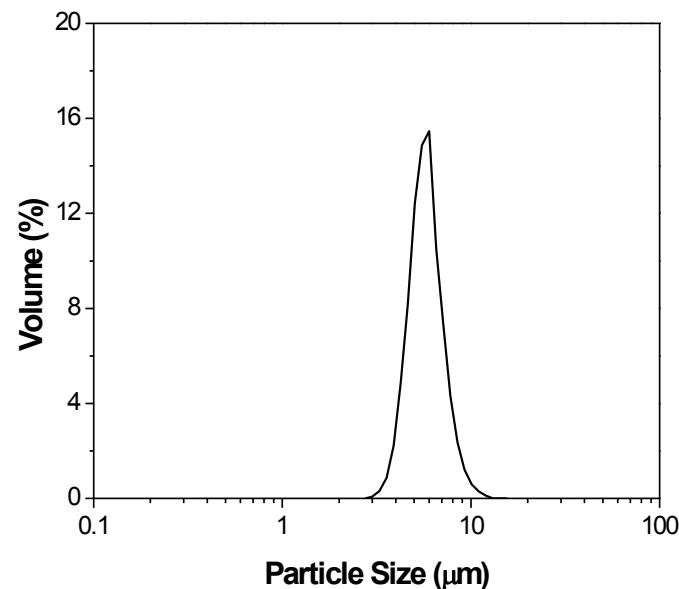


**Supplementary Information**

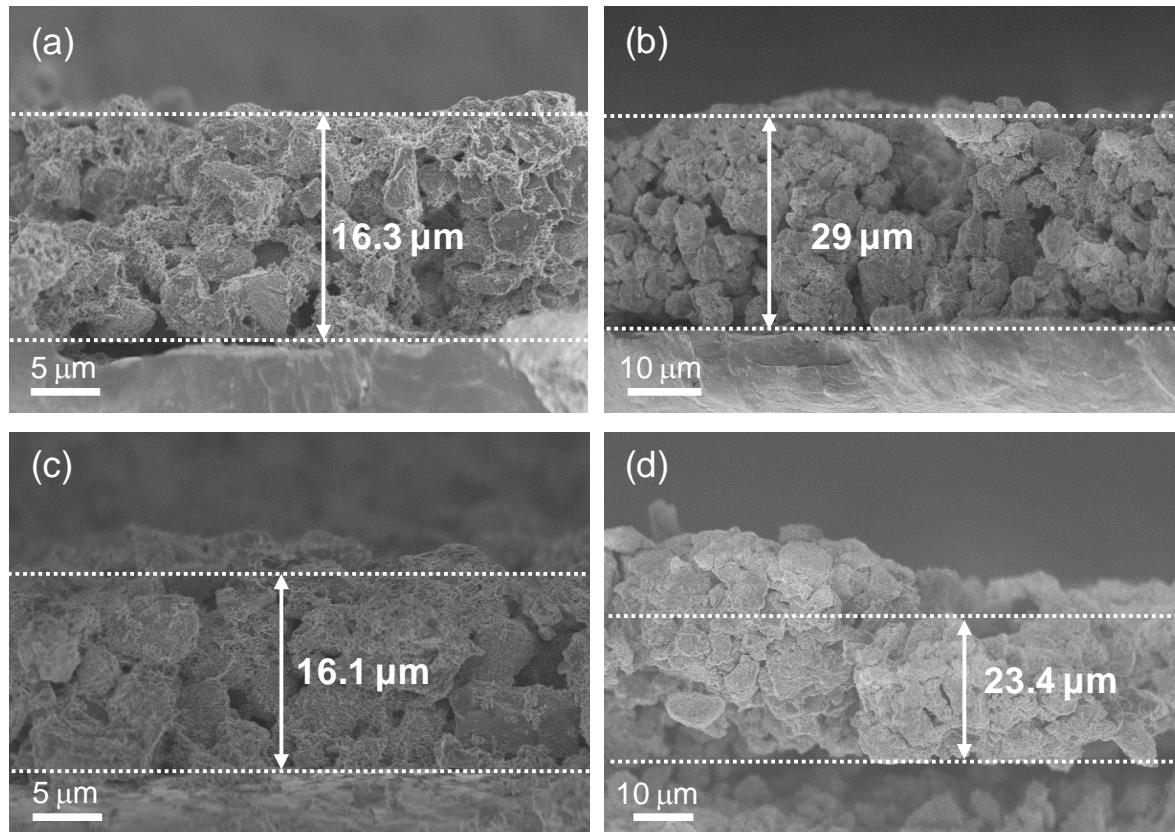
**Surface Engineering of Sponge-like Silicon Particles for High-Performance  
Lithium-Ion Battery Anodes**

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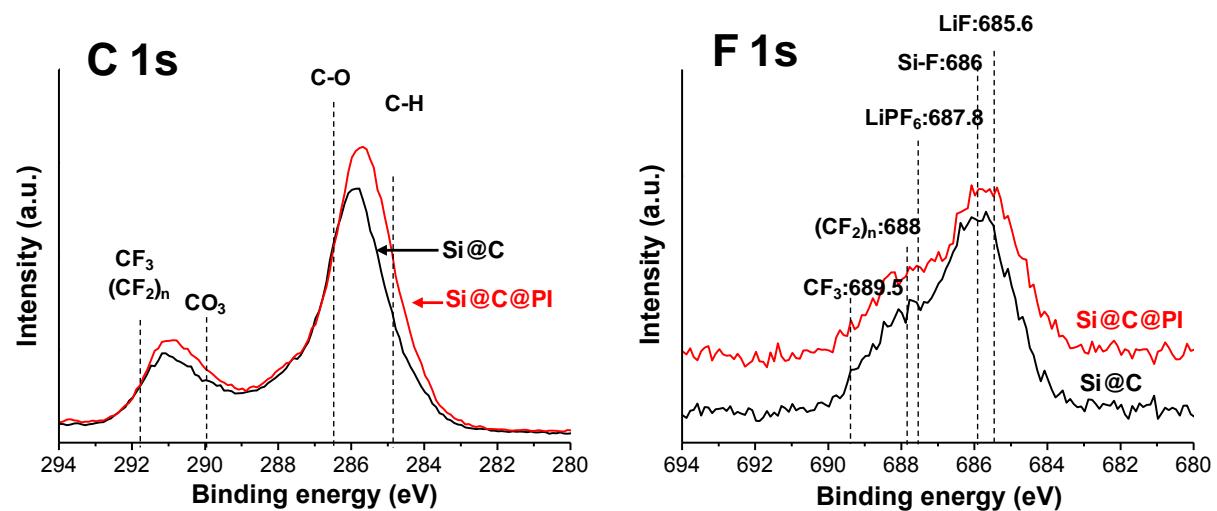


**Figure S1.** Paricle size distribution of sponge-like Si particles after chemical etching. After etching, an average particle size of the Si is ~6  $\mu\text{m}$ .

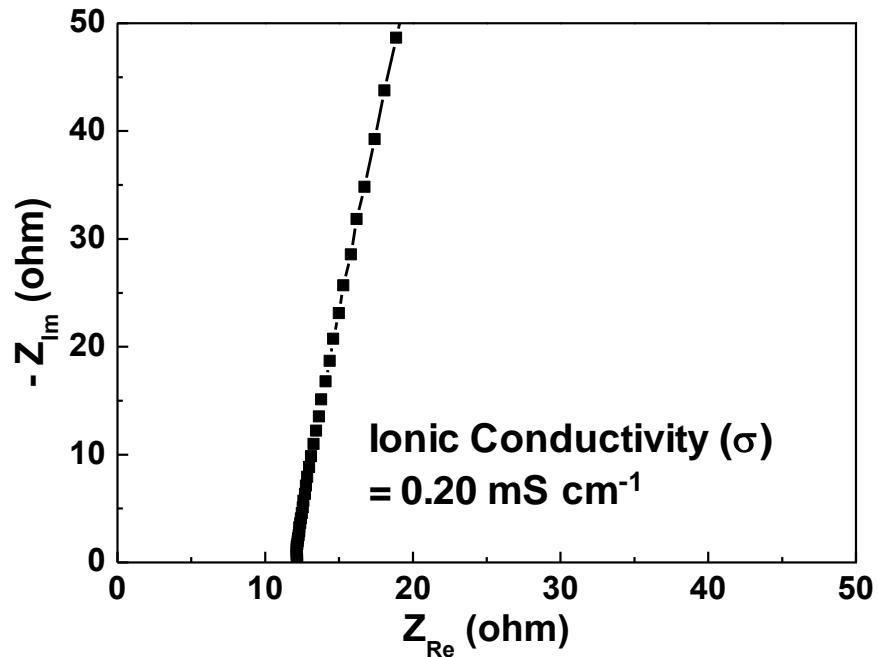


**Figure S2.** SEM images showing volume expansion of electrodes before and after cycling.

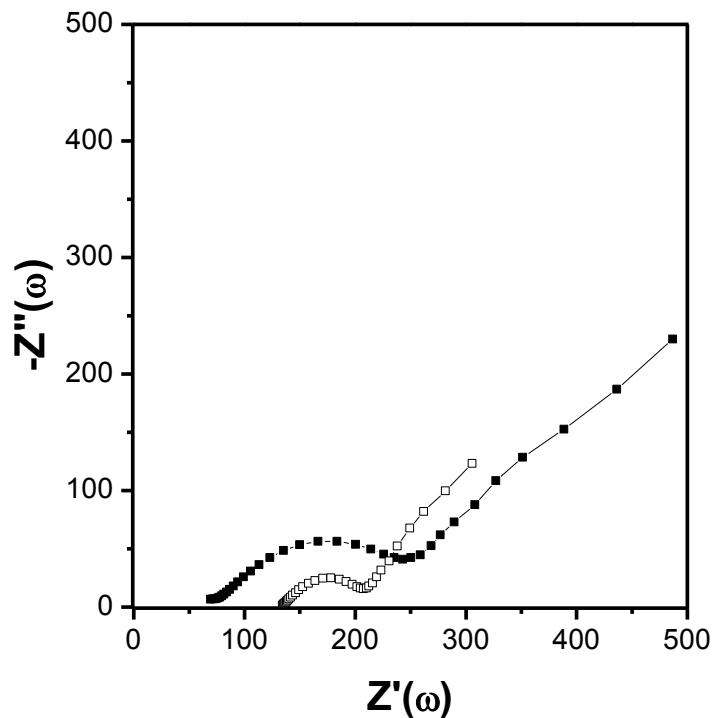
Thicknesses of Si@C (A: before, B: after cycle) and Si@C@PI (C: before, D: after cycle) electrodes were investigated by cross-sectional SEM. The Si@C electrode showed volume expansion of 78%, while the Si@C@PI exhibited less volume expansion of 45% after 80 cycles.



**Figure S3.** XPS spectra of C 1s and F 1s core level obtained from the Si@C (black) and Si@C@PI (red) electrodes after 30 cycles. The peaks of Li<sub>2</sub>CO<sub>3</sub> and ROCO<sub>2</sub>Li at 291 eV, the peak at 136 eV arising from Li<sub>x</sub>POF<sub>y</sub> and Li<sub>x</sub>PF<sub>y</sub>, and the LiF peak at 687.2 eV are typically seen in carbon-coated Si electrodes.



**Figure S4.** Ionic conductivity of PI films swelled with the liquid electrolyte (1.3 M LiPF<sub>6</sub> in EC/DEC = 30/70 v/v). The liquid electrolyte-swollen PI film was found to provide a high ionic conductivity of 0.20 mS cm<sup>-1</sup> at room temperature.



**Figure S5.** AC impedance spectra (after 80 cycles) of cells assembled with Si@C (solid square) and Si@C@PI (open square).