MS 3: Mass Spectrometry Applications

Time: Wednesday 14:30–16:00 Location: F128

Invited Talk MS 3.1 Wed 14:30 F128

Durable, low-temperature and highly-selective catalysis in NO reduction and CO oxidation driven by uni-sized Pt clusters supported on Si and SiC substrates — •Hisato Yasumatsu — Toyota Technological Institute, Ichikawa, Chiba, Japan

It has been found that Pt clusters are fixed to a Si substrate as Pt cluster disks by impact of Pt cluster ions onto the substrate. Electrons accumulated at the sub-nano interface between the Pt cluster and the Si substrate surface enable catalytic NO reduction and CO oxidation at low temperatures with high selectivity.

When the substrate is changed to silicon carbide, SiC, which is well known to possess high chemical and thermal stability, one can obtain durable catalysis as high as 1200 K with maintaining the low-temperature and highly-selective catalytic performance. This study shows a way to utilize this unique and high-performance catalysis driven by the electron accumulation as practical catalysts of electron donation including the gas treatment, and further extension to water-splitting hydrogen production and fuel-cell oxygen reduction reaction (ORR) as well.

Size-selected clusters by passing through a mass filter and cooled through a He collision cell were allowed to collide onto the substrate. Geometry and electron distribution were studied by means of scanning-tunneling microscopy. Electronic structures were obtained through X-ray photoelectron spectroscopy. Catalytic activity was measured with surface-chemistry techniques and fixed-bed gas-flow reaction analysis in combination of mass spectroscopy.

MS 3.2 Wed 15:00 F128

Multi-element isotopic analysis of hot particles from Chornobyl — •Darcy van Eerten[1], Manuel Raiwa[1], Paul Hanemann[1], Laura Leifermann[1], Tobias Weissenborn[1], Wolfgang Schulz[1], Martin Weiss[1], Danielle Ziva Shulaker[2], Peter Boone[2], David Willingham[2], Keenan Thomas[2], Brian Sammis[2], Brett Isselhardt[2], Mike Savina[2], and Clemens Walther[1] — [1]Institut für Radioökologie und Strahlenschutz, Leibniz Universität Hannover, Herrenhäuser Str. 2, 30419 Hannover, Germany. — [2]Chemical and Isotopic Signatures Group, Lawrence Livermore National Laboratory, Livermore, USA.

Nuclear materials that contaminate the environment present an ongoing challenge to characterize due to their small size and diverse morphology. The analysis of isotope ratios in actinides and fission products can provide determination of origin, age and environmental weathering of these materials. Resonance ionisation mass spectrometry (RIMS) utilizes selective laser ionization to target single elements and suppress the isobaric interferences typically found in mass spectrometry. Two specialized instruments were used to analyse single hot particles from Chornobyl: rL-SNMS at the IRS in Hannover, Germany, and LION at LLNL in Livermore, USA. Results from multiple particles are presented with interpretations of isotope ratios in U, Pu, Cs, Rb, Sr and Ba.

MS 3.3 Wed 15:15 F128

Mass spectrometric determination of the speciation of radium in the human digestive tract using ESI-MS — •Linus Holtmann, Ahmadabdurahman Shamoun, Beate Riebe, and Clemens Walther — Institut für Radioökologie und Strahlenschutz, Leibniz Universität Hannover, Hannover, Germany

Radionuclides pose a potential radio- and chemotoxic hazard to humans when ingested. Knowledge of radionuclide interaction in the digestive tract at the molecular and cellular level is necessary for risk assessment and to contribute to an element-specific decontamination strategy.

Synthetic biofluids prepared according to the UBM protocol (BARGE) are used to investigate the speciation of radium in the human digestive tract. The biofluids are analyzed in the presence and absence of Ra(II)/Ba(II) by mass spectrometry using electrospray ionization (ESI-MS). In our experiments, an Orbitrap mass spectrometer allows the measurement of the speciation without any chemical separation. The identification of barium-containing species takes place via specific isotope pattern signatures. Algorithms specifically tailored to the evaluation of complex mass spectra are used.

In addition, the influence of different decorporation and complexing agents on the speciation of Ra(II)/Ba(II) in the simulated digestive process is studied. This way, element-specific decorporation strategies are investigated for their potential efficacy after oral ingestion of radium.

MS 3.4 Wed 15:30 F128

ALPINAC - A non-target screening algorithm for high-resolution mass spectra and its application to the detection of halogenated greenhouse gases. — •Katharina Höveler, Lionel Constantin, Myriam Guillevic, Paul Schlauri, Martin K. Vollmer, and Stefan Reimann — Swiss Federal Laboratories for Materials Science and Technology (Empa), Dübendorf, Schweiz

Efficient and automated screening of gaseous or liquid samples to detect novel compounds based on their mass spectral fingerprints (non-target screening) is an ongoing computational challenge that goes beyond standard library-based approaches. We present a novel algorithm that uses combinatorial and directed graph methods, taking into account chemical rules, to automatically assign high-resolution mass spectral peaks from gas-chromatography-separated time-of-flight mass-spectroscopy (GC-TOF MS) measurements to possible chemical formulas by considering possible fragmentation pathways. In a further step, this information is used to reconstruct the chemical formula of likely molecular parent ions. We show how this technique can be used to detect unknown contaminants in pre-concentrated air samples and how the algorithm can be extended to reconstruct not only the molecular formula but also the chemical structure of the parent ion.

MS 3.5 Wed 15:45 F128

Analysis of the cutting edge of individual 'hot particles' from the Chernobyl Exclusion Zone — •Laura Leifermann[1], Martina Klinkenberg[2], Felix Brandt[2], Paul Hanemann[1], Tobias Weissenborn[1], Sandra Reinhard[1], Manuel Raiwa[3], Wolfgang Schulz[1], and Clemens Walther[1] — [1]IRS, Hannover, Deutschland — [2]FZJ IEK-6, Jülich, Deutschland — [3]LLNL, Livermore, USA

During the Chernobyl reactor accident on April 26, 1986, radioactivity was in part released in the form of nuclear fuel particles. These so-called 'hot particles' have various structures that belong to specific oxidation states of uranium. These oxidation states behave differently in the environment. We obtain individual particles by density separation with a poly tungsten solution. Via radiometric scanning with a Geiger counter we locate the particles. The extraction is performed on tungsten needles with a micromanipulator in a scanning electron microscope (SEM). The particle surface was analyzed by different non-destructive methods such as SIMS, rL-SNMS and EDX. Gamma measurements and optical analyses in SEM were also performed. Micrometer sized particles glued to needles are cut in half with a focused ion beam. We can thus extend our mass spectrometric analysis to the cutting edge and study the particle cross section. Since the particles have been exposed to the environment for over 30 years, weathering effects from outside to inside can be investigated. In addition, it is possible to test to what extent the elemental and isotopic composition of the particles is homogeneous.