Supplementary Materials

Organic Solvent Free Process to Fabricate High Performance Silicon/Graphite Composite Anode

Chen Fang [1,†], Haiqing Xiao [1,2,†], Tianyue Zheng [1,3,*], Hua Bai [2,*] and Gao Liu [1,*]

[1] Energy Storage and Distributed Resources Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Road, Berkeley, CA 94720, USA; cfang@lbl.gov (C.F.); xiaohaiqing@hotmail.com (H.X.)

[2] Institute of Industrial and Consumer Products Safety, Chinese Academy of Inspection and Quarantine (CAIQ), No.11 Ronghua South Road, Daxing District, Beijing, 100176, China

[3] Ningbo Institute of Materials Technology and Engineering, Chinese Academy of Sciences, 1219 Zhongguan West Road, Ningbo, Zhejiang 315201, China

* Correspondence: tyzheng@nimte.ac.cn (T.Z.); baih111@sina.com (H.B.); gliu@lbl.gov (G.L.)

† These authors contributed equally to this work.

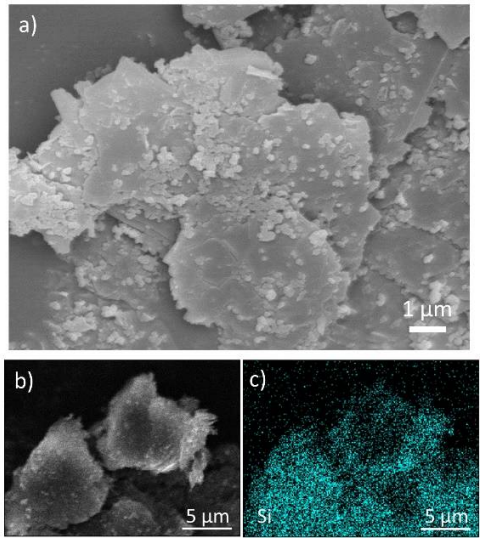

Figure S1. Characterization of silicon/graphite composite after solid mixing a) SEM image and b-c) EDX mapping.

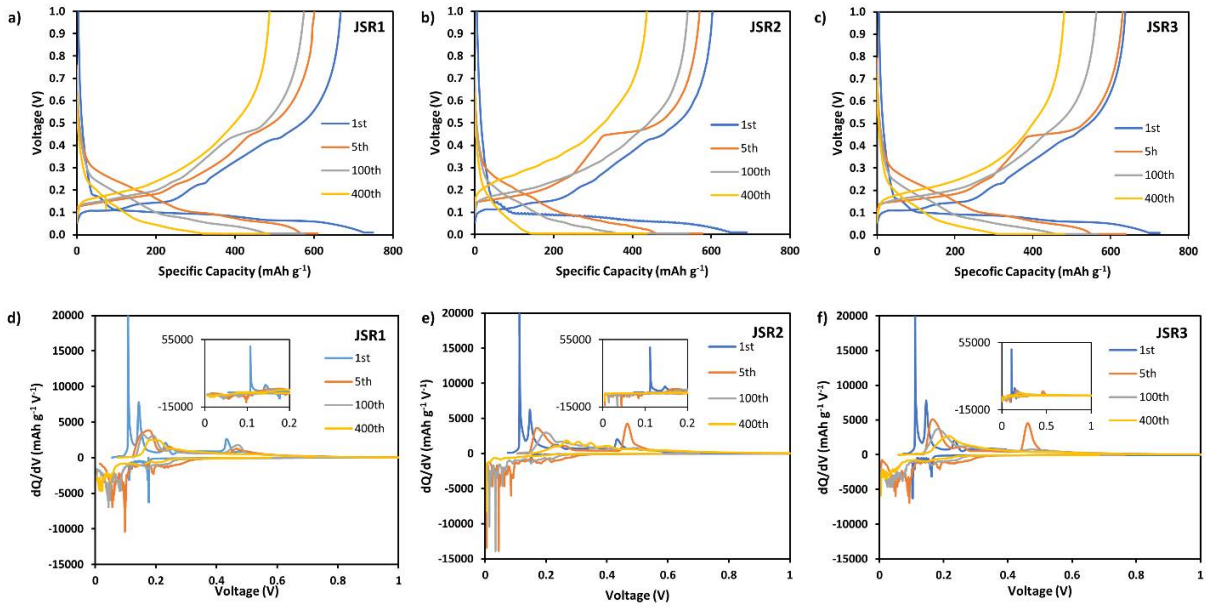

Figure S2. Differential capacity (d*Q*/d*V*) curves and charge/discharge curves of electrodes prepared with JSR1, JSR2 JSR3. (399th cycle is plotted for JSR3 as the 400th cycle because the 400th cycle was interrupted.)

Extended discussion of Figure S2. The charge/discharge curves demonstrated similar features from the initial cycles to the 400th cycles, demonstrating the stability of the composite electrodes. These results are consistent with the stable cycling performance observed. The detailed electrochemical properties of the composite electrodes could be further revealed by dQ/dV profiles. To more accurately reflect the dQ/dV profile throughout the entire cycling process, the 5th cycle of JSR1 electrode is used as an example for discussion (Fig. S2d). Lithiation of the graphite at the 5th cycle was observed with three lithiation plateaus at 0.2, 0.1 and 0.06 V vs Li/Li$^+$. The lithiation peak for silicon at 0.1 V vs Li/Li$^+$ is presumably overlapped with the graphite peaks, and a broad peak at about 0.24 V vs Li/Li$^+$ was observed. As for delithiation, two peaks are observed for graphite at 0.16 and 0.24 V vs Li/Li$^+$ while a 0.46 V vs Li/Li$^+$ peak was observed for silicon. The shifting and broadening of the graphite peaks in both lithiation and delithiation stages indicate the gradual increase of overpotential, likely due to the impedance growth related to repeated electrolyte decomposition on silicon materials. The silicon delithiation peak gradually disappeared in later cycles, which supports the argument above. These results are consistent with the observed capacity decay of the electrode samples.