Supplementary Information



Fig. S1. (a) The evolution of indentation depth during the hold period at peak load. (b) The stress exponents of lithiated Si obtained from the slope of the stress vs strain-rate responses.



Fig. S2. Current and voltage profiles during the first lithiation of Si and the corresponding indentation measurements. The electrical current alternates between zero and galvanostatic discharge at a rate C/10.6 until the cut-off voltage is reached. The drift-rate peaks at the initial application of the current and rapidly converges to a nearly constant value. Additional drift introduced by the environment (circled in orange, early morning in the building) does not compromise the measurements. The elastic modulus and hardness are approximately constant at open circuit (black circle) and decrease during discharge (blue circle). Obvious outliers (black cross) are removed and a settling time of roughly 20 min is granted once the circuit is opened (black cross).



Fig. S3. (a). The load-displacement curves for indentation of Li metal at different depths. (b) Elastic modulus and hardness of Li metal. The large variation in the results is possibly due to the surface roughness, Li creep, and material pile-up around the indentation tip. The inset optical image shows a representative residual impression.



Fig. S4. The load-displacement curves of lithiated Si of different Li compositions.