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Rational design of nanostructured polymer electrolytes and solid-liquid interphases for lithium batteries

ABSTRACT: Understanding basic science and engineering principles that underpin performance of electrochemical energy storage technologies is imperative for significant progress in portable electrical devices. In this regard, metal-based batteries that are composed of a reactive metal (e.g., Li, Na, Al) as anodes have attracted attention because of their potential to improve anode-specific capacity by as much as 10-fold compared to the current state-of- the-art Li-ion batteries that use graphitic anodes. Perhaps their greatest advantage lies in the possibility of using of a Li-free, high-capacity cathodes like oxygen or sulfur that can improve the gravimetric energy density of batteries from ~0.3kWh/kg to ~12kWh/kg (i.e., comparable to the useful energy available from combustion of hydrocarbons). A persistent challenge with batteries based on metallic anodes is their propensity to fail by short-circuits produced by dendrite growth during battery recharge, as well as by runaway of the cell resistance due to internal side reactions with liquid electrolytes. In this talk, I will discuss my thesis research that utilizes multiscale transport modeling and experiments to fundamentally understand and thereby develop rational designs for polymer electrolytes and electrode - electrolyte interphases that overcome these challenges.

There have been several studies in the literature dedicated to the prevention of dendrite growth by means of a high modulus physical barrier. However, electrolytes/ separators with high mechanical strength tend to have low ionic conductivity, thus limiting their practical use. Using continuum analysis of interfacial ion transport, we have shown that the length-scale on which transport occurs

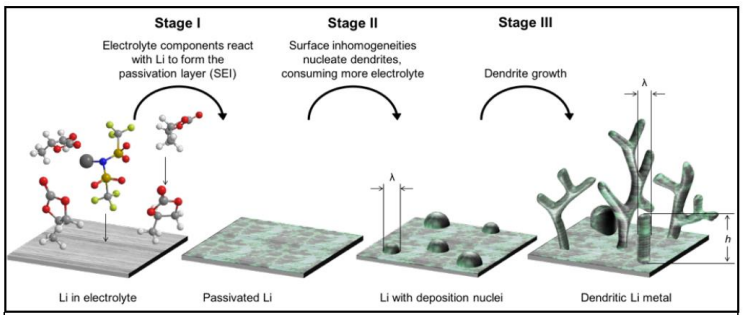

Figure Description: Different stages of instabilities associated with electrodeposition on a lithium metal anode.

near the electrodes could be as important as the electrolyte modulus in stabilizing metals against dendrite formation. In brief, this study concluded that dendrites can be prevented from crossing over to the counter electrode using battery separators with a pore-diameter lower than the critical (smallest) size of the dendritic nucleate. To evaluate this proposal, we designed cross-linked polymer electrolytes with tunable pore size, and quantified the stability of metal electrodeposition. Direct visualization experiments were performed to understand the effect of pore-size on dendrite growth, which showed remarkable agreement with theoretical predictions. Furthermore, when operated in a battery, the crosslinked membrane showed the highest short circuit time compared to similar electrolytes reported in the literature.

Importantly, these studies showed that while the tendency for battery failure by dendrite-induced short-circuits can be reduced in polymer electrolytes, the issue of capacity-fading as a result of continuous reactions of the metal with a liquid electrolyte persists. Recently, there is an emerging school of thought that such reactions can be inhibited in supersaturated electrolyte solutions due to kinetic entrapment of solvent molecules by the crowded ionic species. A drawback of this approach is that it is costly, and it is limited by the solubility of salt in a solvent. In the second part of my talk, I will show how an artificially designed, solid-electrolyte interphase composed of anionic polymers provides a fundamental strategy for extending stability of metal electrodes even with chemically reactive liquid electrolytes. Complementing these experimental findings, I will show that a computational chemistry approach can be utilized to explain the mechanistic processes responsible for the extended stability.

BIOGRAPHY: Snehashis Choudhury received his Ph.D. from the Smith School of Chemical and Biomolecular Engineering at Cornell University in 2018, where he worked in the group of Prof. Lynden A. Archer. He earned his Bachelor of Technology from National Institute of Technology, India in 2013. His PhD research focused on three major themes: (1) understanding the structure and dynamics of self-suspended polymer-grafted nanoparticles, (2) designing nanostructured polymer electrolytes for inhibiting morphological instabilities in lithium metal batteries, and (3) designing solid-liquid interphases on reactive anodes to enhance surface diffusion of metal ions. This work lies at the intersection of chemical engineering science, materials physics, electrochemistry, and synthetic chemistry. His thesis, to date, has led to over 30 publications in leading journals of the field as well as multiple patents.