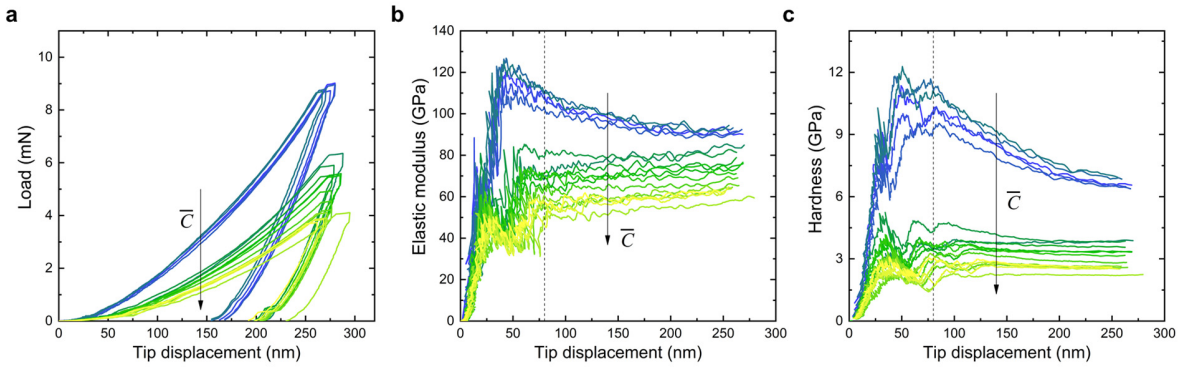

Figure S1. Raw nanoindentation data corresponding to the indentation array in Figure 3a. (a)-(c) shows the load-displacement curves, elastic modulus, and hardness as a function of the tip displacement, respectively. The dashed lines in (b) and (c) indicate the depth of 80 nm used for the extraction of the mechanical properties. The color gradient from blue to yellow indicates the increasing Li concentration.

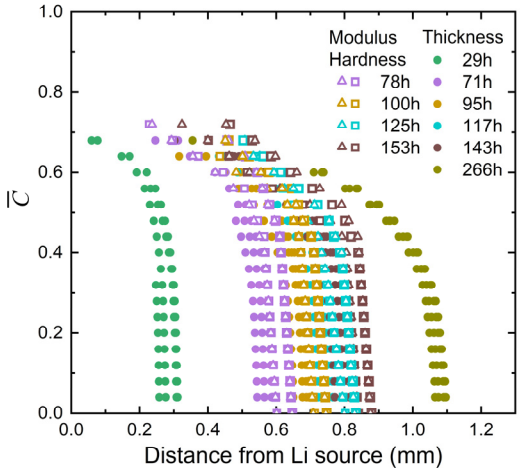

Figure S2. Li concentration profiles determined from the elastic modulus measurements (empty triangles), hardness measurements (empty squares), and film thickness measurements (solid circles).

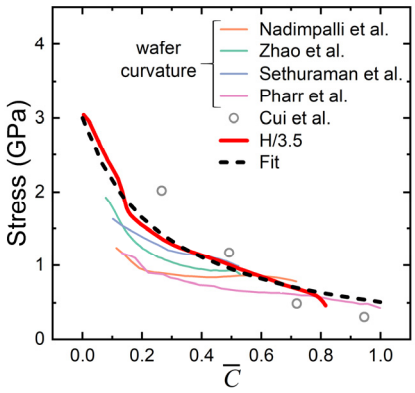

Figure S3. Yield stress of lithiated Si in literature including the nanoindentation hardness measurement (de Vasconcelos et al., 2017), wafer curvature measurements (Nadimpalli et al., 2013; Pharr et al., 2013; Sethuraman et al., 2010; Zhao et al., 2012), and DFT calculations (Cui et al., 2012b).

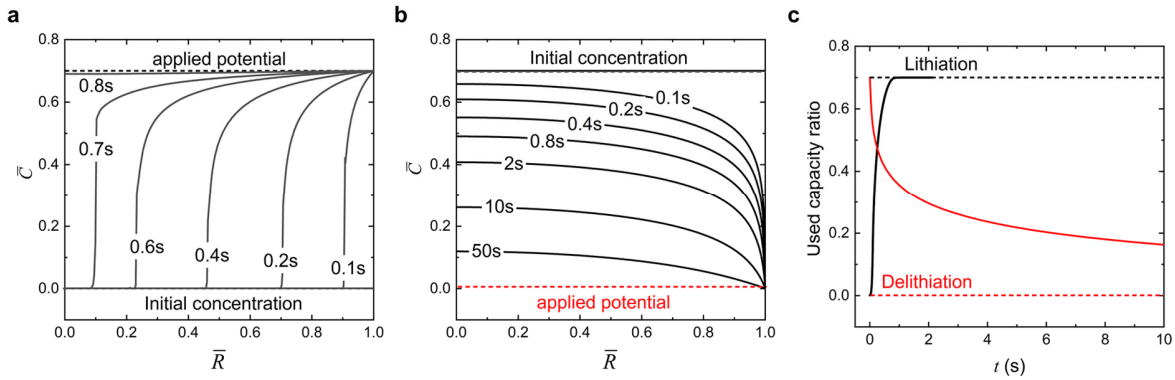

Figure S4. Lithiation and delithiation of a Si particle without considering the stress effect on Li diffusion. (a) Propagation of a sharp reaction front in Si during lithiation, (b) a rather smooth Li concentration gradient during delithiation, and (c) the asymmetric charging (lithiation) and discharging (delithiation) capacities of the Si particle undergoing the potentiostatic cycle.

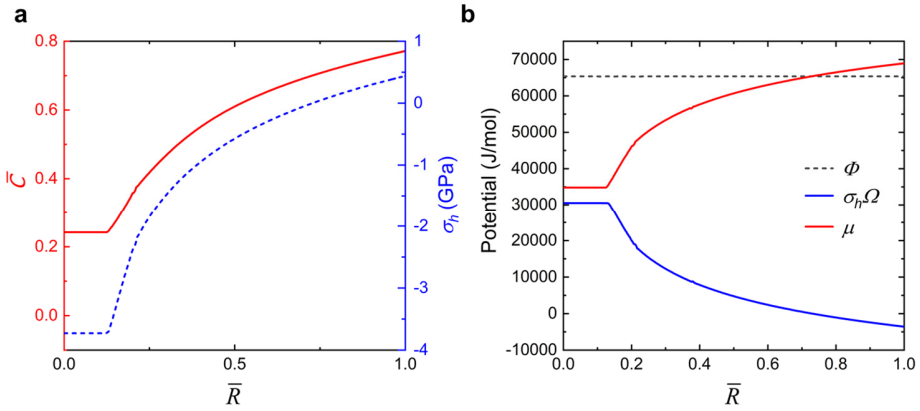

Figure S5. (a) Equilibrium profiles of Li distribution and hydrostatic stress in the Si particle at the end of the potentiostatic charging. (b) Plots of the diffusion potential (dashed line) and its mechanical (blue solid line), and chemical (red dolis line) components across the radius of the Si particle.