MS 1: Accelerator Mass Spectrometry and Applications

Time: Monday 10:30–12:30 Location: U A-Esch 2

Invited Talk MS 1.1 Mon 10:30 U A-Esch 2

Shedding light on Isobars — •Robin Golser — University of Vienna, Faculty of Physics, Isotope Physics

Accelerator Mass Spectrometry (AMS) is an extremely sensitive method to measure minute amounts of radionuclides. An electrostatic accelerator of the Tandem type is utilized to break-up molecules and to provide sufficient energy to identify atoms by their nuclear charge. Abundance sensitivities can routinely be measured as low as 1E-15 if no isobar interferes. Isobars, i.e. atomic or molecular ions with almost the same mass as the ion of interest, are /the/ challenge in (A)MS. Exploiting electronic properties of the isobaric anions at sub-eV kinetic energies is becoming a breakthrough for isobar suppression. Key of a new method implemented at the Vienna Environmental Research Accelerator (VERA) is the (non-resonant) laser photo-detachment of the unwanted isobars in a linear, gas-filled radio-frequency quadrupole. The negative ions of interest remain unaffected by the laser light, when their electron affinity is greater than the photon energy. Although the technique has proven robust and reliable in all cases tried so far, we observe some unexpected phenomena. For example, certain molecules get significantly suppressed without laser, and a strong "chemical reaction" with oxygen occurs when, e.g. Hf-fluoride anions are used in order to measure Hf-182. We envision both new (A)MS isotopes and new (A)MS applications.

MS 1.2 Mon 11:00 U A-Esch 2

Cs isotope measurements by means of ILIAMS — •Johannes Lachner[1], Alexander Wieser[1], Dag Hanstorp[2], Maki Honda[1], Haimei Liang[2], Di Lu[2], Oscar Marchhart[1], Martin Martschini[1], Alfred Priller[1], Peter Steier[1], Julia Sundberg[2], Jakob Welander[2], and Robin Golser[1] — [1]Isotope Physics, Faculty of Physics, University of Vienna, Austria — [2]Department of Physics, University of Gothenburg, Sweden

The sensitive analysis of the trace isotopes ^{135,137}Cs in natural samples, which is of interest for environmental and radio-ecological studies, is one of the aims in the development of Ion Laser InterAction Mass Spectrometry (ILIAMS) at the Vienna Environmental Research Accelerator (VERA). Following the extraction of CsF_2^- and BaF_2^- from the sputter ion source, a suppression of the ^{135,137}Ba isobars can be accomplished using a combination of 1) collisional detachment in a He buffer gas filled radiofrequency quadrupole, 2) chemical reactions by admixing a small fraction of O_2 to the He buffer gas, and 3) photodetachment with 2.3 eV photons (λ=532 nm).

Experiments at the Göteborg University Negative Ion Laser LAboratory (GUNILLA) suggest that increasing the photon energy from 2.3 eV to 3.8 eV or 5.0 eV is not applicable since both CsF_2^- and BaF_2^- are detached at higher energies. On the other hand, the experiments revealed that the interfering isobar BaF_2^- is easier detached in collisions than CsF_2^-. First ILIAMS assisted AMS measurements at VERA resulted in the successful detection of ^{135}Cs and ^{137}Cs using in-house reference materials.

MS 1.3 Mon 11:15 U A-Esch 2

AMS of ^{90}Sr with ILIAMS — •Martin Martschini[1], Oscar Marchhart[1], Maki Honda[1], Dag Hanstorp[2], Johannes Lachner[1], Haimei Liang[2], Alfred Priller[1], Peter Steier[1], Alexander Wieser[1], and Robin Golser[1] — [1]Isotope Physics, University of Vienna - Faculty of Physics, Austria — [2]Department of Physics, University of Gothenburg, Sweden

The fission product ^{90}Sr ($T_{1/2}$ = 28.64 a) is of high environmental interest both for its radiotoxicity as well as its potential as a tracer. Present ^{90}Sr detection limits of mass spectrometric techniques like RIMS, ICP-MS or AMS are similar to the radiometric limit of 3 mBq.

The new Ion Laser InterAction Mass Spectrometry (ILIAMS) technique at the Vienna Environmental Research Accelerator (VERA) overcomes the limiting isobar problem in AMS. It provides near-complete suppression of isobar contaminants in negative ion beams via selective laser photodetachment in a gas-filled radio frequency quadrupole (RFQ). With 12 W of laser power from a 532 nm cw-laser and He-O_2 mixtures as buffer gas, ILIAMS provides a suppression of $^{90}ZrF_3^-$ and $^{89}YF_3^-$ vs $^{88}SrF_3^-$ of $>10^7$. Extraction of SrF_3^- from the ion source and elemental separation in the ionization chamber provide additional suppression of Zr. First measurements on a dilution series from IAEA-TEL-2016-03 reference solution and Zr- and Y-spiked targets were successfully conducted. One of the remaining challenges is the low sputter output from SrF_2 attributed to ionizer poisoning by Sr. In this contribution, we will detail these measurements including efficiency and the present sensitivity limit.

MS 1.4 Mon 11:30 U A-Esch 2

Tracing Nuclear Weapons Fallout Actinides and ^{99}Tc by AMS — •Karin Hain[1], Fadime Gülce[1], Michael Kern[1], Gunther Korschinek[2], Johannes Lachner[1], Martin Martschini[1], Peter Steier[1], Jan Welch[3], Masatoshi Yamada[4], and Robin Golser[1] — [1]Universität Wien, Austria — [2]Technische Universität München, Germany — [3]Technische Universität Wien, Austria — [4]Hirosaki University, Japan

Long-lived radionuclides emitted into the environment by nuclear weapons tests can be used to study their migration behaviour under different physio-chemical conditions. In order to allow a direct comparison, at VERA (Vienna Environmental Research Accelerator), we aim to analyse as many radionuclides as possible from the same sample. This requires a high overall detection efficiency including chemical sample preparation. The current procedure, which allows us to analyze ^{233,236}U, ^{239,240}Pu, ^{237}Np and ^{241}Am from the same 10 L ocean water sample, and depth profiles from the Pacific Ocean analysed accordingly will be discussed. In this context, experiments on the negative ion yield of Uranium from the ion source are being conducted from which the latest results will be presented. We recently started a project on the detection of nuclear weapons fallout ^{99}Tc in different environmental reservoirs. A chemical purification procedure adjustable to the respective sample matrix is currently being developed using a ^{99m}Tc tracer. Preliminary results on the chemical recovery of different approaches and test measurements at the Maier-Leibnitz-Laboratory will be discussed.

MS 1.5 Mon 11:45 U A-Esch 2

Long-term Behavior of Actinides from Global Fallout and In-situ Tracer Tests — •Francesca Quinto[1], Horst Geckeis[1], Karin Hain[2], Markus Plaschke[1], Thorsten Schäfer[1], and Peter Steier[2] — [1]Institute for Nuclear Waste Disposal, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Isotope Research and Nuclear Physics, University of Vienna, Vienna, Austria

In order to evaluate the safety of nuclear waste disposal in deep geological formations, the long-term behavior of the actinides (An) in the environment must be studied. This is possible, e.g., with the investigation of chronologically defined nuclear contaminations like the global fallout and in situ radionuclide tracer tests. At the Grimsel Test Site (GTS) located in the granitic rock of the Aar Massif in Switzerland, we have analyzed the global fallout derived U-236, Np-237 and Pu-239 and the tracers U-233, Np-237, Pu-242, Pu-244, Am-241 and Am-243 employed in several in situ tests. Thanks to the sensitivity of accelerator mass spectrometry, we have determined An concentrations ranging from pg/g down to ag/g in groundwater samples sized 0.1 to 250 g. Such analytical capability has allowed gaining valuable experimental data on the retention and migration of An during more than a decade in the crystalline rock at the GTS.

MS 1.6 Mon 12:00 U A-Esch 2

Development of an analytical method for the determination of actinides in clay systems at ultra-trace levels with accelerator mass spectrometry (AMS) — •D. Glückman[1], F. Quinto[1], K. Hain[2], C. Joseph[1], V. Montoya[1], P. Steier[2], and H. Geckeis[1] — [1]Institute for Nuclear Waste Disposal, Karlsruhe Institute of Technology, Karlsruhe, Germany — [2]Isotope Physics, Faculty of Physics, University of Vienna, Vienna, Austria

A potential host rock for the final disposal of high-level nuclear waste (HLW) is clay rock. In the case of the release of radionuclides from the repository, diffusion represents their main transport process in this formation. Under reducing conditions, which are expected in the repository, actinides are stabilized in low valence states (+III, +IV) normally resulting in low solubility and strong sorption in clay minerals which makes it difficult to study diffusion processes. For this reason, an analytical procedure capable of determining actinides at ultra-trace levels (fg/sample) was developed. For detection, accelerator mass spec-

trometry (AMS) was applied. The analytical procedure comprised the preparation of Opalinus Clay and Callovo-Oxfordian Clay / pore water samples spiked with U-233, Np-237, Pu-244, Am-243 and Cm-248, group extraction of these nuclides and their measurement with AMS. The analysis of these actinide nuclides at concentrations down to 1E+6 atoms/sample (ca. 0.4 fg/sample) from the same sample has been demonstrated by AMS. Such analytical capability will allow for the investigation of actinide diffusion under reducing conditions which will contribute to the safety assessment of HLW repositories.

MS 1.7 Mon 12:15 U A-Esch 2

Test of Projectile X-ray AMS for Nuclear Waste Management — •Markus Schiffer, Lena Bussmann, Claus Müller-Gatermann, Richard Spanier, Susan Herb, Alexander Stolz, Stefan Heinze, Gereon Hackenberg, and Alfred Dewald — University of Cologne, Institute of Nuclear Physics, Germany

Projectile X-ray AMS (PXAMS) is a well-known method for isobar separation by the measurement of characteristic X-rays. The discrimination of AMS nuclide and isobar suffers from low X-ray production yields and detection efficiencies, compared to the detection with particle detectors. New commercial Fast Silicon Drift Detectors (FSDD) with large active area of 50 mm^2 with high energy resolution, ΔE=123 eV at 5.9 keV, allow nowadays a high detection efficiency.

We investigate the projectile X-ray production yield at energies of 2 MeV/amu for medium mass isotopes on heavy stopper materials, like Au or U, to test its suitability as an alternative detection technique to liquid scintillator counting for the determination of isotopic concentrations of e.g. ^{90}Sr, which plays an important role for nuclear waste management.

In this kind of reaction we observed shifts in X-ray energies which are explained in literature by molecular-like electron states for heavy ion collisions. This may became important for the background by K_β, L_β and M_β lines.