Applied Surface Science
Room 321 - Session AS-ThM

Complex Data Analysis For Applied Surface Science

Moderators: Dr. Jonathan Counsell, Kratos Analytical Inc, **Dr. Jeffrey Terry**, Illinois Institute of Technology

11:00am **AS-ThM-13 Data-Driven Analysis Using Explainable AI (XAI) for Surface Analysis Data, Including Mass Images**, ***Satoka Aoyagi,*** Seikei University, Japan **INVITED**

Sophisticated analysis methods, such as secondary ion mass spectrometry, provide rich information about a sample, from which we could generally extract and interpret only a part of it. The effective application of machine learning methods to such data is useful for extracting hidden yet important information with which to characterize the sample. Although there are a variety of methods to analyze complex datasets, the methods that also provide the information on the analysis processes are crucial for analyzing scientific data. This presentation focuses on such methods, known as explainable AI (XAI). Unsupervised learning methods, such as principal component analysis (PCA), non-negative matrix factorization (NMF) and autoencoder (AE), are useful for understanding the outline of the data and sample that are analyzed. Supervised learning methods are powerful for further analysis, such as exploring related factors of a particular material or characteristic in a sample. Combining unsupervised and supervised learning methods provides detailed information on related variables, such as groups of mass peaks from the same material, and on the relationships between particular sample conditions and chemical structures. The application of unsupervised learning methods, such as PCA, NMF and AE, and supervised learning methods, such as artificial neural network (ANN)-based methods [1-3] and Random Forest [4], to organic complex sample datasets is introduced. Moreover, ANN-based systems are useful in dealing with datasets including non-linear factors such as matrix effects, which cause peak intensity changes regardless of the concentration of a target material. The weights of ANN systems indicate which variables are important for the purpose of the analysis, suggesting whether an ANN system outputs answers depending on the appropriate variables. For example, quantitative analysis including interface evaluation of multilayers in depth profiles often requires correction methods. ANN is generally powerful for the ANN-based system developed for secondary ion mass spectrometry (SIMS) data of three-component systems, which is introduced here.

[1] S. Aoyagi, *J. Vac. Sci. Technol. A* **41**, 063202 (2023).

[2] S. Aoyagi, K. Matsuda, *Rapid Commun. Mass Spectrom*., **37**(4), e9445 (2023).

[3] S. Aoyagi et al., *Anal. Chem*., **95**(40), 15078-15085 [tel:15078-15085] (2023).

[4] S. Aoyagi et al., *Anal. Chem*., **93**(9), 4191-4197 [tel:4191-4197] (2021).

11:30am **AS-ThM-15 Data-Driven Automation for X-Ray Photoelectron Spectroscopy (XPS)**, ***David Valley***, *James Johns, Jiayue Lin, Jennifer Mann, Kateryna Artyushkova,* Physical Electronics

We are presenting a set of data-driven tools aimed at simplifying XPS analysis while using data-driven decision making processes to maintain flexibility for advanced use. The approach uses both machine learning and analytical models to automate key steps in both sample setup and data acquisition.

In XPS analysis, typically samples are small, or cut to a smaller size to accommodate the instrument. As a result, multiple samples will fit on a sample holder (platen) and in a high throughput instrument, multiple sample holders can be put in the instrument at once, resulting in potentially 100s of samples in the instrument at any given time.XPS users have identified pain points of the repetitive, tedious work of creating analysis points on each sample mounted on the platen. To address this necessary, but time-consuming step usually performed by the user, we have developed three machine learning models to automatically detect the platen type, platen-photo alignment, and the sample locations on the platen. These are used to automatically qualify the platen upon introduction to the instrument and populate potential analysis positions.

A second set of tools is to use statistical methods to optimize the basic analysis routines that are the starting point for XPS analysis. XPS analysis requires the acquisition of a wide energy range survey spectrum to identify the elements on the surface of the sample, followed by narrow scans that focus on the elements of interest to get higher signal-to-noise for quantification purposes and/or reveal the chemical state of a particular element. We introduce a data-based optimization routine to determine the time balance to get the best data quality for the region acquisitions scans based on a measurement of the survey scan. From the signal-to-noise of the equal time-based survey acquisition we extrapolate the expected error in the atomic percent quantification and choose optimum routines. The optimum is not a unitary choice and there is a pareto optimal front of possible acquisitions states. Based on user input we create an objective function which fits the user's desire for the acquisition result.

11:45am **AS-ThM-16 Computational Assessment of Environment-Dependent Mineral Surface Transformations**, ***Jennifer Bjorklund***, *Sara Mason,* Center for Functional Nanomaterials, BNL

Understanding how mineral surfaces evolve and react under environmentally-relevant conditions is crucial to predicting interfacial processes governing resource recovery, nutrient cycling, and aqueous geochemistry. Minerals like pyrite (FeS_2) and struvite ($NH_4MgPO_4 \cdot 6H_2O$) are environmentally and technologically important systems whose surface chemistry is sensitive to aqueous conditions, with reactivity ultimately controlled by termination, hydration, protonation state, and local atomic configuration. Here, using an approach that combines Density Functional Theory (DFT) calculations with atomistic thermodynamics, we characterize surface stability, transformation pathways, and their impact on reactivity under relevant chemical conditions.

For pyrite, we explicitly compare hydroxylation and protonation of exposed surface sites to determine stable interfacial configurations as a function of aqueous conditions (pH, temperature, pressure). We further assess possible modes and energetics of arsenic (As) incorporation into near-surface and surface sites, identifying preferred structural motifs and their influence on local electronic structure. Building on these structural models, we evaluate gold (Au) adsorption energetics to understand how environmentally induced surface transformations and dopant incorporation modify interfacial binding and reactivity relevant to gold deposition and ore formation processes.

For struvite, we investigate pH-dependent stability of multiple surface terminations derived from the underlying bulk composition, explicitly sampling variations in protonation and hydration states. By constructing thermodynamic comparisons of these stable configurations, we identify dominant surface structures under varying aqueous conditions and quantify how protonation and hydration influence stability and reactivity. These results provide molecular-level insight into the transformations of struvite surfaces under realistic environmental conditions associated with nutrient release.

Together, these insights highlight how aqueous conditions drive surface transformations in mineral systems and how these transformations control downstream interfacial reactivity. By linking surface stability, protonation/hydration, dopant incorporation, and adsorption energetics, this work provides a unified framework for understanding environmentally-relevant responsible mineral interfaces at the atomic scale.

12:00pm **AS-ThM-17 Surface and Subsurface Chemistry of IrOx Electrocatalysts: A Multi–Technique Approach Integrating ToF–SIMS, XPS, and Multivariate Analysis**, ***Lonneke van Eijk***, Colorado School of Mines, USA*; Genevieve Stelmacovich,* LAM Research*; Michael Walker,* Colorado School of Mines, USA*; Rebecca Hamlyn,* Lawrence Berkeley Lab, USA*; Zhao Li, Xiaoping Wang,* Argonne National Laboratory, USA*; Ethan Crumlin,* Lawrence Berkeley Lab, USA*; Alexey Serov,* Oak Ridge National Laboratory, USA*; Deborah Myers,* Argonne National Laboratory, USA*; Svitlana Pylypenko,* Colorado School of Mines, USA

Understanding the surface and subsurface chemistry of IrO_x electrocatalysts is essential for advancing performance of Proton Exchange Membrane Water Electrolyzers (PEMWEs). Commercial IrO_x powders exhibit diverse physicochemical properties and the interplay between various properties and activity and stability is still under investigation. In this work, six commercial IrO_x catalysts were examined. Initial studies used a suite of characterization methods including bulk structural and compositional techniques such as XRD, PDF, SAXS/USAXS, and XAS, as well as surface sensitive structural and compositional methods BET surface area analysis and XPS. These measurements were combined with electrochemical properties, including electrical conductivity and electrochemical measurements in rotating disk electrode configuration to assess activity and stability. This work also includes analysis of the same set of samples with ToF–SIMS, to further access surface and near-surface chemical information, extending into the bulk through depth profiling, to corroborate

and complement other methods. This is accomplished using a three–stage framework for interpreting IrO_x surface chemistry using PCA.

First, ToF–SIMS is examined independently to determine how fragment distributions, depth–dependent signals, evaluated in the context of matrix effects to understand how they correlate with IrO_x composition, oxygen environments, and defect-related chemistry. This step clarifies how ToF–SIMS can be reliably applied to complex oxide electrocatalysts and what chemical information it uniquely provides. Second, ToF–SIMS results are correlated with XPS to refine chemical-state assignments. ToF–SIMS fragment trends are used to validate or challenge XPS peak-fitting models, improving confidence in oxidation-state and oxygen-species interpretation. Finally, ToF–SIMS and XPS parameters are correlated to bulk compositional and structural parameters along with electrochemical metrics. PCA reveals cross–technique correlations that reveal how surface chemistry and subsurface structure relate to catalyst performance and robustness. By combining complementary vacuum–based methods with statistical analysis, this work demonstrates how multi–characterization strategies resolve ambiguities that cannot be addressed by any single technique. The resulting framework provides a generalizable approach for using ToF–SIMS and XPS together to interrogate complex oxide surfaces and highlights the power of integrated surface analysis for understanding structure–chemistry relationships in advanced electrocatalysts.

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