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Controlling conversion reactions with multilayer electrodes

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Battery materials used for conversion reactions are capable of much higher charge capacities than intercalation compounds, but suffer from slow kinetics and irreversibility stemming from diffusion barriers, trapped lithium phases, and agglomeration. All of these problems stem from the complex phase separation/nucleation required to reversibly lithiate and, moreover, delithiate such a material. To study these reactions, we have grown amorphous silicon/chromium multilayers with atomically smooth interfaces. Using synchrotron-based x-ray reflectivity, we are able to watch the lithiation and delithiation of such a multilayer in real-time. Following the multilayer Bragg peaks, we find that the entire structure expands and contracts by over 300% during lithiation. Remarkably, the layering remains intact through the reaction. Drawing from this reversible structural transition, we have tested the rate-capabilities and cycling performance of multilayers in coin cells. Depending on the number of layers and their thicknesses, we find that multilayers can achieve theoretical capacities at slow cycling and can hold half their initial capacity after 1500 cycles at 5C, with an average Coulombic efficiency of 99.7%. Silicon thin films with similar overall thickness lose all capacity by cycle 300. Finally, we will discuss routes toward scaling up silicon-based multilayers to thicknesses well over 1 µm and initial results on germanium and sulfur based heterostructures.

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