MS 9: Actinide Analysis

Time: Thursday 17:30–19:00 Location: HS 2 Chemie

Invited Talk MS 9.1 Thu 17:30 HS 2 Chemie

Development of chemical ionization methods based on plasma driven reactant ion production — •Thorsten Benter, Hendrik Kersten, and Walter Wissdorf — University of Wuppertal, Wuppertal, Germany

Earlier this year, the DPG/professional association mass spectrometry (MS), and the German Society for Mass Spectrometry (DGMS) have reinitiated a collaboration initiative with the goal of fostering current and future research efforts regarding MS, particularly at research institutions, and providing a larger platform for the communication of related results. One tool in this collaboration is the mutual invitation of keynote speeches at the annual conferences of both societies; this contribution is the first in a hopefully long line of future DPG/DGMS interactions.

This talk begins with a brief outline of the current research efforts at the institute for pure and applied mass spectrometry (ipaMS) and the Physical and Theoretical chemistry (PTC) at the university of Wuppertal. We will then discuss selected applications of low to medium pressure, low power plasmas in recently developed chemical ionization (CI) methods used in MS. Some of these methods are tailored towards operation in highly specialized environments, e.g., atmospheric chemistry or semiconductor production research, whereas others have been commercialized, e.g., in the ecTOF of the Swiss company TOFWerk. The latter approach is discussed in more depth, as electron and chemical ionization is (uniquely) running in parallel, providing correlated molecular (CI) and structural (EI) mass spectrometric data.

MS 9.2 Thu 18:00 HS 2 Chemie

Resonant Laser Ionisation Mass Spectrometry on hot particles from the Thule area — •Paul Hanemann[1], Tobias Weissenborn[1], Aaron Lehnert[1], Jixin Qiao[2], Sven Nilsen[2], and Clemens Walther[1] — [1]Leibniz University Hannover, IRS — [2]Technical University of Denmark

In resonant Laser-ionisation Secondary Neutral Mass Spectrometry (rL-SNMS) a set of lasers is used to selectively ionize atoms of a specific element for ToF-MS analysis. This method combines sub-micron spatial resolution with excellent elemental selectivity while keeping the sample structurally intact. In previous work [1-3], rL-SNMS was used for the characterisation of individual nuclear fuel fragments from the Chornobyl Exclusion Zone. To prove the versatility of rL-SNMS we applied this method to samples from the Thule area in Greenland, where a bomber carrying four thermonuclear bombs crashed in 1968. Individual hot particles were isolated from soil samples. In traditional SIMS analysis of Thule hot particles by Ranebo et al. [4] isobaric interferences posed a significant challenge. In this work isobaric interference free rL-SNMS measurements were performed successfully on multiple particles. The particles contain both weapons grade plutonium ($240Pu/239Pu > 0.05$) as well as highly enriched uranium ($235U/238U > 1.0$). The adaption of rL-SNMS from fragments of nuclear fuel to hot particles from nuclear weapon material shows the versatility of this method and its applications for nuclear forensics. References: 1: DOI: 10.1126/sciadv.abj1175 2: DOI: 10.1016/j.sab.2022.106377 3: DOI: 10.1016/j.jhazmat.2023.131338 4: DOI: 10.1017/S1431927607070353

MS 9.3 Thu 18:15 HS 2 Chemie

Nuclear Forensic Analysis of a single Chornobyl Hot Particl via Advanced Mass Spectrometry Techniques — •Laura Leifermann[1], Greg Balco[2], Autumn Roberts[2], Paul Hanemann[1], Tobias Weissenborn[1], Wolfgang Schulz[1], Manuel Raiwa[2], Martina Klinkenberg[3], Felix Brandt[3], Michael Savina[2], Brett Isselhardt[2], and Clemens Walther[1] — [1]Leibniz University Hannover, Hannover, Germany — [2]LLNL, Livermore, USA — [3]Forschungszentrum Jülich, Jülich, Germany

In the field of nuclear forensics, clarifying the origin and age of an unknown sample is the central question and common practice. Nuclear fuel samples rise to a number of new questions regarding burn-up, degree of enrichment, and operational conditions during the service life. The resonant-laser-SNMS method is ideally suited for this purpose. Actinides, fission and breeding products can be analyzed isotope and element selective. If this is combined with noble-gas-MS, many insights can be gained about a fuel sample. This provides a complete picture of the age, burn-up, neutron flux densities during reactor operation and source of a sample. The ratio of U-235/U-238 of a special particle from Chornobyl is 1,22% which indicates a relative low burn-up and matches the Xe-136(131+132) isotope ratios. This is in contrast to the Xe-134(131+132) ratio which indicates a high Pu-fission. This work addresses a unique particle that has raised questions that can be solved by a synthesis of different mass spectrometric analysis techniques.

MS 9.4 Thu 18:30 HS 2 Chemie

Isotopic purification of trans-uranium tracers using RIMS at RISIKO and their characterization with AMS (I) — •Raphael Hasse[1], Sebastian Berndt[1], Christoph E. Duellmann[1,2,3], Sebastian Raeder[2], and Klaus Wendt[1] — [1]Johannes Gutenberg-Universität Mainz, Mainz, Germany — [2]GSI Helmholtzzentrum für Schwerionenforschung, Darmstadt, Germany — [3]Helmholtz-Institut Mainz, Mainz, Germany

Ultra-sensitive accelerator mass spectrometry is regularly used to quantify low levels of actinide concentrations as well as isotopic ratios in environmental samples. Actinide quantification relies on a relative measurement to an added mono-isotopic spike of the respective element. Standard solutions are available at the required purity (up to 10^{-7}) for elements such as U, Pu and Am. However, no corresponding commercial high-purity spike solution exists so far for the trans-uranium elements Np and Cm. In this project, we establish a novel isotopic purification and characterization of isotopic spikes using a combination of RIMS and AMS starting with ^{248}Cm and later extending it to ^{236}Np.

Here, we present this novel isotopic purification approach, carried out at the RISIKO laser mass separator facility in Mainz, using element and isotope selective resonance ionization by two tuneable Ti:Sa lasers. For suitable isotopic purification of a ^{248}Cm spike, an efficient two step ionization scheme was developed. In this way, an overall efficiency well above 10% with a suppression of the neighbouring isotope ^{247}Cm by about 300 was achieved.

MS 9.5 Thu 18:45 HS 2 Chemie

Isotopic purification of trans-uranium tracers using RIMS at RISIKO and their characterization with AMS (II) — •Dominik Koll[1,2], Sebastian Fichter[2], Michael Hotchkis[3], and Anton Wallner[1,2] — [1]TUD Dresden University of Technology, Dresden, Germany — [2]Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany — [3]Australian Nuclear Science and Technology Organisation, Lucas Heights, Australia

Ultra-sensitive accelerator mass spectrometry is regularly used to quantify low levels of actinide concentrations as well as isotopic ratios in environmental samples. Actinide quantification relies on a relative measurement to an added mono-isotopic spike of the respective element. Standard solutions are available at the required purity (up to 10^{-7}) for elements such as U, Pu and Am. However, no corresponding commercial high-purity spike solution exists so far for the trans-uranium elements Np and Cm. In this project, we establish a novel isotopic purification and characterization of isotopic spikes using a combination of RIMS and AMS starting with ^{248}Cm and later extending it to ^{236}Np.

Here, we present the characterization of an isotopically purified ^{248}Cm spike using AMS. A dilution series of purified ^{248}Cm with the initial ^{248}Cm solution containing certified amounts of Cm isotopes was measured for isotopic ratios to quantify the purified ^{248}Cm amount. High-sensitivity measurements of the purified spike were carried out for A=244,247 isotopes to assess the isotopic purity. A suppression factor of about 300 for the neighbouring isotope ^{247}Cm was achieved.