CHEMICAL ENGINEERING

DISTINGUISHED YOUNG SCHOLARS SERIES

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Computational Surface and Interfacial Science of Lithium Ion Batteries

ABSTRACT: Lithium ion batteries are widely used in consumer electronics and continue to impact the transportation sector through increased use of electric vehicles. In recent years, ab intio methods – primarily based on density functional theory (DFT) - have played a pivotal role in describing the materials physics of Li^+ insertion and removal in different electrode materials. These methods have furthermore enabled high-throughput screening of promising Li-ion battery chemistries based on bulk material properties. Recent advances notwithstanding, the practical performance of Li-ion batteries remains limited by a variety of complex reactions at surfaces and interfaces. These reactions, however, remain poorly understood at the molecular scale. In this talk, I will describe theoretical and computational analyses of model interfacial reactions in Li-ion batteries. The reactions of interest are specifically selected in order to target widespread problems relevant to various battery chemistries.

I will first describe how ab initio thermodynamics can be used to determine the surface structure of lithium manganese oxide (LMO), a common Li-ion cathode material.[1] The LMO surfaces identified from this study are subsequently used as model substrates for computational analysis of (1) side reactions with the electrolyte and (2) LMO surface passivation with protective coatings.[2–5] Building off this work, I will then discuss a multi-scale modeling approach for analysis of solid-solid interfaces in Li-ion batteries.[6] In particular, I will focus on the interface between lithium lanthanum titanate (LLTO) solid electrolytes and Li metal anodes. LLTO is known to be unstable against Li metal, making this a compelling model system to both understand the onset of interfacial decomposition chemistry and to develop rational strategies to stabilize these solid-solid interfaces. Analogous to the thermodynamic analysis of LMO surfaces, a Li grand potential ensemble is used to evaluate Li/LLTO models that are likely to exist under experimental conditions. Ab initio molecular dynamics simulations are then used to elucidate trends in interfacial reconstruction mechanisms. Based on

these dynamic simulations and a DFT-based electronic structure analysis, we propose that a thin La_2O_3 interlayer could stabilize LLTO against Li metal. This theory-guided interfacial design strategy is validated by in situ X-ray scattering measurements. These experimental studies reveal that the degradation of the LLTO surface following Li deposition can be effectively mitigated by ~2 nm of a La_2O_3 coating.[7]

Taken together, this work highlights fundamentals of surface and interfacial reaction chemistry in Li-ion batteries based on principles of materials thermodynamics, kinetics, molecular dynamics, and electronic structure. The insights gained through these computational studies open new avenues to rationally tailor interfaces for energy storage devices with higher energy density and longer lifetimes.

BIOGRAPHY: Robert Warburton received his B.S. in Chemical Engineering from The Ohio State University in 2014. He then moved on to Purdue University to pursue his Ph.D. under the supervision of Prof. Jeff Greeley. His research at Purdue focused on first principles modeling of interfaces in lithium ion batteries, and he received his degree in 2019. During his graduate studies, he also spent 8 months working under the guidance of Dr. Larry Curtiss in the Materials Science Division at Argonne National Laboratory as a Department of Energy Office of Science Graduate Research (SCGSR) Fellow. While at Argonne, he studied computational methods to model charge transfer in solids and at interfaces for applications in lithium ion and lithium air batteries. Robert is currently a Postdoctoral Associate with Prof. Sharon Hammes-Schiffer in the Department of Chemistry at Yale University where his research focuses on theoretical and computational modeling of proton-coupled electron transfer and electric fields at electrochemical interfaces.

LECTURE 1:00 - 2:00 Zoom
Networking Hour on Zoom Following

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