Chemical Analysis and Imaging at Interfaces
Room 303 - Session CA2-FrM

Chemical Analysis and Imaging at Interfaces Oral Session II
Moderators: Carles Corbella, NIST-Gaithersburg, **Xiao-Ying Yu**, ORNL

10:15am **CA2-FrM-9 Modulating Active Site for Water Electrolysis***, **Sen Zhang**,* University of Virginia **INVITED**

This presentation focuses on modulating active sites for oxygen evolution reaction (OER) through precise control of single atom/cluster sites and nanocrystal surfaces and interfaces. Emphasis is placed on the use of in situ and operando spectroscopic techniques, including X-ray absorption spectroscopy (XAS), Raman spectroscopy, and surface sensitive IR, to directly probe dynamic surface reconstruction, oxidation state evolution, and formation of catalytically active species under working conditions. These real-time insights reveal how applied potential induces active-phase transformations and how tuning active site coordination environment can optimize adsorption energetics and reaction intermediates. By correlating spectroscopic signatures with electrochemical performance, we establish robust structure–activity relationships that guide rational active-site engineering for highly efficient and durable OER electrocatalysts.

10:45am **CA2-FrM-11 Synthesis and in Situ Characterization of Simulated Hanford Tank Waste Models in Aqueous Solution***, **Tobias Misicko**,* Oak Ridge National Laboratory, USA*; Mackenzie Savage,* University of Virginia*; Gabriel Parker, John Lasseter,* Oak Ridge National Laboratory, USA*; Kaleigh Louque, Yang Xiao,* Louisiana Tech University*; Xiao-Ying Yu,* Oak Ridge National Laboratory, USA

The nuclear waste stored at the US Department of Energy's Hanford Site as an environmental remediation facility presents many major challenges. Mainly, the remediation of aluminum-containing tank waste generated from the separation of aluminum cladding of spent fuel rods requires processing and its complex dissolution chemistry is still not well understood. These wastes typically consist of three aluminum oxide/oxy-hydroxide species, namely gibbsite (γ-$Al(OH)_3$),[1] boehmite (α-AlOOH),[2] and bayerite (α-$Al(OH)_3$).[3] Targeted remediation of these solids is complicated by their dissolution and stability in solution as well as a wide range of particle sizes. Physical property information on the tank waste is scarce due to computational simulation limits and the radioactivity of the waste preventing laboratory handling. Herein we report a versatile hydrothermal synthesis technique enabling tunable and scalable production of gibbsite, boehmite, and bayerite in submicrometer for use as simulant materials for in situ analysis and related experimental investigations. Synthesized samples were characterized ex situ using X-ray Diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), time-of-flight secondary ion mass spectrometry (ToF-SIMS), and physisorption to verify structural and chemical properties. *In situ* particle analysis and imaging were subsequently investigated using the System for Analysis at the Liquid-Vacuum Interface (SALVI) microfluidic platform, which enabled probing of the vacuum-liquid interface with SEM and ToF-SIMS.[4] The insights gained from this work are expected to inform the development of remediation approaches and formulate remediation methodologies that are safe for both site personnel and the environment.

References:

[1] Page, J.S., *et al.*, *Journal of Hazardous Materials,* 384 (**2020**) 121318

[2] Peterson, R.A., *et al., Separation Science and Technology,* 42 (**2007**) 1719-1730

[3] Herting, D.L., *et al., US DOE Office of Environmental Management,* (**2002**), OSTI ID: 808264

[4] Yang, L., *et al., Lab on a Chip*, 11 (**2011**), 2481-2484

11:00am **CA2-FrM-12 Studying Electrified Catalysis at the Laboratory and at Synchrotron End Stations***, **Patrick Lömker**,* Scienta Omicron, Sweden*; Andrew Yost, Daniel Beaton, Xin Zhang,* Scienta Omicron

Catalysts are central to the transition from fossil-based to renewable energy systems, with both thermal and electrocatalysis playing key roles in enabling carbon-neutral production of chemicals and fuels. However, their investigation presents significant technical challenges. This contribution discusses advanced sample environments for Dip+Pull methodologies and cell-based electrocatalytic systems.

In Dip+Pull catalysis, recent developments emphasize improved usability and experimental efficiency. Sample cell designs have evolved from in-line electrode configurations to on-axis arrangements, thereby reducing electrolyte transport distances. This approach has been successfully applied to a range of systems, including CO reduction catalysis and studies of battery materials and their degradation mechanisms.

The second part focuses on electrochemical cell geometries, particularly configurations in which the catalyst acts as a barrier between the liquid phase and the vacuum environment. Such setups enable the investigation of membrane transport properties and the operando surface states of platinum catalysts and their intermediates under oxygen reduction reaction conditions.

A unifying aspect of both approaches is the investigation of the electrochemical double layer, which plays a critical role in governing catalytic activity and interfacial processes. Its characterization under these experimental conditions is discussed in detail.

11:15am **CA2-FrM-13 Energy Flux from Non-Thermal Discharges Measured via Nanocalorimetry: Toward Plasma Standard Development***, **Carles Corbella**,* National Institute of Standards and Technology (NIST)/ University of Maryland, College Park*; Aritra Sil, Berc Kalanyan, James Maslar,* National Institute of Standards and Technology (NIST)*; Lakshmi Ravi Narayan,* National Institute of Standards and Technology (NIST)/ University of Maryland, College Park*; William Osborn, Feng Yi, Andrei Kolmakov,* National Institute of Standards and Technology (NIST)

The use of rapid thermal probes to characterize plasma processes has been under development for a few decades. Despite a growing interest in quantifying the energy fluxes from low-temperature discharges used for surface processing, the decoupling of the elementary contributions from the different plasma species remains a challenge. Primary heat fluxes carried by ions, electrons, photons, and neutrals, are usually convoluted with secondary fluxes from chemical reactions, phase transitions, and recombination processes in calorimetry measurements. Nanocalorimeter sensors, which in our design consist of a Pt thin film microresistor lithographically defined on a free-standing, ultrathin SiN_x membrane with a thermal mass down to nJ/K, constitute an alternative solution to obtain instantaneous energy fluxes given their sensitivity, selectivity, and short response time. In this presentation, the potential application of nanocalorimetry to elucidate and control fundamental surface plasma-induced processes is motivated by the current needs of the coatings industry, where substrate temperature is a critical parameter. Here, we will discuss nanocalorimetric methods to discriminate the individual thermal fluxes from plasma species, such as the use of catalytic coatings to detect plasma radicals, the application of DC bias voltages to select impinging charged particles, and the exploration of interactions with light using optical filters. The presented prototype has been tested under hydrogen, helium, argon, and oxygen RF glow discharges operated in both inductive and capacitive modes. The energy flux, including the dominant participation of energetic ions in Ar plasma, ranges from 10 W/m^2 up to 1700 W/m^2. The experiments were performed in a dedicated research setup equipped with a Langmuir probe, retarding field energy analyzer, optical emission spectroscopy, and quadrupole mass spectrometry, to support and validate the nanocalorimetry measurements. The main objective is to develop plasma energy flux metrology as a standard plasma diagnostics tool that quantifies the thermal effect on plasma-exposed materials across different plasma sources and applications. The relevance of this emerging metrology to the plasma-materials community is justified by the nanocalorimeters' real-time and sensitive performance, robustness, and integrability to other metrology platforms.

Author Index

Bold page numbers indicate presenter

— B —
Beaton, Daniel: CA2-FrM-12, 1
— C —
Corbella, Carles: CA2-FrM-13, **1**
— K —
Kalanyan, Berc: CA2-FrM-13, 1
Kolmakov, Andrei: CA2-FrM-13, 1
— L —
Lasseter, John: CA2-FrM-11, 1
Lömker, Patrick: CA2-FrM-12, **1**
Louque, Kaleigh: CA2-FrM-11, 1
— M —
Maslar, James: CA2-FrM-13, 1
Misicko, Tobias: CA2-FrM-11, **1**
— O —
Osborn, William: CA2-FrM-13, 1
— P —
Parker, Gabriel: CA2-FrM-11, 1
— R —
Ravi Narayan, Lakshmi: CA2-FrM-13, 1
— S —
Savage, Mackenzie: CA2-FrM-11, 1
Sil, Aritra: CA2-FrM-13, 1
— X —
Xiao, Yang: CA2-FrM-11, 1
— Y —
Yi, Feng: CA2-FrM-13, 1
Yost, Andrew: CA2-FrM-12, 1
Yu, Xiao-Ying: CA2-FrM-11, 1
— Z —
Zhang, Sen: CA2-FrM-9, **1**
Zhang, Xin: CA2-FrM-12, 1