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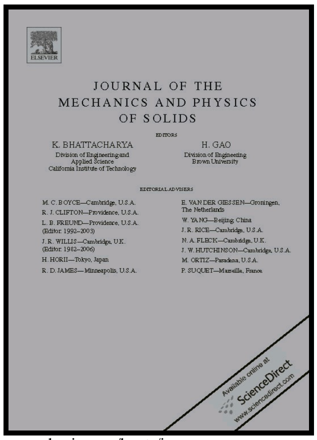

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Intrinsic Stress Mitigation via Elastic Softening during Two-Step Electrochemical Lithiation of Amorphous Silicon

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Abstract

Recent experiments and first-principles calculations show the two-step lithiation of amorphous silicon (a-Si). In the first step, the lithiation progresses by the movement of a sharp phase boundary between a pristine a-Si phase and an intermediate $Li_\eta Si$ phase until the a-Si phase is fully consumed. Then the second step sets in without a visible interface, with the $Li_\eta Si$ phase continuously lithiating to a $Li_{3.75}Si$ phase. This unique feature of lithiation is believed to have important consequences for mechanical durability of a-Si anode in lithium ion batteries, however the mechanistic understanding of such consequences is still elusive so far. Here, we reveal an intrinsic stress mitigation mechanism due to elastic softening during two-step lithiation of a-Si, via chemo-mechanical modeling. We find that lithiation-induced elastic softening of a-Si leads to effective stress mitigation in the second step of lithiation. These mechanistic findings allow for the first time to quantitatively predict the critical size of an a-Si anode below which the anode becomes immune to lithiation-induced fracture, which is in good agreement with experimental observations. Further studies on lithiation kinetics suggest that the two-step lithiation also results in a lower stress-induced energy barrier for lithiation. The present study reveals the physical underpinnings of previously unexplained favorable lithiation kinetics and fracture behavior of a-Si anodes, and thus sheds light on quantitative design guidelines toward high performance anodes for lithium ion batteries.

Keywords: Amorphous silicon; Two-step lithiation; Chemo-mechanical modeling; Fracture; Lithium-ion battery; Anode

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