Dynamical Phase Transitions on the Surface of Negative Electrodes – The Role of SEI

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Surface structures on negative electrodes are of paramount importance for the lifetime of lithium batteries. Growth of the solid-electrolyte interphase (SEI) is a major degradation mechanism for graphite, silicon and lithium metal electrodes through lithium/electrolyte consumption and capacity decay [1]. For next-generation electrodes like lithium metal, the growth of lithium whiskers during lithium deposition is an equally important limitation of battery cyclability [2]. In this talk, we will discuss the close chemo-mechanical coupling between SEI and lithium whiskers [3]. Fascinatingly, we observe dynamical instabilities in the SEI structure [4,5] as well as during dissolution of lithium whiskers [3].

The emergence of a dual-layer structure of SEI during its growth is important for understanding capacity fade and SEI mechanics. We discuss how the interplay of different electrolyte reduction reactions can lead to a compact inner SEI layer and a porous outer SEI layer [4]. This line of argument is the standard explanation for its emergent structure. We discuss an alternative, yet more fundamental explanation. When electron transport through the SEI slows down, kinetics of the reduction reaction favours a more heterogeneous growth of SEI and consequently leads to porous growth for the outer SEI [5]. We discuss predictions of such a dynamic instability.

Furthermore, we model the dissolution of nanoscale lithium whiskers [3]. Observations show that at very low rates the tip of whiskers does not dissolve and remains as dead lithium in the electrolyte. Our model explains the dynamic instability leading to this kind of dead lithium considering the impact of SEI on the whisker. In this way, we discuss an aspect of the intricate connection between SEI and lithium deposition.

References

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