MS 3: Actinide Analysis

Time: Tuesday 11:00–13:00 Location: N 6

Invited Talk MS 3.1 Tue 11:00 N 6

Flying viruses - Native mass spectrometry meets X-rays — •Charlotte Uetrecht — CSSB Centre for Structural Systems Biology, Deutsches Elektronen-Synchrotron DESY & Leibniz Institute of Virology (LIV) & University of Lübeck, Notkestraße 85, 22607 Hamburg, Germany — Institute of Chemistry and Metabolomics, University of Lübeck, Ratzeburger Allee 160, 23562 Lübeck, Germany

Native mass spectrometry (MS) enables ionisation and transfer of structurally intact non-covalent protein complexes into the gas-phase. As such, it is a perfect tool to study proteins and their assembly intermediates in a mass and conformation specific manner, albeit with limited structural resolution. Accordingly, other experimental approaches such as X-ray diffractive imaging are necessary to get a full understanding of proteins, their assemblies and dynamic processes. Therefore, it seems natural to combine native MS with X-ray diffraction in the gas phase. In particular, well established methods from MS like m/z selection, ion trapping or ion mobility are incorporated in the MS SPIDOC sample delivery system. In contrast to conventional diffractive imaging of crystallised proteins, the proteins here are delivered as single particles without the need for crystallisation. This increases naturally the requirement on the X-ray source: high-resolution single-particle X-ray diffractive imaging (SPI) can only be conducted at X-ray free electron lasers, lower resolution information can be obtained from small angle X-ray scattering (SAXS) at synchrotrons. This talk highlights first results from leveraging such synergies, and how this will improve our understanding of virus structure and dynamics.

Invited Talk MS 3.2 Tue 11:30 N 6

Precision mass measurements of exotic fission fragments — •Arthur Jaries for the JYFLTRAP and ISOLTRAP-Collaboration — Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Direct measurement of the mass of single nuclides allows to determine their binding energy and gives information on the evolution of nuclear structure away from stability. Among the available experimental methods, Penning-trap mass spectrometry provides the most precise technique to probe this fundamental nuclear property, typically reaching a relative uncertainty of one part per billion. Additionally relevant for astrophysics, precision mass measurements produce valuable inputs for the theoretical models describing the stellar nucleosynthesis processes such as the rapid neutron capture (r) process. The latter is responsible for the production of more than half of the elemental abundances above iron, making mass spectrometry of radionuclides involved a tool to better understand the origin of heavy elements in the Universe. In the recent years at the JYFL Accelerator Laboratory in Finland, the use of the JYFLTRAP Penning traps combined with the fast and universal IGISOL production method, led to the determination of more than 100 atomic masses across the nuclide chart, with a specific focus on the rare-earth and the ^{132}Sn neutron-rich regions. This presentation highlights the latest measurement campaigns of exotic fission fragments of uranium performed with the JYFLTRAP, discussing their implications for nuclear structure and r-process modeling.

MS 3.3 Tue 12:00 N 6

Age dating of thermonuclear bomb fragments by combination of rL-SNMS and gamma spectroscopy — •Paul Hanemann[1], Jan Schimansky[1], Tobias Weissenborn[1], Aaron Lehnert[1], Manuel Raiwa[1], Jixin Qiao[2], Sven Nielsen[2], and Clemens Walther[1] — [1]Leibniz Universität Hannover, IRS, Germany — [2]Technical University of Denmark, Roskilde, Denmark

In 1968 a US-bomber armed with thermonuclear warheads crashed close to the Thule airbase, releasing plutonium fragments into the environment. [1] Using the ^{241}Pu ^{241}Am chronometer it is possible to determine the production date of the Pu. For the determination of the atomic ratios of both isotopes in the past a complex and destructive radiochemical preparation of the sample and two measurements over the span of multiple years were necessary. [2] By utilizing the nondestructive isobar free measurements of resonant Laser Secondary Neutral Mass Spectrometry (rL-SNMS) and traditional gamma spectrometry we are able to age date multiple fragments of nuclear material from the Thule region in a comparatively fast way while keeping them intact for further analysis. The date of the chemical purification of the Pu in the samples was determined to be around the year 1960. This result agrees well with the existing values from the literature by Eriksson et al. [2]. Compared to [2], due to passing time nearly $\frac{3}{4}$ of the ^{241}Pu has decayed until our measurements, yet our results have comparable uncertainties. This high sensitivity shows a clear advantage of using this rL-SNMS based method for age dating. References: 1: DOI:10.1016/S1569-4860(91)80004-8; 2: DOI:10.1021/es800203f

MS 3.4 Tue 12:15 N 6

Characterizing a cryogenic ion mobility spectrometer for actinides with measurements on their lighter homologues — •Felix Katzer[1], Michael Block[1,2,3], Premaditya Chhetri[1,2,3], Biswajit Jana[4], Mustapha Laatiaoui[5], Sebastian Raeder[2,3], and Elisabeth Rickert[1,2] — [1]JGU Mainz, Germany — [2]HI Mainz, Germany — [3]GSI Darmstadt, Germany — [4]BARC Trombay, India — [5]GANIL Caen, France

The investigation of the ion mobility of heavy elements in noble gases provides valuable insights into the influence of relativistic effects and electron correlation on their electronic structure. To enable such studies, a cryogenic drift tube based actinide ion mobility spectrometer (AIMS) has been developed as a powerful tool to better understand the chemical and physical properties of heavy elements. To characterize the set up, various transition metals and lanthanides, such as Gd^+, Ho^+, Tm^+ and Yb^+, were studied with regard to their low-field reduced ion mobility in helium as a buffer gas. In the measurements, the mobility behavior of the homologous f-block elements was investigated by varying measurement parameters in order to understand the underlying mechanisms and to determine system uncertainties. Further modifications to the system are being carried out in preparation for future mobility measurements of the elements in the actinide series.

MS 3.5 Tue 12:30 N 6

Assessment of the behaviour and source of anthropogenic actinides on HZDR's research campus — •Shruti Dabke[1], Sebastian Fichter[1], Dominik Koll[1], Michael Hotchkis[2], and Anton Wallner[1] — [1]Helmholtz-Zentrum Dresden Rossendorf (HZDR), Dresden, Germany — [2]Australian Nuclear Science and Technology Organisation (ANSTO), Lucas Heights, Australia

Understanding actinide behaviour in environmental reservoirs is essential for assessing safety and potential plant or groundwater uptake. To study the accessibility of actinides by different chemical conditions, a sequential leaching technique was employed. Hence, the fractionation of actinides, specifically U, Pu and Am, in one soil sample collected from the campus of HZDR was analysed for five established fractions and an additional refractory fraction accessed by HF digestion. The concentrations of the actinide isotopes were measured using ICP-MS and AMS. Preliminary Pu data showed a non-negligible percentage of the total plutonium concentration in the refractory fraction. Acid leaching, which is commonly used for such environmental analyses, would not typically give access to this refractory plutonium. Moreover, the ratio of ^{240}Pu/^{239}Pu in the refractory fraction was determined to be 0.047 ± 0.002 which is significantly lower than the global fallout ratio found in the other fractions. This suggests a source of refractory weapons-grade plutonium. Further experiments on soil samples and the IAEA-385 reference sediment will focus on the refractory fraction, and the results of isotopic fractionation of the other actinide elements will be studied using the new AMS facility, HAMSTER, at HZDR.

MS 3.6 Tue 12:45 N 6

Traceable Reference Material for mass spectrometry: The EURAMET project Metrology for the harmonisation of measurements of environmental pollutants in Europe — •Aaron Lehnert[1], Lucille Chambon[2], Ben Russell[3], Dirk Arnold[4], and Clemens Walther[1] — [1]Leibniz Universität Hannover, Germany — [2]Université Paris-Saclay, CEA, List, Laboratoire National Henri Becquerel, France — [3]National Physical Laboratory, United Kingdom — [4]Physikalisch-Technische Bundesanstalt, Germany

The European Green Deal's ambition for zero pollution requires the development of highly sensitive techniques to detect ultra-low amounts of pollutants and to determine their isotope ratios, where mass spectrometry is a key method for determination of non-radioactive polluting elements and long-lived radionuclides. The MetroPOEM project

bridges the traceability gap between activity and mass-based measurements - particularly estimation of mass bias.

A solid silica-based reference material, produced by sol-gel synthesis at CEA spiked with 234,235,236,238U, 237Np, 239,240Pu and 241Am, as well as ~10 kg of an inactive material for testing dissolution procedures. Additionally, ČMI generated ~50*0,5L liquid RM aliquots of sea water spiked with natural U, 237Np, 239,240Pu and 241Am.

These materials were characterised in accordance with the requirements of ISO 17034 by interlaboratory studies between the project partners, using techniques developed in the project.