Chemical Analysis and Imaging at Interfaces
Room 205 ABCD W - Session CA+AS+SS-WeM

Chemical Analysis and Imaging at Interfaces Oral Session

Moderators: Gabriel Parker, Oak Ridge National Laboratory, **Xiao-Ying Yu**, Oak Ridge National Laboratory, USA

8:00am **CA+AS+SS-WeM-1 Heterogeneous Chemistry at Liquid-Vapor Interfaces Investigated by X-Ray Photoelectron Spectroscopy*, Hendrik Bluhm,*** Fritz Haber Institute of the Max Planck Society, Germany **INVITED**

Aqueous solution-vapor interfaces govern important phenomena in the environment and atmosphere, including the uptake and release of trace gases by aerosols and CO_2 sequestration by the oceans. A detailed understanding of these processes requires the investigation of liquid-vapor interfaces with chemical sensitivity and interface specificity. [1] This talk will discuss opportunities and challenges for investigations of liquid-vapor interfaces using X-ray photoelectron spectroscopy and describe recent experiments that have focused on the propensity of certain ions and the role of surfactants at the liquid-vapor interface.[2-4] The talk will also discuss the utilization of photoelectron angular distributions for the investigation of the depth of solvation of surfactants at the interface. [5-7]

[1] R. Dupuy, et al, J. Chem. Phys. **154**, 060901 (2021).

[2] S. Gholami, et al., Environmental Science: Atmospheres **5**, 291-299 (2025).

[3] C. Richter, et al., Phys. Chem. Chem. Phys. **26**, 27292-27300 (2024).

[4] T. Buttersack, et al., Nat. Commun. **15**, 8987 (2024).

[5] R. Dupuy, et al., Phys. Chem. Chem. Phys. **24**, 4796-4808 (2022).

[6] R. Dupuy, et al., Acc. Chem. Res. **56**, 215-223 (2023).

[7] R. Dupuy, et al., Phys. Rev. Lett. **130**, 156901 (2023).

8:30am **CA+AS+SS-WeM-3 Probing Chemical and Catalytic Interfaces Using Operando Soft X-Ray Spectroscopy*, Jinghua Guo,*** Lawrence Berkeley National Laboratory **INVITED**

Synchrotron based X-ray spectroscopic techniques offers unique characterization of energy, catalysis and chemical process in regards to the functionality, complexity of material architecture and chemistry. In the operando soft X-ray spectroscopy study of interfacial phenomena, it has been found that the microstructure and composition of materials as well as the microstructure evolution process have a great influence on performances in a variety of fields, such as the energy conversion and energy storage materials in the chemical and catalytic processes. This presentation will show how to best use the X-ray spectroscopy characterization techniques, including X-ray absorption spectroscopy (XAS) and resonant inelastic X-ray scattering (RIXS) to investigate the real interfacial reaction mechanism during the operation. The experimental results show how operando soft X-ray spectra uncover the phase conversion, chemical and structure change of solid/liquid and solid/gas interfaces in real time, thus further enhance the understanding of real reaction mechanism.

9:00am **CA+AS+SS-WeM-5 Capturing Electrical Double Layer in Action with Xps on a Graphene Coplanar Capacitor with an Ionic Liquid*, Sefik Suzer,*** Bilkent University, Chemistry Department, Ankara, Turkey

Time-dependent XPS data is recorded for capturing the dynamics of the Electrical Double Layer formation on electrified two Multilayered-Graphene electrodes, configured as a coplanar-capacitor, having an ionic liquid as the electrolyte. The device is subjected to 2 V biasing cycle changing its polarity every hour, while iteratively recording the O1s peak representing the anion with 0.5 s steps. Variations in the O1s peak's binding energy position on the electrified electrode report directly the electrical potential of the IL medium, which suddenly jumps to the opposite polarization, if the electrode is grounded. The peak eventually returns to its equilibrium position with a relatively long time constant. The complementing action is also captured on the drain electrode, which exhibits mirrored but oppositely polarized temporal variations. Grounding the electrode allows separating the fast electronic components from those slow ionic ones, which is the key process introduced in this work, which is vital for better understanding of the function of the numerous components of the devices investigated. Experimental details will be given, these novel findings will be discussed and their implications for understanding the mechanism of the EDL formation will be presented.

9:15am **CA+AS+SS-WeM-6 Studying Tungsten and Alloys as Candidate Plasma Facing Material Using ToF-SIMS*, Xiao-Ying Yu,*** *Gabriel Parker, Tobias Misicko, Yan-Ru Lin,* Oak Ridge National Laboratory*; Tanguy Terlier,* Rice University*; Yutai Katoh,* Oak Ridge National Laboratory

Plasma facing materials (PFMs) are important in realizing fusion power. Tungsten (W) and alloys are considered primary candidates of PFMs due to their high melting points, high thermal conductivities, good neutron irradiation resistance, fast diffusion of hydrogen, low retention, and sputtering behaviors. However, technical challenges remain in adopting W and alloys as PFMs. In this presentation, we will share recent study cases of W and alloys using advanced microanalysis and chemical imaging, primarily time-of-flight secondary ion mass spectrometry (ToF-SIMS). SIMS is a powerful imaging mass spectrometry tool, and it can be used to reveal surface composition with high sensitivity or probe the material layer-by-layer and reveal spatial distributions in two-dimension or three-dimension. Due to parallel data acquisition, full spectral information consisting of elements, isotopes, and molecule permitted in the duty cycle is available in SIMS' chemical mapping. We will present a few case studies of potential PFMs using SIMS. First, High Flux Isotope Reactor (HFIR) irradiated single crystal tungsten (SCW) specimens from the FRONTIER collaboration campaigns were selected for spectral analysis and depth profiling. SCW coupons were subjected to shielded and unshielded neutron irradiation in HFIR. Prior to ToF-SIMS analysis, specimens were prepared using focused ion beam (FIB). To assure reliable peak identification and assignment, we performed analysis of pristine single crystal tungsten as a control. Mass spectra reconstructed from depth profiling show a variety of transmutation products in unshielded W, such as Rhenium, Osmium, and Tantalum. In contrast, not as many transmutation products were detected in the shielded irradiated W spectra. Second, W alloys were studied to verify the trace doping quantity of Boron (B) using the high mass sensitivity of SIMS spectroscopy, and measurements were verified using Raman. Depth profiling was used to verify the distribution of B within the W matrix. Third, we show that depth profiling with high spatial resolution can be used to map the grain boundaries in W alloys and assist the development of new materials and validate the engineering process. These recent studies provide results of the structural and compositional changes in W and alloys as potential PFMs, showing that SIMS can be a useful tool on elucidating alloy property changes and supporting material development for sustainable fusion in the future.

11:00am **CA+AS+SS-WeM-13 ToF-SIMS Acquisition Multiplexing - Concept, Applications, and Data Analysis*, Henrik Arlinghaus,*** 1) ION-TOF GmbH, Germany; 2) Institut für Hygiene, WWU, Germany*; Alexander Pirkl, Derk Rading, Julia Zakel, Ewald Niehuis,* ION-TOF GmbH, Germany **INVITED**

Time of Flight Secondary Ion Mass Spectrometry (ToF-SIMS) is a versatile technique for 2D and 3D analysis of surfaces. During the acquisition process, secondary ions are desorbed from the sample using one or more primary ion beams. These secondary ions are used to acquire a full mass spectrum at each voxel. Typically, a single acquisition mode is used when acquiring data, optimized for one specific potential aspect of the (unknown) sample. This becomes problematic when the number of acquisitions is constrained by the amount of sample available or the instrument time available. In the acquisition multiplexing approach, multiple acquisition modes, each of which is optimized for a specific performance aspect, are utilized during the acquisition process, resulting in multiple co-located datasets. Each of these acquired datasets may be analyzed individually in the traditional manner, or via algorithmic techniques such as Multivariate Statistical Analysis (MVSA) or Machine Learning (ML). Additionally, by taking advantage of the dataset's co-location property, it is possible to analyze all of the acquired data at once, finding aspects of the sample which span the data spectrally, spatially, and across acquisition modes.

We have applied this approach to vary numerous performance parameters of ToF-SIMS instruments, such as the primary ion beam current, the primary ion species, the focus of the beam, etc. Subsequently, analysis routines optimized for the parameter that was varied were applied to the datasets to make full use of the resulting data. One example is acquiring datasets using different primary ion beam currents, and then generating a High Dynamic Range (HDR) like dataset. High primary ion currents result in intense peaks which may saturate the detector signal. At the same time, low intensity signals are noisy at low primary ion currents. Another parameter which we varied is whether to optimize the primary beam for high mass resolution (and lower spatial resolution), or high spatial resolution (and lower mass resolution). This results in two datasets with fully complementary information. These may then be analyzed using machine learning based image fusion to generate a single high mass high

spatial resolution dataset.

11:30am **CA+AS+SS-WeM-15 Diamond Surface Analysis for Electronics and Quantum Applications**, ***Alastair Stacey***, Princeton Plasma Physics Laboratory **INVITED**

Characterising and controlling surface electronic and quantum states is an almost ubiquitous challenge for electronic and quantum technologies. The diamond material system is a particular example, where bulk states can be created with extreme purity but surface states, chemical and physical, are not yet well controlled or even understood.

In this presentation I will detail our efforts to analyse the diamond surface, with a variety of vacuum science techniques and theoretical analyses, and reveal some of the chemical challenges being faced in passivating and functionalizing this surface. I will show evidence that these surfaces remain significantly disruptive for quantum devices and present recent efforts in the development of high performance hydrogen terminated diamond transistors. Finally, I will forecast the remaining challenges and next steps for improving the surface science of this important quantum electronic material.

12:00pm **CA+AS+SS-WeM-17 ToF-SIMS Spectral Analysis Using Python**, ***Tobias Misicko***[1], Louisiana Tech University and Oak Ridge National Laboratory; *Nan Jiang, Xinghang Zhang, Yexiang Xue,* Purdue University; *Xiao-Ying Yu,* Oak Ridge National Laboratory

Time-of-Flight Secondary Ion Mass Spectrometry (ToF-SIMS) is a powerful surface analysis technique that enables spatially resolved chemical characterization of materials with high mass resolution and accuracy. However, analyzing ToF-SIMS data remains challenging due to the high dimensionality and large size of datasets resulting from parallel data acquisition. Previous efforts have largely depended on manual interpretation and the analyst's prior experience to apply dimensionality reduction techniques for material composition analysis. This process demands substantial human supervision and is hindered by the lack of open-source datasets and comprehensive, end-to-end code implementations for multivariate analysis pipelines, particularly for principal component analysis (PCA) and non-negative matrix factorization (NMF). In this work, we integrate both established and emerging methods tailored for ToF-SIMS spectral analysis, delivering an open-source, Python-based framework for intelligent mass spectral analysis to the ToF-SIMS research community. We demonstrate the application of PCA and NMF for spectral analysis and benchmark their performance using a quality-assured SIMS dataset.

[1] **JVST Highlighted Talk**

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