When Electrochemistry Meets Mechanics: Impact of the SEI on Lithium Metal and Silicon Anodes

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Next-generation anode materials such as lithium metal and silicon promise substantial improvements in energy density, making them attractive for future high-performance lithium-ion batteries. However, both materials undergo very large volume changes during lithiation and delithiation, which generate significant stresses within the electrode and at the interface to the solid-electrolyte interphase (SEI). Consequently, understanding the coupled chemical and mechanical processes that occur during cycling is essential for accurately describing the behavior of these next-generation anodes and for designing strategies to improve their performance and stability.

In this work, we investigate the plating and stripping processes of lithium metal anodes using continuum simulations explicitly accounting for mechanical effects. Our model demonstrates that inhomogeneous stripping of lithium whiskers can lead to the formation of electrically isolated lithium, providing a physical explanation for the emergence of dead lithium [1]. During plating, the mechanical interplay of lithium metal and SEI causes stress-driven extrusion of lithium and the formation of lithium whiskers, highlighting the critical role of mechanical stresses in morphological evolution [2]. For silicon anodes, the large volumetric expansion and contraction considered in our core-shell model of active silicon surrounded by an inactive shell causes severe stresses inside the silicon anode. Due to the chemo-mechanical coupling, the emerging stresses impact the anode voltage, explaining the experimentally observed voltage hysteresis during cycling and after relaxation periods [3]. In addition, our model captures the logarithmic voltage relaxation that occurs over extended timescales, often persisting for weeks after cycling, which arises from slow relaxation of the viscous stress contribution [4]. Moreover, we have investigated the degradation of silicon-graphite composite anodes driven by conventional SEI growth and several aspects arising from the mechanical degradation of silicon.

These results emphasize that the chemo-mechanical interplay is a key factor governing the behavior of next-generation anode materials. Incorporating mechanical effects into electrochemical models enables a more comprehensive understanding of degradation mechanisms and voltage response in lithium metal and silicon electrodes. Our model-based insights can guide the development of strategies to mitigate the major drawbacks of these materials, thereby helping to unlock their full potential for high-energy-density battery technologies.

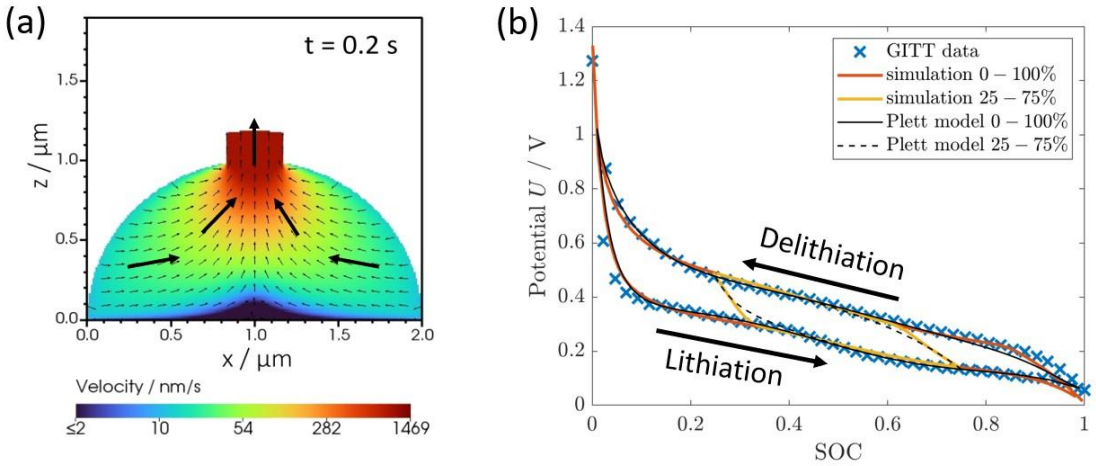

Fig1. (a) Lithium velocity profile during plating and whisker formation [2]. (b) Voltage hysteresis of silicon anode [3]. Reprinted and modified from stated reference, licensed under CC BY 4.0, https://creativecommons.org/licenses/by/4.0/.

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