANIUM

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The knowledge of complexation reactions of early actinides with nitrogen donor ligands serves not only as fundamental research in this underrepresented field of chemistry but also contributes to a deeper understanding of their reactivity and coordination chemistry. In contrast to the lanthanides, with the dominating oxidation state of +III, actinides, especially the early actinides up to Plutonium, can exist in a variety of different oxidation states ranging from +II to +VI. The coordination chemistry of tri- and tetravalent actinides with selective soft nitrogen donor ligands is of special interest, with a potential use as extraction and/or decontamination agents. In order to understand the bonding trends and electronic structure, the nitrogen donor ligand 2,6-bis(1-(4-bromo-2,6-dimethylphenyl)-1H-1,2,3-triazol-4-yl)pyridine, a BPTP-type ligand, was used as the compound of choice in this contribution. BPTP-type ligands are based on the commonly known BTP-ligand, a tridentate chelating ligand which was designed for the purpose of separating lanthanides from actinides.[1] The BPTP ligand was synthesized by a copper mediated click reaction of 2,6-diethynylpyridine with the corresponding azide. Within this ongoing study, we focus on the synthesis and characterization of tetravalent actinides, which are readily available for all of the early actinides from Thorium up to

Plutonium. The obtained U(IV) complex was characterized by single crystal X-ray diffraction (SC-XRD). Further characterization of the novel coordination complex using NMR, IR, and EPR, as well as an expansion to the transuranic elements will complete this study.

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ID: 0920

THE USE OF ^{152}Eu(III) TOWARDS THE ELUCIDATION OF Pu(III) BEHAVIOR WITH PHOSPHATES

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Phosphate species from natural (e.g. phosphate mineral like apatite) as well anthropogenic (e.g. fertilizers) sources are ubiquitous in the environment[1]. Phosphoric acid is also present in the PUREX process as ultimate degradation product of tributylphosphate[2]. Phosphate species are expected to strongly interact with plutonium ions, plutonium being one of the main environmental pollutants associated with nuclear power generation and nuclear weapon testing[3]. In order to improve the models of Pu chemical behavior in the presence of phosphate species, it is necessary to collect fundamental data on interaction of Pu_3^+, Pu_4^+, PuO_2^+, and $Pu_2O_2^+$ ions with these inorganic ligands. However, due to difficulties in handling Pu (redox properties, radiotoxicity), the aim is first to develop an experimental protocol using analogs in the same oxidation state[4]. The present work focuses on the determination of, on one hand, the speciation of Eu(III) at ultra-trace scale using ^{152}Eu in the presence of $H_2PO_4^-$ and, on the other hand, of the solubility product of Nd-based rhabdophane. The partition of ^{152}Eu in the system HDEHP/Toluene/NaCl/HCl/NaH_2PO_4 has been studied at constant ionic strength and temperature. The mean composition of the formed complexes and their associated stability constants have been deduced from the variations of the distribution ratio of ^{152}Eu as function of phosphate concentration. Concerning the solubility product of $PuPO_4$, stable rhabdophane $Nd_{0.99}Eu_{0.01}PO_4 \cdot nH_2O$ and the ^{152}Eu doped one have been synthesized. The stable solid has been characterized by XRD, TGA and SEM, and its dissolution has been proved to be congruent. Dissolution experiments have been then conducted on the doped phosphate compounds reducing the amount of solid (from 50 to 2 mg). The process was monitored by γ-spectrometry. The same behavior was observed in all the studied systems: the equilibrium is reached in about ten days and the solubility product obtained in these conditions agrees with literature data[5]. This validates the use of a small amount of $PuPO_4$ in future dissolution experiments.

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ID: 0933

SYNTHESIS AND CHARACTERIZATION OF TETRAVALENT ACTINIDE AMIDINATE HALIDE COMPLEXES

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Amidinate ligands have attracted considerable attention in the field of coordination chemistry over the last decades as versatile soft N-donor ligands to stabilize the transition metal complexes in various oxidation states. The additional advantages of employing amidinates over other ligand systems are their high modularity and easy access to a variety of analogs by altering the substitution patterns through straightforward synthetic procedures. There have been a number of studies on the transition metal amidinate complexes including lanthanides, and even some early actinides. Still, these studies are mainly limited to thorium(Th) and high valent(V, VI) uranium(U) complexes. Here we focused on the interaction of An(IV) complexes (An= Th, U, and Np) with benzamidinate ligands to provide a comprehensive understanding of the electronic properties of actinide compounds. In this study, we successfully synthesized a series of tetravalent actinide tris-benzamidinate chloro complexes [AnCl(amid)$_3$] (An= Th, U, and Np; amid= iPr$_2$BA and (S)-PEBA). Furthermore, we also obtained additional halide complex series (F, Br) by halogen exchange reactions on chloro complexes as precursors to investigate the conformational stability of the complex. The crystal structures of the model actinide complexes were determined by SC-XRD, showing three benzamidinates and one halide ligand coordinated to the actinide metal center in a monocapped distorted octahedral coordination geometry, resulting in a propeller-like shape with the halide lying on the rotation axis. The actinide complexes were also characterized in solution by using paramagnetic NMR spectroscopy to elucidate structural and chemical bonding situations with an increasing number of unpaired electrons along the 5f series.

ID: 0943

ADVANCEMENTS IN THE FABRICATION AND CHARACTERIZATION OF ACTINIDE TARGETS FOR SUPERHEAVY ELEMENT PRODUCTION

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All isotopes of the elements with atomic numbers 114 and above as well as many of the longer-lived isotopes of lighter superheavy elements are only accessible in fusion reactions with actinide targets. Accordingly, the production of actinide targets is an important topic in the field of SHE research, especially with new accelerator facilities coming online that deliver ever more intense heavy-ion beams. After a brief review the current status of actinide target production with a focus on the activities at Johannes Gutenberg University Mainz, new developments towards the production of thicker and more beam-tolerant targets will be presented. Main current activities focus on improved analytics of irradiated targets to better understand beam-induced modifications and the effects that ultimately lead to target failure, and on the development of novel electrochemical methods that promise to allow overcoming some of the limitations of the current standard technique of molecular plating.

ID: 0945

OFF-LINE SINGLE-ATOM GAS CHROMATOGRAPHIC ADSORPTION STUDIES OF BISMUTH

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In preparation of gas phase chemical experiments with element 115, we studied the chemical behavior of single short-lived bismuth atoms in rare gases (helium and argon) and in oxygen atmosphere. For that purpose, we performed off-line isothermal gas chromatography experiments at room temperature. The short-lived volatile ^{219}Rn precursor, provided from an ^{227}Ac-source, was transported into the miniCOMPACT setup (mini Cryo-Online Multi detector for Physics And Chemistry of Transactinides) with different carrier gases. Chromatograms of ^{211}Bi were recorded as a function of various parameters like carrier gas type and gas flow rate, thus characterizing the novel miniCOMPACT detector array. This aids to optimize the conditions for future experiments with superheavy elements. All measured species deposited on the SiO_2 surface of the miniCOMPACT via diffusion-controlled deposition. Furthermore, we performed adapted Monte Carlo simulations, which account for the precursor effect, and compared them to experimental results to determine the lowest limit value of the adsorption enthalpy of bismuth on silicon dioxide.

ID: 0952

FUNDAMENTAL BEHAVIOR OF METALS IN THE PRESENCE OF NON-CONVENTIONAL SOLVENTS

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Understanding of fundamental aspects of metal behavior is a key component of controlling the separation and purification process. This knowledge is based on speciation and oxidation state information leading to numerical thermodynamic parameters[1-3]. Surprisingly, there are still chemical systems with unknown stability constants of metals in the generic mineral acids. However, this situation becomes more complex when a new class of compounds is introduced, so-called non-conventional solvents[4]. This new approach utilizes application of ionic liquids and deep eutectic mixtures. The former is a salt with a melting temperature below 100 °C and they are composed of discrete ions. The latter is a mixture of at least two compounds, resulting in a melting temperature depression in comparison with that of individual moiety, and they consist of hydrogen bond donors and acceptors. These new systems also can be hydrophobic and applicable for metal extraction[5-7]. Recent results on In, Tl, Rh, and Ir behavior in the presence of non-conventional solvents will be discussed.

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ID: 0998

EVALUATION OF ADSORPTION ENTHALPIES IN ISOTHERMAL CHROMATOGRAPHY EXPERIMENTS UNDER NON-OPTIMAL CONDITIONS - THE CASE OF TL AND TLOH ON FUSED SILICA

RUGARD DRESSLER[a], **JENNIFER WILSON**, **ROBERT EICHLER**[a,b], **PATRICK STEINEGGER**[a,c]

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The determination of the enthalpy of adsorption of an analyte on a defined stationary surface is one of the most important methods for the determination of chemical properties of superheavy elements (SHEs). Isothermal gas-adsorption chromatography setups have been used in various experiments to chemically study compounds of rutherfordium (Rf, Z=104), dubnium (Db, Z=105), seaborgium (Sg, Z=106), and bohrium (Bh, Z=107). In all these experiments, Monte Carlo simulations were used to derive the adsorption enthalpy for the investigated chemical system from the experimental data. This approach allows for predicting the breakthrough yield of a chemical species on a specific stationary phase as a function of the applied fixed experimental parameters e.g. gas flow and temperature profile. Including the decay (half-life) of the used radioisotope, leaves the enthalpy of adsorption as the only free parameter in the model. If the measurements can be performed over a sufficiently wide temperature range, saturation of the breakthrough yields at about 100 % can be achieved. Therefore, data evaluation based on relative chemical yields of the external chromatograms was introduced in the past. These relative chemical yields were calculated with respect to a reference measurement at sufficiently high temperature, assumed to represent the 100 % breakthrough yield. The adsorption enthalpy was then deduced by determining the Monte Carlo simulation that reproduced the experimentally observed external chromatogram best. Recently, in preparation for experiments with nihonium (Nh, Z=113), model studies were performed with its lighter homolog thallium, using both isothermal vacuum as well as isothermal gas-adsorption chromatography. It became evident that it was difficult to reliably identify the saturation level of the external chromatogram for the species studied due to experimental limitations regarding the maximum temperatures, isothermal temperature profile stability as well as the statistics of the detected events. All of these limitations are clearly due to non-optimal experimental conditions and make the use of established data analysis questionable. This has motivated the development of an algorithm to predict the breakthrough yield independently of a reference measurement, which can be used in connection with Monte-Carlo simulation to estimate the enthalpy of adsorption. Here, we will present this evaluation algorithm and its application to isothermal chromatography experiments using the example of past experiments performed at the JAEA tandem accelerator in

Tokai-mura (Japan) and at the JINR 400U Cyclotron in Dubna (Russia).

ID: 1026

ONLINE GAS ADSORPTION CHROMATOGRAPHY OF TL/TLOH FOR FUTURE NH/NHOH EXPERIMENTS

JENNIFER WILSON[a], **ALEXANDER BODROV, GOSPODIN BOZHIKOV**[b], **NIKOLAY AKSENOV**[c], **NATALIA GUSTOVA, VLADAMIR ZOBNIN, ALEXEY SABELNIKOV, DOMINIK HERRMANN**[d], **MARINA VORONYUK, ALEXANDER SVIRIKHIN, ALEXANDER MADUMAROV**[b], **OLEG MALYSHEV, ILYA CHUPRAKOV, VICTOR IVANOVIC CHEPIGIN, YURY ALBIN**[b], **BERNOIT GALL, ANDREY ISAEV, ANDREI ASTOKHOV, ROBERT EICHLER**[d,e], **ZOUHAIR ASFARI, YURA POPOV, SERGEY DMITRIEV**[b], **ELIZAVETA PISHCHALNIKOVA, PATRICK STEINEGGER**[d,f]

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To adequatly chemically characterize superheavy elements, such as nihonium (Nh, Z=113) which has a production rate on the order of atoms per day, thorough prestudies must be completed. Gas adsorption chromatography of cyclotron-produced ^{184}Tl were conducted as preptory experiments for future Nh chemistry studies. ^{184}Tl (5 neutron evaporation channel) was produced at the ^{400}U cyclotron facility of the Flerov Laboratory of Nuclear Reactions (FLNR) from a 255 MeV ^{48}Ti ion beam bombarding a ^{141}Pr target (prepared on a 1.5 µm Ti-backing). Recoiling nuclear reaction products were velocity-separated in vacuum with the SHELS separator before entering the chemistry setup by passing through a 7 µm thin Mylar window. There, the isotopes are thermalized in a concial, Ar-flushed recoil transfer chamber which tapers to a 4 mm fused silica column. In the downstream direction of the column a hot tantalum foil (1000 °C), a reactive gas inlet (O_2 + H_2O), the isothermal chromatography oven (temperature range from room temperature – 850 °C), and a quartz wool plug aligned with a HPGe γ-ray detector are placed. A key component of the setup was the hot tantalum foil placed after the recoil transfer chamber but before the inlet. This ensured all ^{184}Tl entering the isothermal chromatography section is in the metallic ground state. It was expected that the interaction between Tl and the hydroxyls on the fused silica surface readily promote the TlOH formation, as stated by previous studies. Unexpectedly, the influence of fused silica dehydroxylation can been seen on the external chromatogram. Shown here is the analysis of the experimental data employing isothermal gas adsoption chromatography and its impact for designing future experiments targeting an unambiguous chemical characterization of Nh.

ID: 1031

CHEMISTRY WITH SUPERHEAVY ELEMENTS AT FLNR AND AT PSI

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The Superheavy Element Factory in Dubna offers unique prospects regarding future chemistry experiments with superheavy elements. The very first chemistry experiment at the Superheavy Element Factory will focus on the chemical characterization of elements copernicium (Z=112) and flerovium (Z=114) in their respective elemental state, employing online gas adsorption thermochromatography. Further efforts are directed towards the first unambiguous chemical characterization of nihonium, based on through preparatory experiments with the lighter homolog thallium. Here, we will present the planned commissioning experiment with copernicium and flerovium, followed by a status report regarding the chemical characterization of nihonium as well as future plans.

ID: 1057

SEPARATIONS OF THE HEAVIEST ACTINOIDS

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Overall goal of this project is to contribute to the knowledge of the heaviest actinoids. The research was carried out in two main directions: development of a method for radiochemical separation of actinoid series by liquid chromatography, including a model for determining separation parameters in non-equilibrium processes at one-atom-at-a time level (JINR Dubna), and novel methods for An(II) and An(III) separations for the studies of Md, No and Lr, including assembly of a setup for the electrochemistry studies of NCA radionuclides (CTU in Prague). During development of the model algorithm for describing the chromatographic peak, the numerical integration of the mass transfer equations with and without diffusion was performed under conditions corresponding to the processes in the CIX - α-HIB chemical system. The main result of this work is the proposed machine learning algorithm for determining the distribution coefficients of α-HIB for Am, Cm and Cf. Chromatographic experiments were carried out on a glass column at room temperature. The cation exchange resin was used as a sorbent. The initial solution of α-HIB at a given pH containing the radioisotopes Am, Cm, and Cf was fed into the column under an inert gas. The equilibrium concentration of the ligand at a given pH of the solution was calculated via the α-HIB dissociation constant. Analysis of the data obtained on the effect of diffusion on the chromatographic process allows to conclude that the process is in equilibrium when studying the behavior of actinide at micro-concentrations in the cationite - α-HIB system. To simulate the conditions in experiments with transfermium elements, the studies of methods for An(II) and An(III) separations were performed in a radiochemistry laboratory at the U-120M cyclotron (Institute of Nuclear Physics, Řež) equipped by a He gas-jet system. The nuclides used were produced by irradiating self-supporting metallic target foils with $3He_2^+$ ion-beam. Products recoiling out of the target foil were stopped in a recoil chamber filled with He-gas, transported by a He gas-jet seeded with KCl aerosols and collected on glass-microfiber filters. The aerosols with the radionuclides were then dissolved and used for the liquid–liquid extraction experiments. Thulium, prepared by irradiation of holmium, was used as a simulant of trivalent ions while cadmium, prepared by irradiation of palladium, was used as a divalent ion. HDEHP in kerosen was used as the extractant. The extractions were carried out from the medium of nitric acid. The data obtained confirmed that HDEHP is a suitable candidate for the separation of carrier-free di- and trivalent ions of cyclotron-produced short-lived radionuclides in acidic solution. An extensive search of methods already used in practice for influencing the oxidation states of (radio)nuclides at low concentrations was performed and completion and commissioning of an electrochemical equipment was carried out. The basic components of this electrochemical apparatus are the potentiostat, electrochemical cells, and a large spectrum of electrodes. The first experiments were aimed at verification of the correct functionality of all present components, including the correctness of the calibrations.

This work was supported by the Centre for advanced applied science, project No. CZ.02.1.01/0.0/0.0/16019/0000778, and the “JINR 3+3” project, both supported by the Ministry of Education, Youth and Sports of the Czech Republic.

AUTHOR INDEX

11. Errata & corrigenda

Revision I - Added missing abstracts:

ID: 997 GALLUCCIO, Francesco	*Combined radiochemical separation and multicollector ICP-MS approach to determine* ^{135}*Cs and* ^{135}*Cs/*^{137}*Cs isotopic ratio*
ID: 1004 DENKOVA, Antonia	*Fast, efficient and simple method to radiolabel polymeric micelles with radionuclides*
ID: 1010 RAJEC, Pavol	*Production of cyclotron-based Gallium-68 radioisotope and related radiopharmaceuticals*

We wholeheartedly apologise to the affected authors and contributors.

REMARKS AND NOTES

REMARKS AND NOTES

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On behalf of the local organizing committee

Dr. Marketa Hola, Masaryk University
Chair of Local Organizing Committee

Prof. Viktor Kanicky, Masaryk University
Conference Chairmen

Prof. Pavel Matejka, University of Chemistry and Technology Prague
Conference Chairmen