

MS 4: Accelerator Mass Spectrometry

Time: Wednesday 10:30–12:30

Location: MS-H9

Invited Talk

MS 4.1 Wed 10:30 MS-H9

Isobar separation with cooled ions and laser light for compact AMS facilities — ●JOHANNES LACHNER^{1,2}, STEFAN FINDEISEN¹, ROBIN GOLSER², MICHAEL KERN², OSCAR MARCHHART², MARTIN MARTSCHINI², ANTON WALLNER¹, and ALEXANDER WIESER² — ¹HZDR, Dresden — ²University of Vienna, Faculty of Physics, Austria

Ion-Laser InterAction Mass Spectrometry (ILIAMS) slows down anions to thermal kinetic energies in a radiofrequency quadrupole (RFQ) filled with He buffer gas. Laser light (e.g. 532 nm) is overlapped with the decelerated anions to separate isobars via photodetachment.

Here, we present two applications of ILIAMS at the 3 MV Vienna Environmental Research Accelerator (VERA): ²⁶Al is an established AMS nuclide but its detection can be improved using AlO⁻, which is formed more likely than the customarily applied Al⁻. ILIAMS suppresses the isobar ²⁶Mg by neutralization of MgO⁻ and overcomes the disadvantage of AlO⁻ compared to Al⁻, where Mg⁻ is not extracted from the ion source. This enhances the sensitivity of ²⁶Al detection and the prolific AlO⁻ beam can be used at facilities with terminal voltages < 10 MV. ^{135,137}Cs measurements are presented as an example of highly sensitive detection of novel AMS nuclides. In this case, we use ^{135,137}CsF₂⁻ anions and ILIAMS suppresses the isobaric ^{135,137}BaF₂⁻.

We furthermore present a new design of a modular ion cooler with multiple RFQ sections. With more control of the ion energy during their passage through the RFQ we want to improve the transport efficiency for molecular anions. This ion cooler will be integrated in a new 1 MV AMS facility at Dresden in 2023.

MS 4.2 Wed 11:00 MS-H9

Why and how producing an isotopic spike for the analysis of environmental ²³⁷Np — ●KARIN HAIN¹, MARTIN MARTSCHINI¹, AYA SAKAGUCHI², PETER STEIER¹, ANDREAS WIEDERIN¹, AKAHIKO YOKOYAMA³, and ROBIN GOLSER¹ — ¹University of Vienna, Faculty of Physics, Austria — ²University of Tsukuba, Faculty of Pure and Applied Science, Japan — ³Kanazawa University, Institute of Science and Engineering, Japan

The quantitative analysis of the potential oceanographic tracer ²³⁷Np by mass spectrometric techniques such as Accelerator Mass Spectrometry (AMS) still suffers from the lack of an isotopic spike. Measurements using non-isotopic spikes for normalisation, such as ²⁴²Pu, have achieved acceptable results for selected environmental materials, but require careful control of the oxidation states during chemical separation and monitoring of the relative ion source output. In our joint project with the University of Tsukuba we successfully produced mass 236 by the irradiation of Th foils with a ⁷Li beam in the 30-40 MeV range at the RIKEN Nishina Center. First AMS measurements using fluoride molecules indicate that the observed surplus of mass 236 above background is indeed ²³⁶Np. The by-production of mass 237, however, is higher than expected from model calculations with the EMPIRE code and needs further investigation. This contribution will discuss environmental levels of ²³⁷Np obtained with AMS using non-isotopic normalisation and the present status of the ²³⁶Np spike production.

MS 4.3 Wed 11:15 MS-H9

Separation of U and Np Isobars by ILIAMS — ●ANDREAS WIEDERIN¹, KARIN HAIN¹, MARTIN MARTSCHINI¹, AYA SAKAGUCHI², PETER STEIER¹, and ROBIN GOLSER¹ — ¹University of Vienna, Faculty of Physics - Isotope Physics, Austria — ²University of Tsukuba, Faculty of Pure and Applied Science, Japan

²³⁷Np (T_{1/2}=2.1 Ma) is the second most abundant anthropogenic actinide in the environment, but a reliable quantification by AMS independent of the environmental matrix would require an isotopic spike for normalization. We currently consider ²³⁶Np the most suitable candidate to serve this purpose. Such a spike material is currently under development and needs a careful characterization regarding interfering by-products. Possible isobaric background from ²³⁶U necessitates the separation of U and Np by chemical means and/or during the AMS measurement. Ion Laser InterAction Mass Spectrometry (ILIAMS) uses selective laser photodetachment inside an RFQ ion cooler to neutralize anions of the interfering isobar. By applying a 2.33 eV laser, UF₄⁻ has been suppressed by at least four orders of magnitude in recent experiments, while NpF₄⁻ passed unaffected. The comparatively low formation rate and high sensitivity to interactions with the buffer

gas of UF₄⁻ further increased the suppression relative to NpF₄⁻. This result represents the first isobar separation in the mass range of the actinides in AMS. The U suppression achieved is already sufficient for the application of ²³⁶Np to quantitatively determine ²³⁷Np by isotope ratio measurements.

MS 4.4 Wed 11:30 MS-H9

Study of Actinide Signatures as Potential Markers for the Anthropocene — ●JANIS WOLFF¹, KARIN HAIN¹, MARIA MESZAR², MICHAEL STRASSER³, MICHAEL WAGREICH², and ROBIN GOLSER¹ — ¹University of Vienna, Faculty of Physics, Währinger Str. 17, 1090 Vienna, Austria — ²University of Vienna, Department of Geology, Althanstraße 14, 1090 Vienna, Austria — ³University of Innsbruck, Department of Geology, Innrain 52f, 6020 Innsbruck, Austria

The Anthropocene is the proposed geological epoch that follows the Holocene and is defined by the predominance of human impact on the Earth System. An epoch-defining impact must produce stratigraphic signals that are unique, distributed globally, and well preserved for a long time. Long-lived radionuclides released in atmospheric nuclear weapon testings may have produced a suitable signal. The proposed markers for the Anthropocene, Pu-239 and Am-241, and additionally U-233, U-236, Np-237 and Pu-241 were analyzed in the different reservoirs, urban strata and Austrian lake sediments, using the AMS facility VERA. The anthropogenic radionuclides have been successfully detected in layers corresponding to the active phase of nuclear weapons testing. In the urban strata, the isotopic ratio U-233/U-236, a new signature for nuclear weapons fallout, marks the onset of the Anthropocene whereas the concentrations of the other radionuclides in general gradually increase towards younger ages.

MS 4.5 Wed 11:45 MS-H9

Towards the Redetermination of the Half-life of ³²Si - Isobar Separation of ³²Si from the Isobar ³²S — ●MATTHIAS SCHLOMBERG, DR. CHRISTOF VOCKENHUBER, and PROF. DR. HANS-ARNO SYNAL — Laboratory of Ion Beam Physics, ETH Zürich

The ³²Si is a cosmogenic, long-lived radionuclide with potentially interesting applications for dating the recent past. However, its half-life of about 150 years is still not known with sufficient precision despite several independent measurements over the past four decades. The SINCHRON collaboration with partners from PSI, CHUV, PTB and ETH aims at a comprehensive redetermination of the half-life of ³²Si. The Laboratory of Ion Beam Physics (LIP) at ETH Zurich will perform the AMS measurements using the 6 MV-Tandem facility for the determination of the number of ³²Si atoms in the samples used for the activity measurement. In addition to the challenge of performing an absolute measurement without having standards available, ³²Si must be separated from its intense isobar ³²S.

We developed a method based on a passive gas absorber in front of a gas ionization detector that allows us detection of ³²Si by stopping the isobar ³²S at 30 MeV. However, background from light recoils from the absorber material and deviations of the stopping power at low energies still pose challenges.

An overview of the SINCHRON project will be presented. The setup and the obtained data will be discussed with respect to an absolute measurement.

MS 4.6 Wed 12:00 MS-H9

Improved ⁴¹Ca AMS measurements at DREAMS — ●CARLOS VIVO-VILCHES, GEORG RUGEL, JOHANNES LACHNER, ANTON WALLNER, DOMINIK KOLL, KONSTANZE STUEBNER, SEBASTIAN FICHTER, and STEPHAN WINKLER — Helmholtz-Zentrum Dresden-Rossendorf, AcceleratorMass Spectrometry and Isotope Research, Dresden, Germany

Sensitivity of ⁴¹Ca measurements at the 6 MV AMS system at HZDR, DREAMS, using calcium fluoride (CaF₂) targets, is mainly limited by 2 factors: the total efficiency of the measurements; and the fraction of ions of its isobar ⁴¹K which mimic the signal of ⁴¹Ca in the gas ionization chamber detector.

The addition of lead fluoride (PbF₂) to the target mixture has been proven to boost the production of different (MF_n)⁻ ions. At DREAMS, changing the previously used mixture of CaF₂+Ag (1:4 w/w) by CaF₂+Ag+PbF₂ (1:4:4 w/w), ionization efficiency is increased from

$\sim 0.15\%$ to $\sim 0.45\%$.

The ^{41}K suppression by the detector can also be improved, even without changes in the instrumentation itself. With an optimized analysis of the 4-dimensional signals from the gas ionization chamber detector, the suppression factor can be increased, at least, a factor 2: from 2×10^4 to 4×10^4 .

The reported changes improve the total efficiency of ^{41}Ca detection as well as the suppression of the ^{41}K isobar and lead to a $^{41}\text{Ca}/^{40}\text{Ca}$ sensitivity of $2\text{-}3 \times 10^{-15}$ with an overall efficiency of $\sim 0.03\%$.

MS 4.7 Wed 12:15 MS-H9

Normalization methods for the analysis of environmental

^{99}Tc — ●STEPHANIE ADLER, KARIN HAIN, MARTIN MARTSCHINI, FADIME GÜLCE, and ROBIN GOLSER — University of Vienna, Faculty of Physics

Quantification of the anthropogenic radionuclide ^{99}Tc ($t_{1/2} = 2.1 \cdot 10^5$

yr) in the general environment by AMS requires suppression of the stable isobaric background of ^{99}Ru and a reliable normalization method to overcome the lack of a stable Tc isotope.

At VERA, previous research has shown that extracting TcF_5^- from the ion source is suitable for Ion Laser InterAction Mass Spectrometry (ILIAMS) as RuF_5^- can be suppressed by a laser by up to 10^5 .

Experiments at other AMS-facilities using TcO^- normalized the ^{99}Tc to the ^{93}Nb -current of the matrix material with a precision of 30%. Using this approach, our experiments showed Tc-deficits of $> 40\%$, indicating major loss of Tc during sample preparation. This led to thorough investigations of the final target preparation steps utilizing ^{95}Tc and γ -Spectrometry. The chemical recovery was improved to reliable yields $> 92\%$, by lowering the calcination temperature.

When extracting $^{99}\text{TcF}_5^-$ from the source as required for ILIAMS, the Nb-normalization scatters by a factor of 10 and seems less reliable. Thus research on using an alternative spike material ^{103}Rh for normalization is currently ongoing.