Mesoscale Priority Research Direction

Rational Design of Electrode/Electrolyte/Membrane Interfaces for Energy Storage

Opportunity

Scientific challenge: The key challenge is to understand charge transport in electrode materials and interfacial charge transfer, selective charge and molecular transport, and reactions at electrode-electrolyte interfaces in energy storage systems. The essential issues include: i) coupling of electron/ion transport in electrode materials; ii) formation, composition, and stability of the solid electrolyte interface (SEI); iii) design of electrolytes and membranes for optimum performance and compatibility with existing electrodes; iv) influence of membrane chemistry and morphology on selective transport; v) factors influencing the pore network in membranes; vi) detrimental dendrite formation and growth; vii) interaction of electrolytes with polymers; viii) mechanism of electrolyte decomposition and membrane degradation; ix) and chemical and mechanical stability of membranes under normal and extreme operating conditions.

Current state of understanding: There is only a phenomenological understanding of electrochemical processes at the mesoscale, which precludes rational selection of materials and design of interfaces for next-generation energy storage.

Meso Challenge

What makes it meso? Interfacial processes, such as SEI formation and dendrite growth, lie at the mesoscale for two reasons: a) the physics and chemistry are not correctly described by continuum models; and b) they occur at time and length scales that they cannot be treated with density functional theory (DFT) alone.

Approach

What can be done to address the challenge? Integrate *predictive modeling* starting at the DFT level up to coarse-grained reactive and kinetic models with *novel synthesis* methods and high resolution time resolved *in situ* characterization using aberration corrected microscopes and advanced light sources to understand interface evolution and transport processes in energy storage systems. Models must be coarse-grained systematically using molecular dynamics with reactive force fields, kinetic Monte Carlo simulation, and phase field modeling. The modeling and experimental results to be collected in an open source database of morphology, binding and migration energies, diffusion coefficients, interface thickness and stability, and mechanical properties. Data to be analyzed for trends toward identifying promising compositions and interfacial characteristics. Experiments can serve to verify and validate the models, while modeling can guide the synthesis efforts.

Impact

How will pursuit of the research direction, including the meso opportunity, impact the scientific challenge? Fundamental understanding of electrochemical interfaces will enable scalable synthesis of energy storage materials, including ionic liquid (IL)-based systems, that feature selective ion and molecular transport, enhanced safety and performance, and low cost.

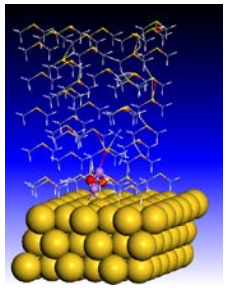
References: **Girishkumar *et al*, *J. Phys. Chem*. (2011); Yang *et al*, *Chem Rev*. (2011) ; Li *et al*, *Adv. Energy. Mat*. (2011)**

U.S. DEPARTMENT OF ENERGY | Office of Science

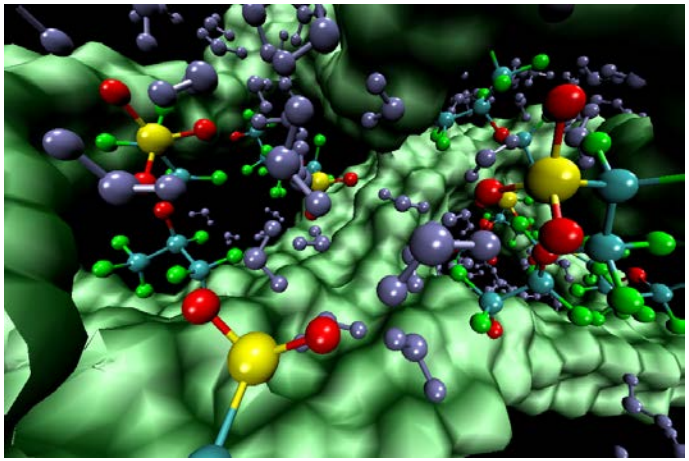
Contact: Michel Dupuis, michel.dupuis@pnnl.gov , and Ram Devanathan, ram.devanathan@pnnl.gov , Pacific Northwest National Laboratory

Mesoscale Priority Research Direction

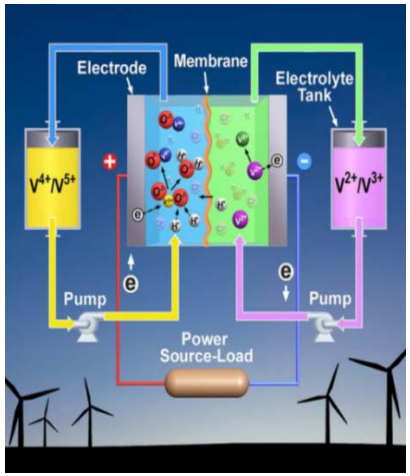
Mesoscale is a bridge between molecular and device scales



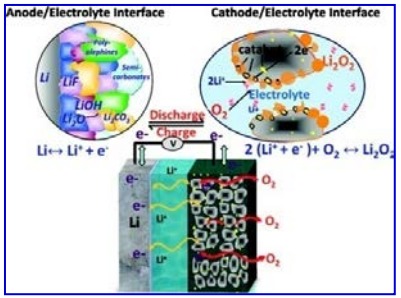
Molecular scale:
DFT & NMR, QENS



Interfaces at mesoscale:
Modeling involving coarse-grained MD, Kinetic MC, and Phase field models, classical DFT (cDFT) for appropriate time and length scales for reactivity and transport in high ionic concentrations
High resolution *in situ* characterization with time / ensemble sampling is critical to model validation



Yang *et al*, *Chem Rev*. (2011)
Li *et al*, *Adv. Energy. Mat*. (2011)



Girishkumar *et al*, *J. Phys. Chem*. (2011)

Device scale:
Continuum models of heat, charge, and mass transfer; *Device characterization*

U.S. DEPARTMENT OF ENERGY | Office of Science

Contact: Michel Dupuis, michel.dupuis@pnnl.gov , and Ram Devanathan, ram.devanathan@pnnl.gov , Pacific Northwest National Laboratory