Department of Electrical and Computer Engineering
Materials Engineering Program
Center for Integrated Bio and Nano Systems
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Determination of Structural Evolution during the Initial Formation of the Solid Electrolyte Interphase in Lithium-Ion Batteries Using in situ Liquid SIMS

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Abstract: The solid–electrolyte interphase (SEI) dictates the performance of most batteries, but the understanding of its chemistry and structure is limited by the lack of in situ analysis tools. In this presentation, we show a dynamic picture of the SEI formation in lithium-ion batteries using in situ liquid secondary ion mass spectrometry (SIMS). [%5Cl%20%22_ENREF_1%22%20%5Co%20%22Zhu,%202015%20#4%22]1-2 One of the unique advantages of in situ liquid SIMS approach is simultaneous examination of reactants, products, reaction intermediates and electrode surface under operation conditions on a molecular level. More importantly, excellent depth resolution of this new technique allows operando characterization of formation and dynamics of an electric double layer. Our data suggest that before any interphasial chemistry occurs (during the initial charging), an electric double layer forms at the electrode-electrolyte interface due to the self-assembly of solvent molecules. The formation of the double layer is directed by Li+ and the electrode surface potential. The structure of this double layer predicts the eventual interphasial chemistry; in particular, the negatively charged electrode surface repels salt anions from the inner layer and results in an inner SEI that is thin, dense, inorganic and fluorine-depleted in nature. It is this dense layer that is responsible for conducting Li+ and insulating electrons, the main functions of the SEI. An electrolyte-permeable and organic-rich outer layer appears after the formation of the inner layer, which may help de-solvation and solvation of Li+ during charging-discharging. In the presence of a highly concentrated, fluoride-rich electrolyte, the inner SEI layer has an elevated concentration of LiF due to the presence of anions in the double layer. These real-time nanoscale observations will be helpful in engineering better interphases for future batteries. (1. Nano Lett 2015, 15 (9), 6170; 2. Nat Nanotechnol 2020, 15 (3), 224).

Short Bio: Dr. Zihua Zhu is a Senior Chemist of Environmental Molecular Sciences Laboratory at Pacific Northwest National Laboratory. He received B.Sc. & M.Sc. from Peking University, and Ph.D. from Pennsylvania State University, all in chemistry. He joined PNNL in 2006. Dr. Zhu's research has focused on the secondary ion mass

spectrometry (SIMS) field for over 20 years, and he has published 160+ articles in high impact journals. He was a key inventor (with 4 USA patents) of in situ liquid SIMS and is leading the applications of this new capability on better understanding complex chemistries occurring at solid-electrolyte interphase in novel Li ion batteries, electro-catalysis, biofilm growth, cell attachment, aerosol formation, ion solvation and initial nucleation of nanoparticle formation. His awards include American Vacuum Society-ASSD Peter M.A. Sherwood Mid-Career Professional Award, Federal Laboratory Consortium for Technology Transfer (FLC) Awards, and R&D 100 Award.

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