CHEMICAL ENGINEERING

DISTINGUISHED YOUNG SCHOLARS SERIES

NAV NIDHI RAJPUT

Monday, July 31, 2017

Postdoctoral Fellow
LBNL

Designing Optimal Electrolytes for Energy Storage Devices Using Coupled High Throughput *Ab Intio* Calculations and Molecular Dynamics Simulation

ABSTRACT: Today, the pursuit of transformative gains in the performance of electrical energy storage (EES) systems and industrial technologies are intrinsically a material's problem, which requires development of novel electrode materials, electrolytes, and architecture. Such innovations of novel electrode as well as electrolytes for future EES systems lie inherently in the computationally driven design of materials by obtaining a fundamental understanding of the interplay between events scaling over wide spatial and temporal ranges. My work comprises of developing infrastructure for high throughput computations for the Joint Center for Energy Storage (JCESR) and Electrolyte Genome (Fig.1), which is a coupled first-principle and classical molecular dynamics code-base for rapid understanding and design of optimal electrolytes. The composition of electrolytes has critical implications for the performance of current and future energy storage systems; from the formation and stability of the electrode-electrolyte interface to the transport properties, speciation, and viscosity of the bulk electrolyte. In this work, I present a multi-scale modeling approach for salts and solvents important to multivalent (e.g., Mg^{2+}, Ca^{2+} and Zn^{2+}), chemical transformation (e.g., Li-S), and redox flow batteries. We uncover a novel effect between concentration dependent ion pair formation and anion stability at reducing potential, e.g., at the metal anode. We find that both Mg and Zn electrolytes are highly prone to ion pair formation, even at modest concentrations, for a wide range of solvents with different dielectric constants, which have

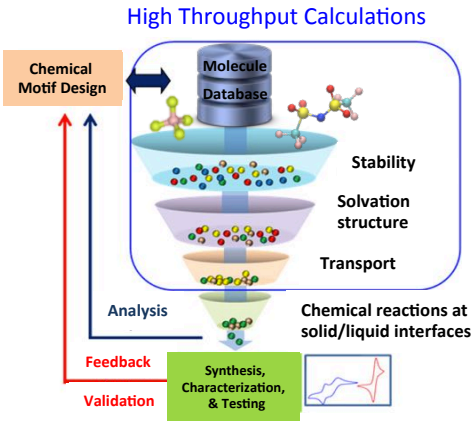

Figure 1 Schematic of down-selection of candidate molecules for electrical energy storage applications based on high-throughput computations using quantum chemical and molecular dynamics simulations combined with experimental validation and feedback.

implications for dynamics as well as charge transfer. Specifically, we observe that, at Mg metal potentials, the ion pair undergoes partial reduction at the Mg cation center ($Mg^{2+} \rightarrow Mg^{+}$), which competes with the charge transfer mechanism and can activate the anion to render it susceptible to decomposition. We also study the effect of salt anion and the solvent on the solvation structure and dynamics of Li-polysulfide in the solution for application in Li-S battery. It is observed that the polysulfide chain length has a significant effect on the ion-ion and ion-solvent interaction as well as on the diffusion coefficient of the ionic species in solution. We considered ionic liquid tethered ferrocene catholyte as redox center utilizing the Fc/Fc^{+} reaction for improving the performance of non-aqueous redox flow battery materials. It was observed that at solubility limit, the precipitation of solute is initiated through agglomeration of contact-ion pairs due to overlapping solvation shells. This works shows that the combination of multi-scale modeling with experimental techniques provides unprecedented insight into the origin of the electrochemical, structural, and transport properties of electrolytes, which is crucial in designing optimal electrolytes for beyond Li-ion batteries.

BIOGRAPHY: Nav Nidhi Rajput is a postdoctoral research associate working on Electrolyte Genome project with Dr. Kristin A. Persson in Energy Technologies Area division at Lawrence Berkeley National Laboratory. She received her M.S. and Ph.D. degrees in Chemical Engineering as a member of Prof. Francisco Hung's group at the Louisiana State University. Her research focuses on predicting and understanding the unique physical properties of liquid solutions, nanoporous materials and confined fluids—using computer simulations—for applications in nanostructured materials and energy storage.

LECTURE 4:00 – 5:00 (PAA) A110
Happy Hour in Benson Hall Lobby Following

CHEMICAL ENGINEERING
UNIVERSITY of WASHINGTON
Knowledge and solutions for a changing world