Advances in Battery Engineering, Interface Design, and Characterization Mini-Symposium
Room 319 - Session BT-ThA

Advanced Characterization of Battery Interfaces

Moderators: Samantha Rosenberg, Kairos Power, **Xiao-Ying Yu**, Oak Ridge National Laboratory, USA

2:15pm **BT-ThA-1 Designing Materials for High Performance Energy Storage Applications at Solid-Solid and Solid-Liquid Interfaces***, Ajay Karakoti,* Pacific Northwest National Laboratory **INVITED**

The performance of energy storage materials is fundamentally governed by interfacial phenomena, where ion transport, nucleation, and chemical degradation dictate cycle life and energy density. Probing these buried, dynamic interfaces requires combining advanced characterization, computational modeling, and augmented by data-driven workflows. This talk highlights two complementary efforts that leverage multiscale characterization, in-situ probes, and emerging computational frameworks to elucidate and engineer interfaces in next-generation solid-state and grid-scale lead-acid batteries.

The first part of the talk demonstrates a discovery framework for designing solid–solid interfaces in lithium-ion solid-state batteries by coupling state-of-the-art machine learning potentials with physics-based simulations on cloud high-performance computing. Screening across millions of candidate compositions yielded a tractable, shortlisted set of promising solid electrolyte candidates that were subsequently synthesized and experimentally validated. Postmortem characterization of cycled cells using SEM-EDS, XPS, and PXRD reveals how cathode–electrolyte interfacial stability and structural evolution govern long-term performance, informing design rules for composite electrolytes.

The second part of the talk identifies opportunities for utilizing data-driven frameworks to bridge our understanding of failure mechanisms in lead-acid batteries. The complex chemistry of this system necessitates probing reactions using a multi-modal toolkit spanning in-situ atomic force and optical microscopy, solid-state NMR, XPS, and synchrotron methods that link macroscopic failure modes, such as, irreversible sulfation and electrolyte stratification to atomic-scale interfacial structure. The rich, multimodal datasets emerging from these studies present a unique opportunity to bridge length scales and provide inputs for future machine learning and autonomous workflows aimed at formulating an overarching mechanistic understanding of battery failure and accelerating the discovery of new materials.

2:45pm **BT-ThA-3 Advances in Neutron and X-Ray Imaging and Tomography Methods for in Situ Characterization of Lithium-Ion Batteries at NIST***, David Jacobson, Jacob LaManna, Elias Baltic, Daniel Hussey,* National Institute of Standards and Technology (NIST) **INVITED**

Neutron imaging has played an important role in characterizing electrochemical systems in the last 20 years. Neutrons penetrate metallic components of battery systems yet remain sensitive to changes in lithium concentration and/or hydrogenous electrolyte concentration, thanks to the large attenuation cross-section for ^{6}Li and H. When combined in operando with X-rays, it is possible to obtain a multi-modal picture of the electrochemical system, allowing for phase separation that is not possible with individual probes alone.

At NIST, the neutron imaging program operates two instruments, both with simultaneous neutron/X-ray tomography capabilities: the BT2 NIST Neutron Imaging Facility (NNIF), and the NG6 Cold Neutron Imaging Instrument (CNII).

In this presentation, we will discuss the current status of these facilities, planned future upgrades, the impacts on battery characterization, and examples of the techniques as applied to imaging lithium batteries. During the NIST Center for Neutron Research (NCNR) shutdown, we performed experiments at other facilities that will be discussed here. In addition, we have pursued upgrades to the NIST imaging facilities to improve spatial resolution, field-of-view, data acquisition, and data analysis speeds.

Because neutron sources are lower in intrinsic brightness than synchrotron X-ray sources, they require longer data collection times for 3D data sets. To improve neutron data acquisition speeds NIST has deployed machine-learning methods to reduce image noise allowing for shorter image acquisition times. NIST is also incorporating a high-speed server and network to provide near real-time data analysis to facility users. Furthermore, we will discuss new cold neutron imaging methods for multi-scale characterization and the status of the NIST neutron microscope project.

3:15pm **BT-ThA-5 Airless Transfer for Battery Materials: Preserving Native Surface Chemistry for Analysis***, Forrest Nichols, Dan Sullivan,* Eurofins EAG

Analysis of materials that react with atmospheric chemicals, such as batteries, require airless preparation and airless transfer into the materials analysis tools. In this paper we describe the sample preparation to remove a cathode from a Li ion battery in a glove box in a dry room and placed into an airless transfer box. The sample is airlessly transferred into an XPS tool for analysis. This is followed by exposure of the sample to air for one minute and then another XPS analysis. The sample is then exposed to air for two days and a final analysis is performed. The change in the surface composition and chemistry is investigated as a function of air exposure time. Airless transfer enables accurate surface analysis of battery materials by preserving native chemistry.

3:30pm **BT-ThA-6 ASSD Student Award Finalist Presentation: Strategies for ToF-SIMS of Buried Interfaces in Solid Polymer Electrolyte Batteries***, Reyhane Shavandi*[1]*, Meghan Burns, Neelam Sunariwal,* University of Illinois at Chicago*; Sanja Tepavcevic,* Argonne National Laboratory*; Luke Hanley,* University of Illinois at Chicago

The solid electrolyte interphase (SEI) formed at lithium metal interfaces plays a critical role in determining the stability and performance of solid state batteries with polymer-based electrolytes. Time-of-flight secondary ion mass spectrometry (ToF-SIMS) combined with ion sputtering was used here to investigate the composition and spatial distribution of the buried SEI formed with a mixed of bis(fluorosulfonyl)imide (LiFSI) and poly(ethylene oxide) (PEO) electrolytes between lithium electrodes, Li|PEO-LiFSI-$LiNO_3$|Li. Gaseous cluster ion beam (GCIB) sputtering revealed that the SEI remained adhered to the Li electrode rather than to the polymer electrolyte, indicating its mechanical robustness and chemical stability. Depth profiles from ToF-SIMS analyses further revealed that addition of small amounts of $LiNO_3$ to the LiFSI/PEO electrolyte led to the formation of a nitrogen-rich SEI layer adjacent to the Li electrode surface, characterized by a strong CN^- signal identified by high mass accuracy peak assignment (S.R. Shavandi, *et al.*, *J. Vac. Sci. Technol. B* 44, 2026, 012401). The presence of CN^- signal indicated the presence of nitrogen-containing species within the inner SEI and a corresponding reduction of LiF-content within the SEI. Based on these findings, an anode free system using a Cu electrode, which both less expensive and nonflammable was examined. The Cu electrode was coated with a thin Au layer and Al_2O_3 nanoparticles were incorporated into the solid-state electrolyte to form Li|PEO-LiTFSI-$LiNO_3$+Al_2O_3|Au/Cu. $LiNO_3$ was added to create a nitrogen-rich interfacial layer with excellent Li-ion transport ability, while Al_2O_3 nanoparticles served to mechanically reinforce the pure polymer electrolyte. Li plating onto bare Cu during electrochemical cycling is hindered by a large nucleation overpotential that leads to non-uniform and dendritic growth, but the Au coating onto Cu is thought to create a lithiophilic interface by forming Au–Li intermetallics that reduce the nucleation barrier and enable uniform Li deposition. Current efforts focus on combining the benefits of the nitrogen-rich interface with this lithiophilic Au-alloying strategy to better understand where Li preferentially plates and how the nitrogen-rich layer forms and evolves on Au/Cu versus metallic Li surfaces. ToF-SIMS directly probed Li nucleation and interfacial chemistry with depth profiling using either O_2 or GCIB sputtering guns to achieve high sensitivity to Li and buried interfacial species. Both two-dimensional depth profiling and three-dimensional chemical imaging were utilized to resolve the spatialdistribution of Li relative to the Au interlayer.

3:45pm **BT-ThA-7 Correlative Imaging and Surface Analysis of Air-Sensitive Materials***, Lu Ping,* Thermo Fisher Scientific, USA*; Hsiang-Han Tseng, Helen Oppong-Mensah, Tim Nunney,* Thermo Fisher Scientific, UK

Understanding complex materials increasingly requires the correlation of structural, morphological, and chemical information acquired across multiple analytical platforms. Imaging techniques such as scanning electron microscopy provide essential context on microstructure, defects, particles, and regions of interest, while surface-sensitive spectroscopies such as X-ray photoelectron spectroscopy reveal local chemistry, bonding, and elemental composition. For air-sensitive materials, however, transferring specimens between instruments can alter the very surfaces and interfaces under investigation, making reliable correlation especially challenging.

[1] **ASSD Student Award Finalist**

This presentation describes recent developments enabling vacuum-protected transfer between electron microscopy and surface analysis instrumentation. The approach combines SEM-based imaging and site selection with subsequent XPS analysis using a two-way vacuum transfer capability, allowing selected regions to be analyzed while minimizing atmospheric exposure. The ability to move samples between instruments under controlled conditions supports iterative, correlative investigation without compromising reactive surfaces.

Examples will be presented from air-sensitive materials, including Li-ion battery components, where preservation of native surface chemistry is critical for interpreting degradation, interphase formation, and chemistry-structure relationships. These results illustrate how vacuum-integrated correlation of imaging and spectroscopy can provide a completer and more reliable picture of materials that are difficult to characterize using isolated techniques alone.

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