

Designing functional polymeric electrode/electrolyte interphases for high energy density lithium metal batteries

Sanjuna Stalin¹

1. Cornell University College of Engineering, Ithaca, New York, United States

Reactive metal anodes are known to electrodeposit in the form of irregular morphological features on planar substrates. Formed during the earliest stages of deposition, these features are thought to eventually result in non-planar, mossy structures that proliferate in the electrode spacing, hampering electrode reversibility. A growing body of work suggests that the mechanics, structure, ion transport properties, reductive stability, and interfacial energy of interphases formed spontaneously on the metal electrode play important, but separate roles in regulating nucleation, growth, and reversibility of these non-planar structures. In this study, we examine the effect of polymeric interphases on the early stages of lithium metal deposition and subsequent growth rate of the deposit front. By performing a theoretical linear stability analysis of metal electrodeposition across elastic interphases, we find that interphase thickness, mechanics, ion transport and interfacial properties all play precise and differentiated roles in setting the optimal interphase design. Motivated by this analysis, we focus specifically on thermosetting polymer interphases because their mechanical and chemical properties can be readily manipulated. Our goal is to develop design rules that can be implemented without adding substantially to the weight or volume of the metal electrode. By systematically varying the physical and chemical properties by manipulating monomer chemistry at the molecular level, we try to elucidate the effect of the properties of the polymer on the morphology of electrodeposited lithium. Using characterization techniques that include electroanalytical tools and operando visualization of the metal deposition, we find that parameters like polymer thickness, metal-polymer interfacial energy and elasticity have a profound effect on the morphology of electrodeposited lithium and correlate that with the theoretical results. These findings can potentially guide the design of artificial interphases as well as electrolyte components that lead to specific compositions of the SEI.

