

Supporting Information

Carbon-coating strengthens solid electrolyte interphase to inhibit Si pulverization

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1.Coating characterizations

As shown in the HRTEM and EDS images (Fig. S1a-b), Si NPs are uniformly coated by nanocrystalline carbon, and the carbon coating layer is about 10 nm thick (see Fig. S1c).

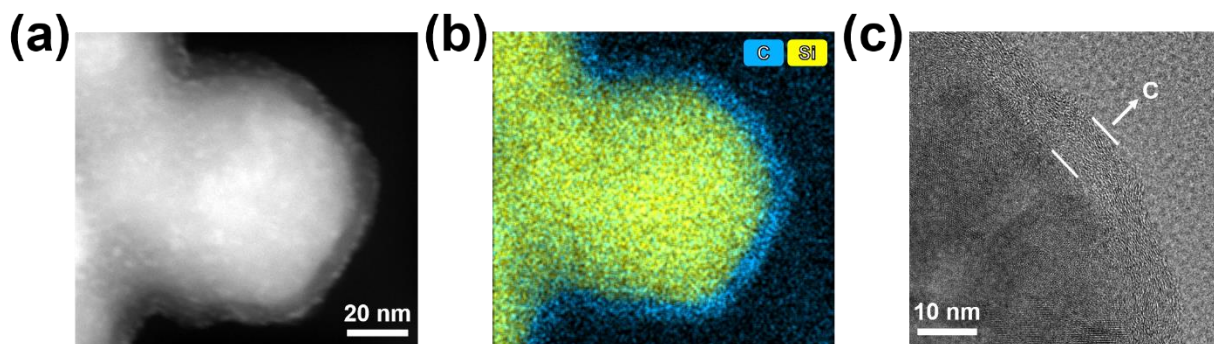


Fig. S1 HRTEM (a) and EDS (b) images of pristine Si@C NP; (c) lattice diagram of the nanocrystalline carbon coating layer.

The Raman spectra (Fig. S2) illustrates a carbon coating layer on the Si NPs.

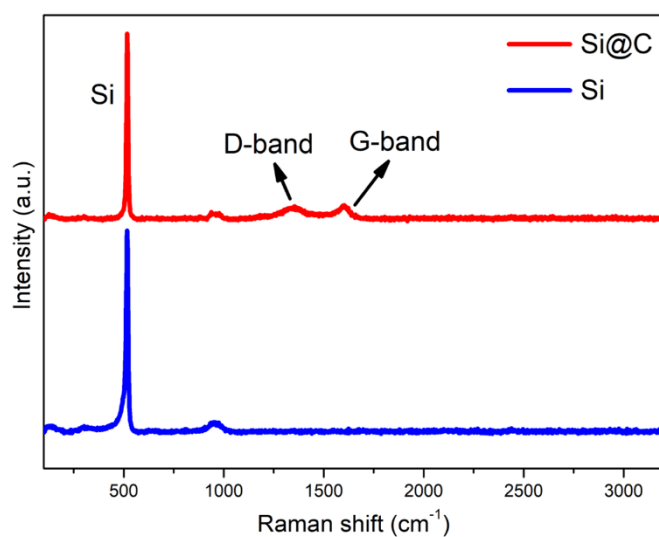


Fig. S2 Raman spectra of bare Si NPs and Si@C NPs.

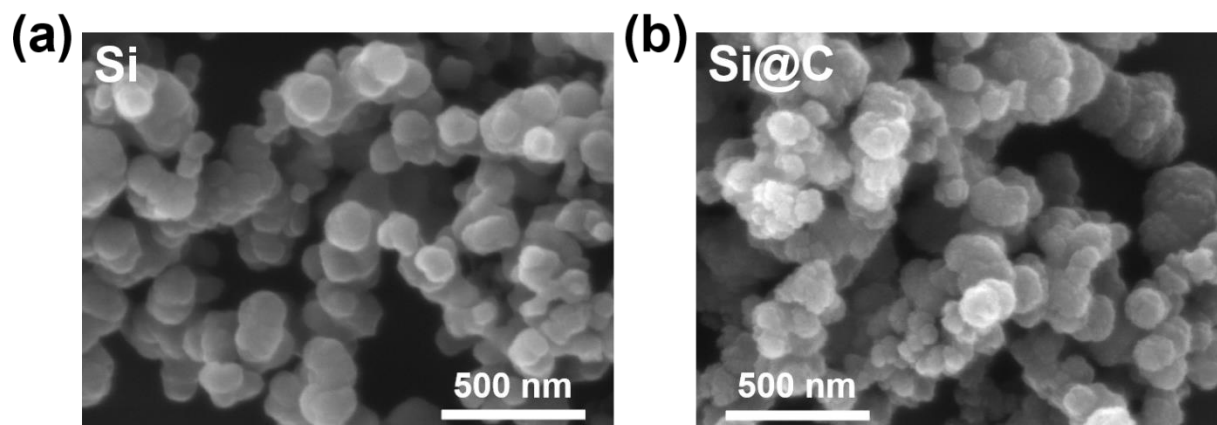


Fig. S3. SEM images of bare Si NPs (a) and Si@C NPs (b).

2. Morphology of bare Si and Si@C electrodes before cycling

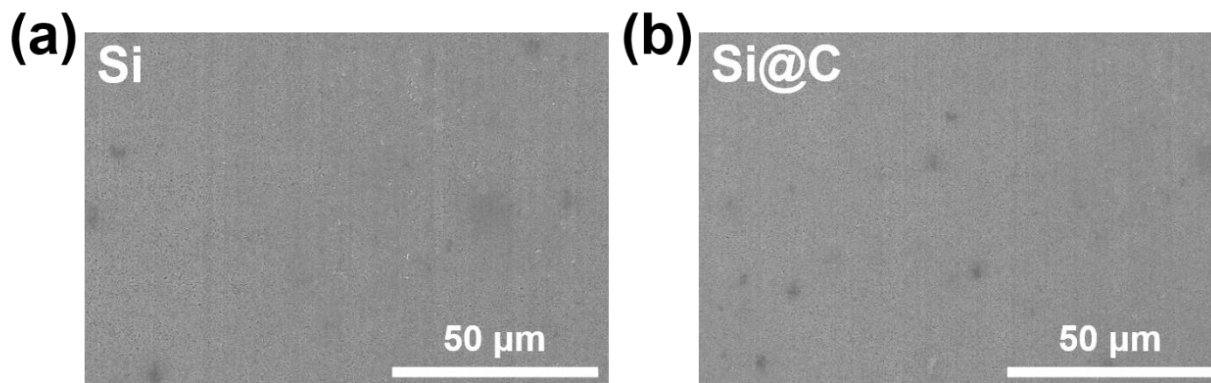


Fig. S4 SEM morphology of the bare Si (a) and Si@C (b) electrodes before cycling.

3. Crossing-section morphology of bare Si and Si@C electrodes after 100 cycles

After 100 cycles, according to the crossing-section morphology images of the bare Si and Si@C electrodes (Fig. S5a-b), the Si particles cannot be clearly distinguished, which means a universal pulverization of Si particles. While for the Si@C particles, their profiles are still can be recognized, indicating the integrity of Si@C particles is well-kept.

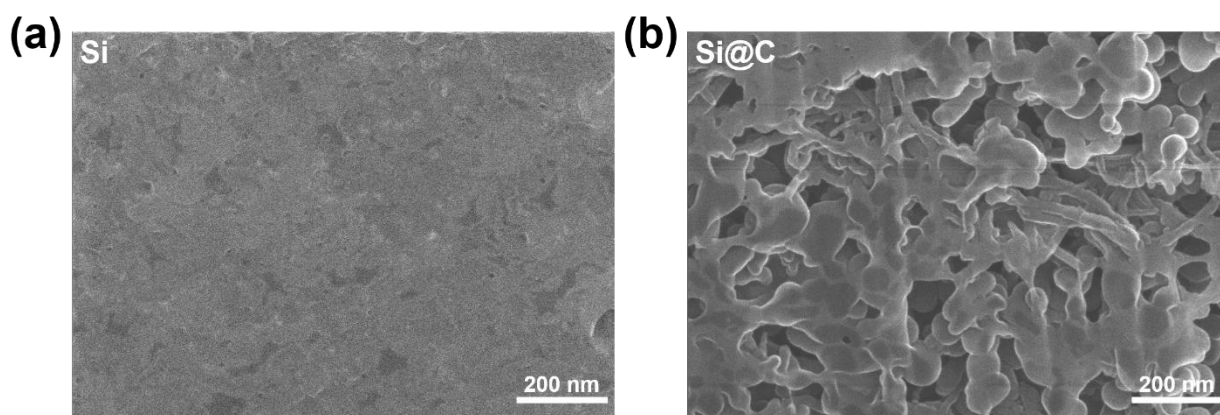


Fig. S5 Crossing-section morphology of the bare Si (a) and Si@C (b) electrodes after 100 cycles.

4.AFM topography

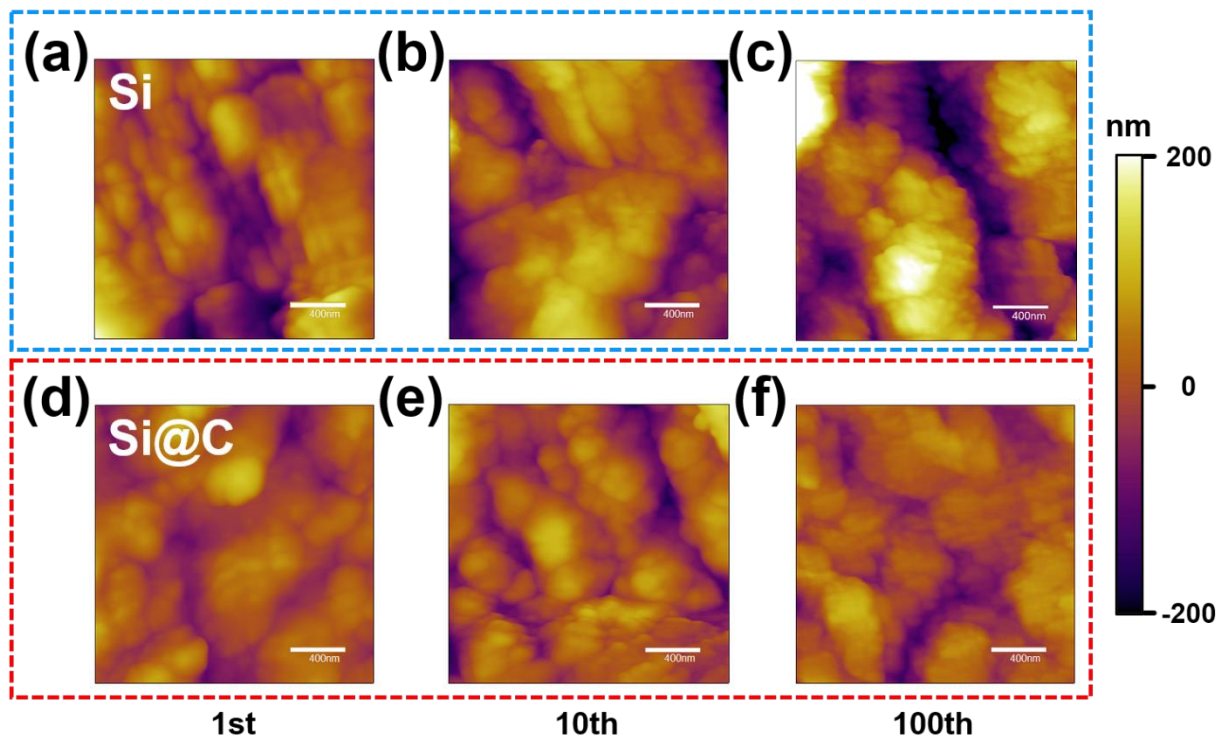


Fig. S6 AFM topography images of the bare Si (a-c) and Si@C (d-f) anodes after the 1st, 10th and 100th cycle.

5. CV curves

Three peaks around 1.63 V, 1.43 V and 1.35 V are observed in the first cathodic scan of the Si@C electrode, and then disappear in the following scans, which can be attributed to the formation of SEI due to the decomposition of electrolyte additive (FEC) and solvent (EC and DMC), respectively.^{1,2} Compared with the Si@C electrode, the reduction peaks of the bare Si anode shift to the lower potential, meaning the electrode undergoes larger potential hysteresis owing to the relatively low electronic conductivity. The lower peak current of the Si@C electrode indicates that carbon-coating facilitates the formation of SEI, and effectively reduces electrolyte decomposition.

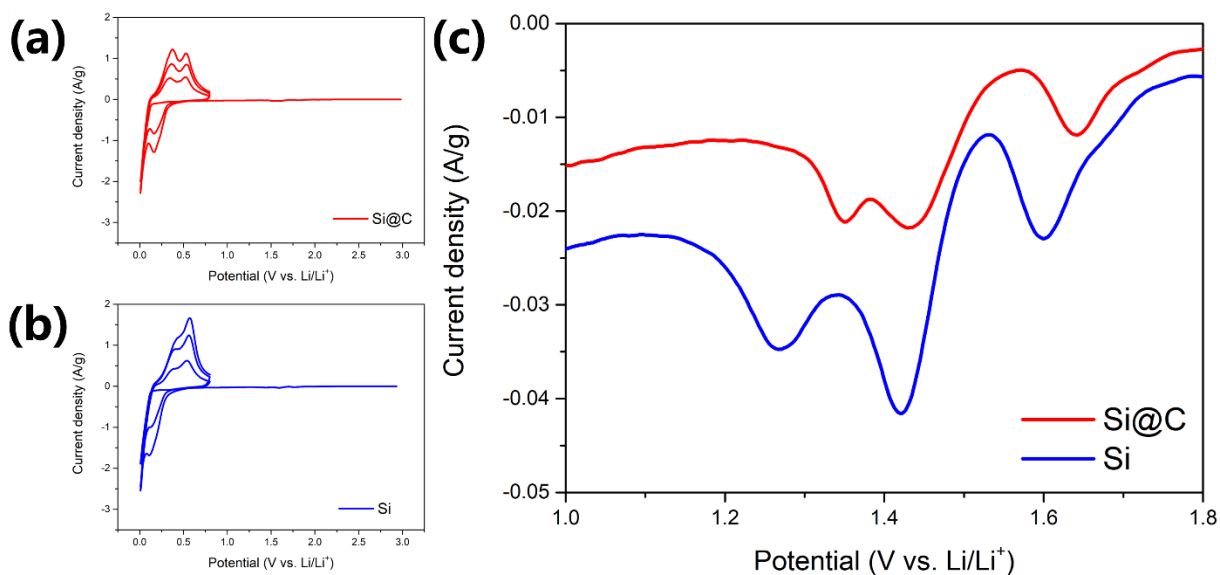


Fig. S7 CV curves of the Si@C (a) and bare Si (b) electrodes at a scan rate of 0.1 mV s^{-1} , (c) Enlarged curves of the selected potential regions in (a) and (b).

6. The deconvoluted Si 2p spectra and detailed C 1s spectra for the bare Si and the Si@C electrodes after 100 cycles

The supplementary Si 2p spectra of the bare Si and the Si@C electrodes after 100 cycles are shown in Fig. S8, in which the Si 2p spectra are deconvoluted into $2p_{1/2}$ and $2p_{3/2}$.³

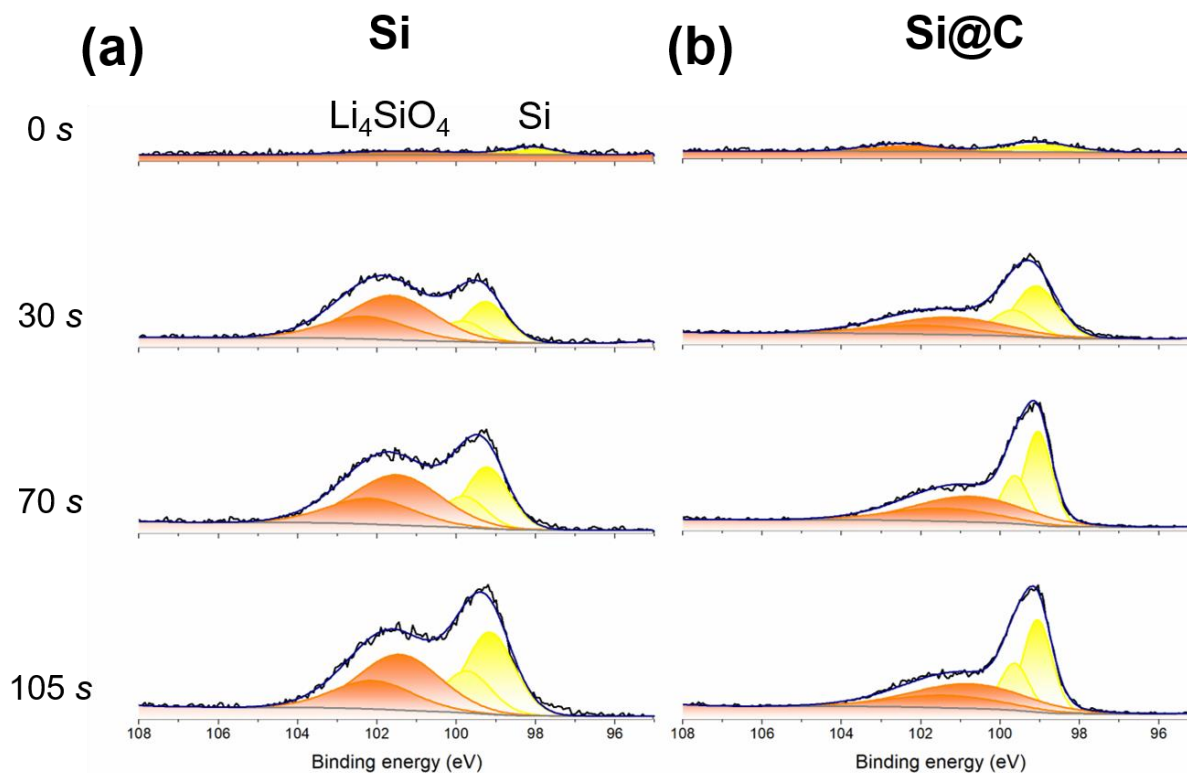


Fig. S8 The deconvoluted Si 2p spectra for the bare Si (a) and the Si@C (b) electrodes after 100 cycles.

The detailed C 1s spectra of the bare Si and the Si@C electrodes after 100 cycles are shown in Fig. S9a-b.⁴⁻⁶ The C-O peak in the C 1s XPS spectra indicates the presence of organic compounds with oxygen-containing groups in the SEI. Besides, a new peak located at around 288 eV is revealed, which can be ascribed to RCH₂OLi.^{5,6} As shown in the C 1s XPS spectra, the SEI on the Si@C electrode contains more RCH₂OLi than the SEI on the bare Si electrode.

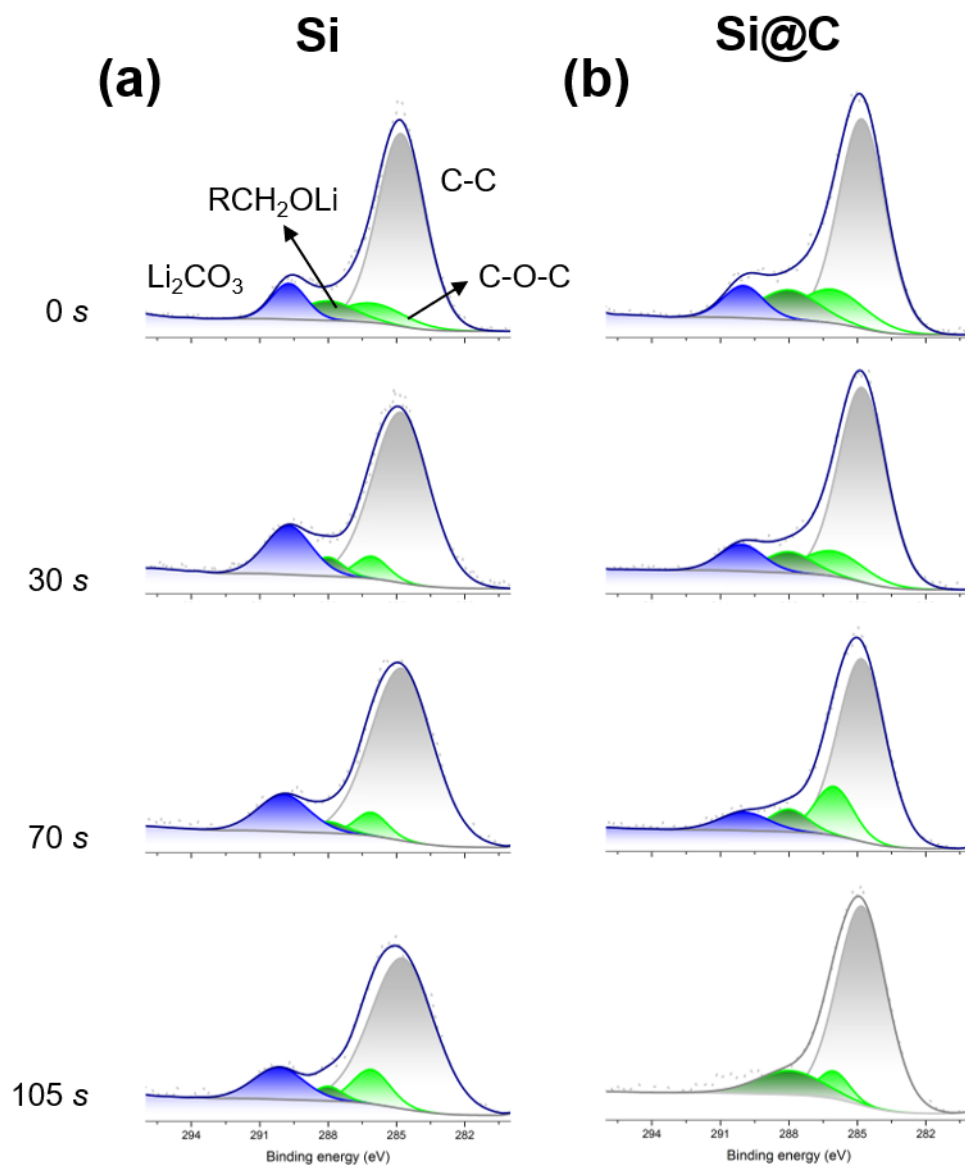


Fig. S9 The detailed C 1s spectra for the bare Si (a) and the Si@C (b) electrodes after 100 cycles.

7.XPS full spectra of the SEI on the Si electrode before etching and after etching for 30 s

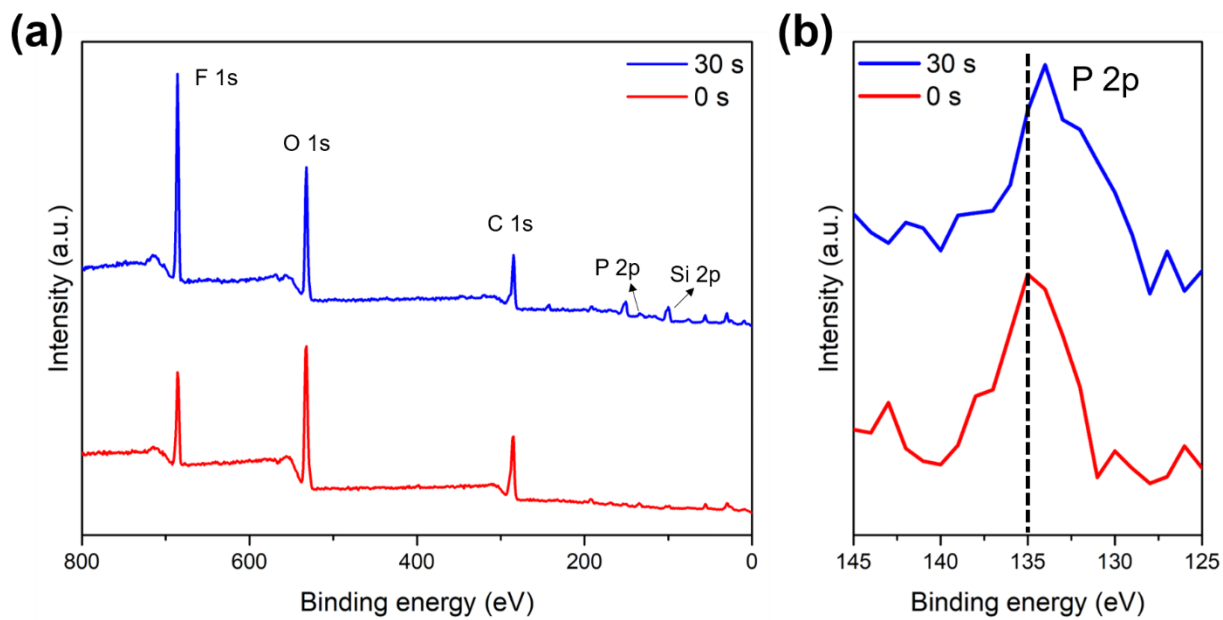


Fig. S10 (a) XPS full spectra and (b) selected region (P 2p peak) of the SEI on the Si electrode before etching and after etching for 30 s.

7. The values of R_s , R_{SEI} , and R_{ct} for the bare Si and the Si@C electrodes after different cycles

Table S1: The values of R_s , R_{SEI} , and R_{ct} for the Si and Si@C electrodes after different cycles

	Cycle number	R_s (Ω)	R_{SEI} (Ω)	R_{ct} (Ω)
Si	1st	9.3	82.0	77.0
	10th	3.4	138.7	20.7
	100th	7.6	238.1	90.7
Si@C	1st	3.3	22.6	66.0
	10th	6.4	52.6	56.1
	100th	7.3	80.5	49.0

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