

Supporting Information

B- and F-Enriched Buffering Interphase Enables High-Rate and High-Stability SiO_x/C Anode

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Experimental Section

Materials Synthesis

The SiO_x/C@LFB composites were obtained by a simple solution mixing method with heat treatment. First, lithium difluoro-oxalate borate (LiDFOB, $\geq 99\%$, Aladdin) was dissolved in N,N-dimethylformamide (DMF, AR, 99.5%, Macklin) to form a 0.1 mol/L clear solution, and then added SiO_x/C material with a 5:1 mass ratio to LiDFOB. After mixing and stirring for 24 h, drying the DMF solvent in a vacuum drying oven, to obtain the composite material with tight adsorption of LiDFOB on the SiO_x/C surface (denoted as SiO_x/C+LiDFOB). Secondly, the SiO_x/C+LiDFOB was sent into a tube furnace so that it was subjected to an annealing process (400 °C, 3 h) under argon atmosphere to obtain the SiO_x/C@LFB composite. Thirdly, SiO_x/C@LFB, carbon black and sodium carboxymethyl cellulose (CMC) were mixed well in the ratio of 8:1:1 and formulated into a viscous slurry. Then, the slurry was cast onto copper foil and vacuum-dried at 80 °C for 12 h. After drying, a punching machine was used to prepare copper foils with a diameter of 12 mm. In order to reduce the experimental variables, SiO_x/C materials without added lithium salts and undergoing the same annealing process were used as the main active materials for the comparison electrodes. The preparation process of SiO_x/C-based electrodes in the universality validation experiment is consistent with the above, and only LiDFOB needs to be replaced with the corresponding lithium salts including LiOTF, LiTFSI and LiBOB. Note that the mass loadings of the anodes were set between 1.0 and 1.1 mg cm⁻².

Material Characterizations

The results of thermogravimetric analysis (TGA) were obtained by a simultaneous thermogravimetric analyzer (STA449F5, Netzsch, Germany) from 25 to 600 °C at 5 °C/min heating rate (nitrogen atmosphere). SEM images were taken by field emission scanning electron microscope (SU8220, Hitach, Japan). The results of TEM-EDS element mapping images were all obtained by field emission transmission electron microscope (Talos F200S, FEI, Thermo, Czech). XPS spectra were generated using X-

ray photoelectron spectroscopy (Escalab 250Xi, Thermo Fisher, UK). XRD patterns obtained by X-ray diffractometer (D8 VENTURE, Bruker, Germany).

Electrochemical Measurements

All batteries were tested with CR2016 coin cells, in which half cells are electrochemically measured in the 0.01-2 V voltage range. The half-cell was assembled with $\text{SiO}_x/\text{C}@\text{LFB}$ as the working electrode and lithium foil as the counter electrode, and the electrolyte was 1 M LiPF_6 dissolved in a volume ratio of 1:1 mixture of ethylene carbonate (EC) and diethyl carbonate (DEC) with an additional 5 wt% fluoroethylene carbonate (FEC). The amount of the electrolyte was 100 μL and Celgard 2400 microporous membrane was used as the separator. All charge-discharge tests were done using a Neware battery cycler (Shenzhen, China). To obtain the CV curves and EIS results, the Gamry electrochemical workstation was used for testing, where the EIS measurement frequency range was set from 100 kHz to 0.01 Hz. All cells were assembled in a glove box filled with Ar ($\text{H}_2\text{O}<0.5$ ppm, $\text{O}_2<0.5$ ppm).

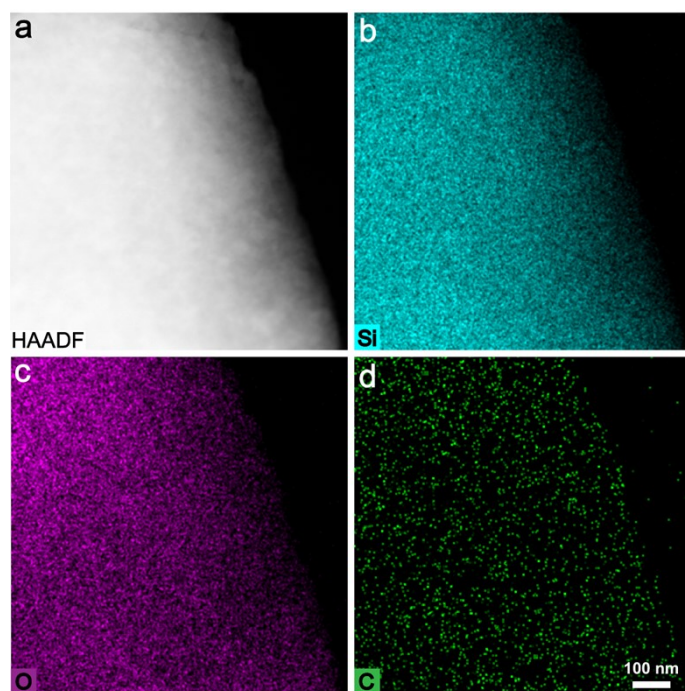


Fig. S1 (a-d) TEM-EDS element mapping images of Si, O, and C for SiO_x/C .

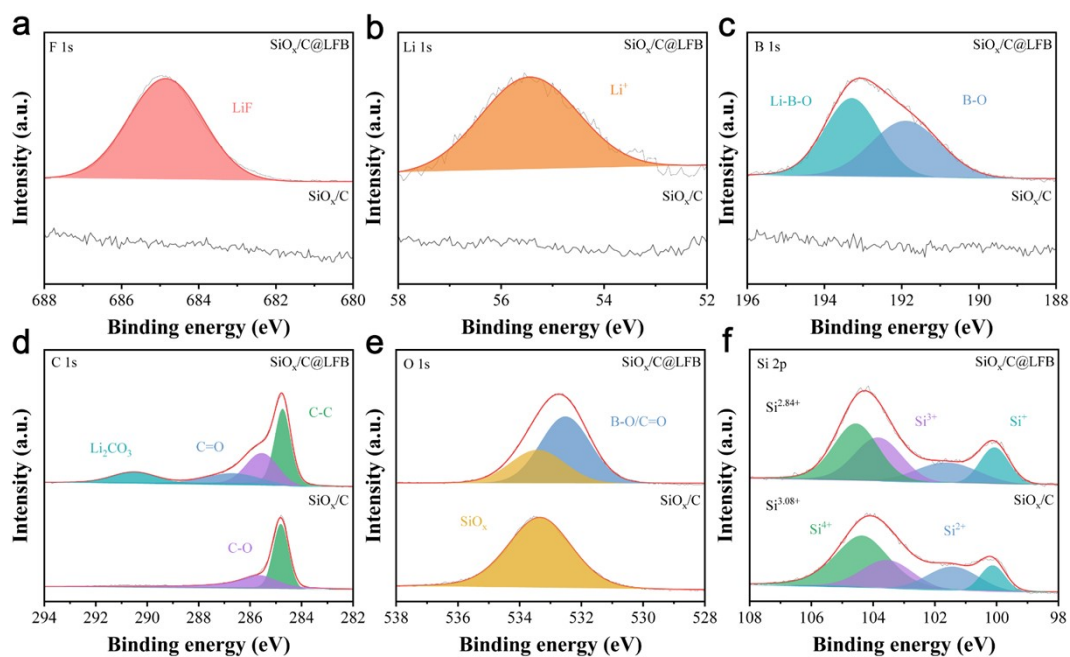


Fig. S2 (a-f) XPS spectra analyses of SiO_x/C and $\text{SiO}_x/\text{C}@LFB$ for F 1s, Li 1s, B 1s, C 1s, O 1s, and Si 2P.

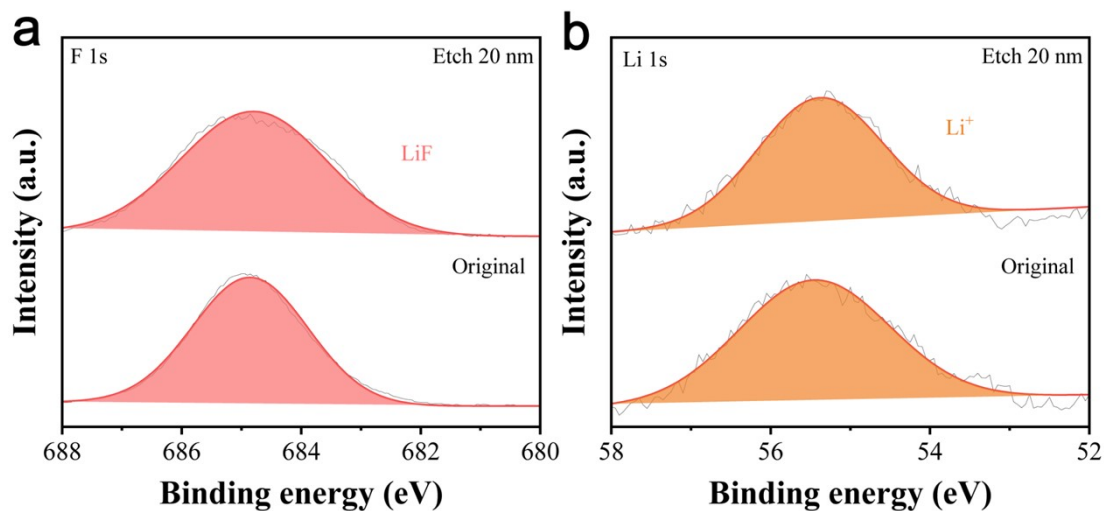


Fig. S3 XPS spectra analyses of pristine and etched 20 nm $\text{SiO}_x/\text{C}@L\text{FB}$ electrode for (a) F 1s and (b) Li 1s.

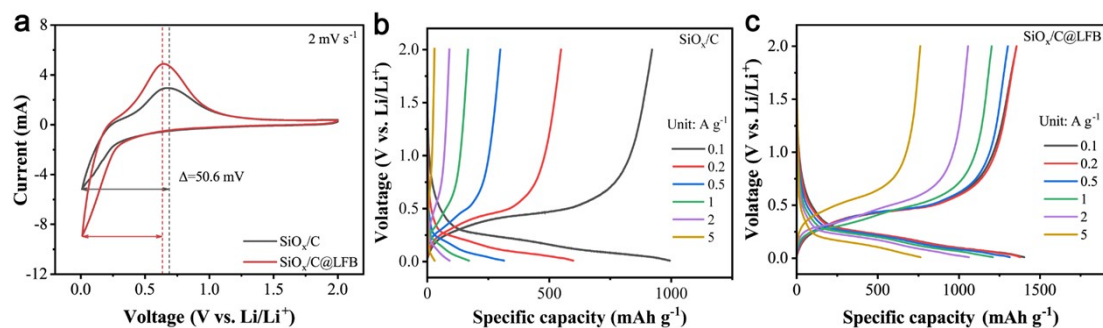


Fig. S4 (a) The CV curves of SiO_x/C and $\text{SiO}_x/\text{C}@L\text{FB}$ electrodes at 2 mV s^{-1} . Capacity-voltage curves of (b) SiO_x/C and (c) $\text{SiO}_x/\text{C}@L\text{FB}$ electrodes at various current densities.

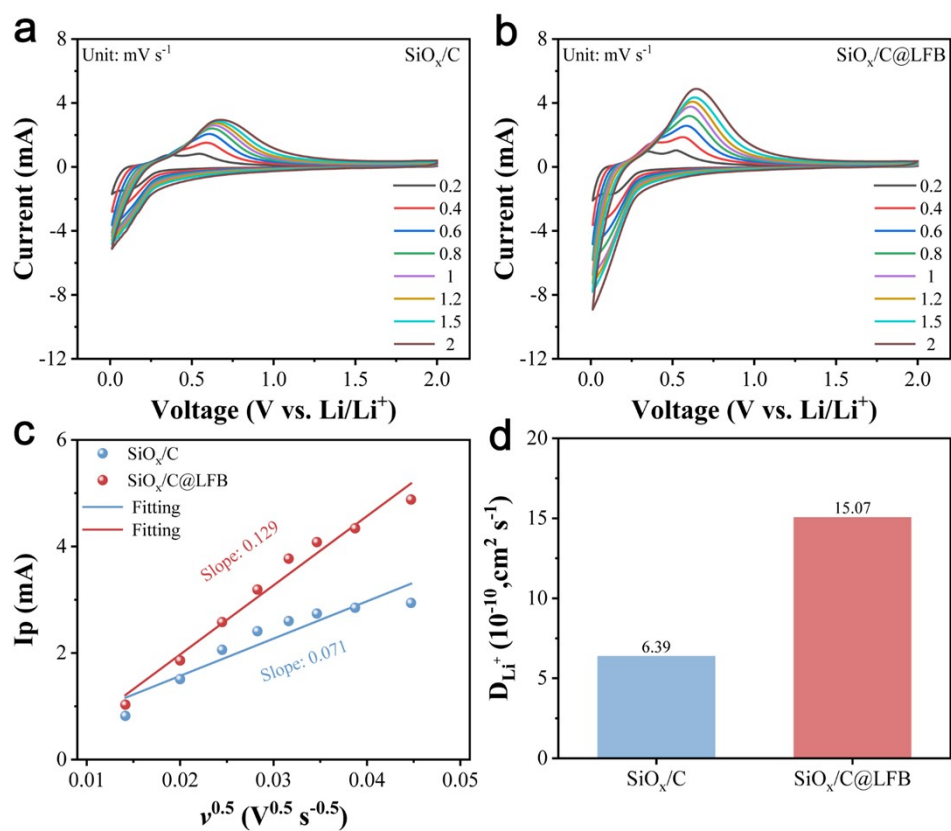


Fig. S5 (a-b) CV curves and (c) I_p versus $\nu^{0.5}$ plots of SiO_x/C and $\text{SiO}_x/\text{C}@LFB$ electrodes at different scan rates. (d) Calculated Li^+ ions diffusion coefficient.

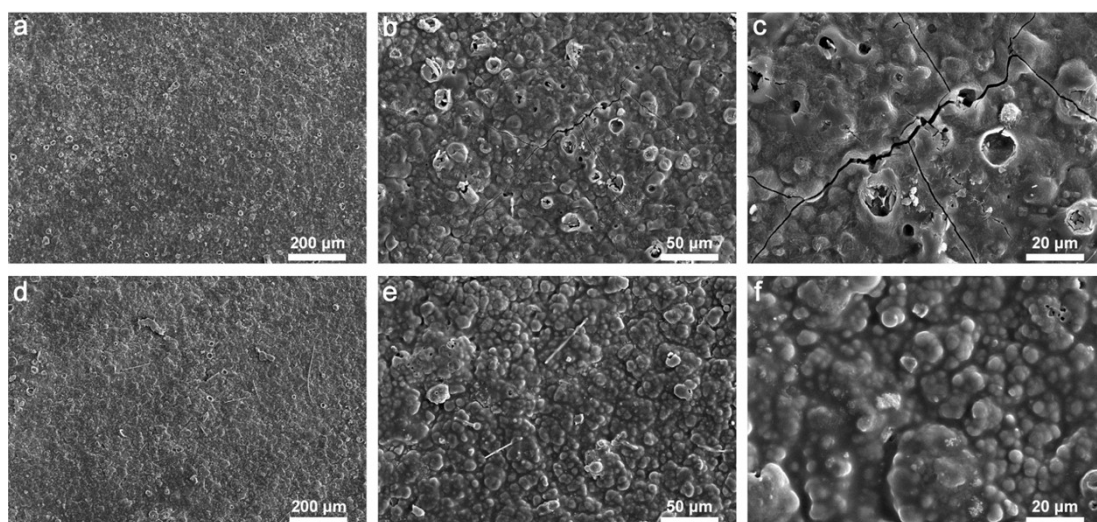


Fig. S6 SEM images of (a-c) SiO_x/C and (d-f) $\text{SiO}_x/\text{C}@LFB$ electrode surface after 50 cycles at 0.5 A g^{-1} .

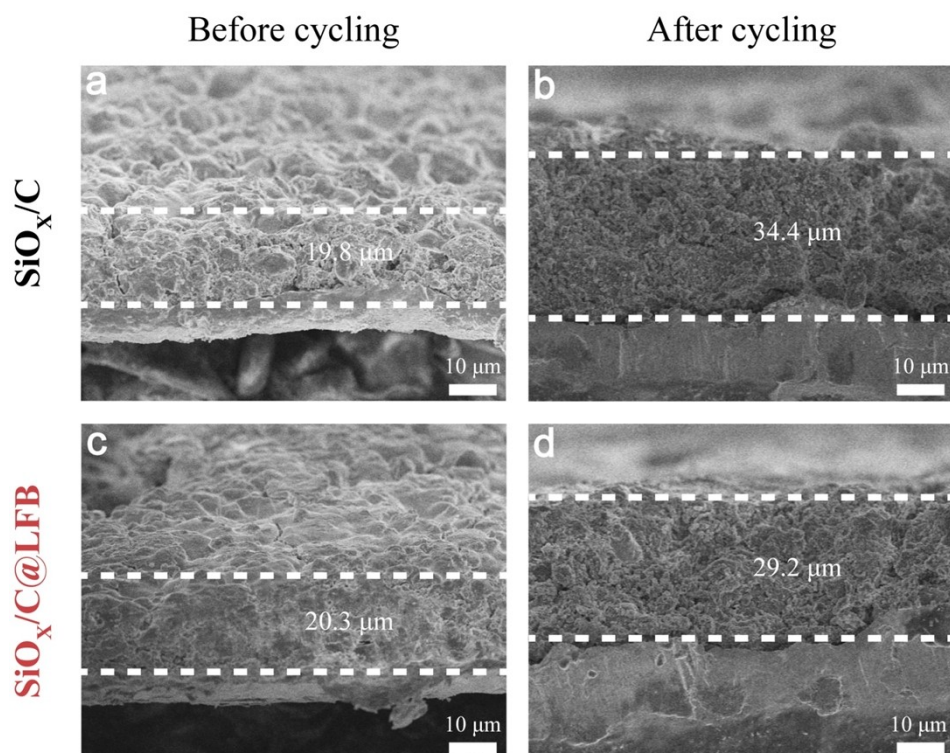


Fig. S7. SEM images of the (a-b) SiO_x/C and (c-d) SiO_x/C@LFB electrode cross-sectional before and after cycling.

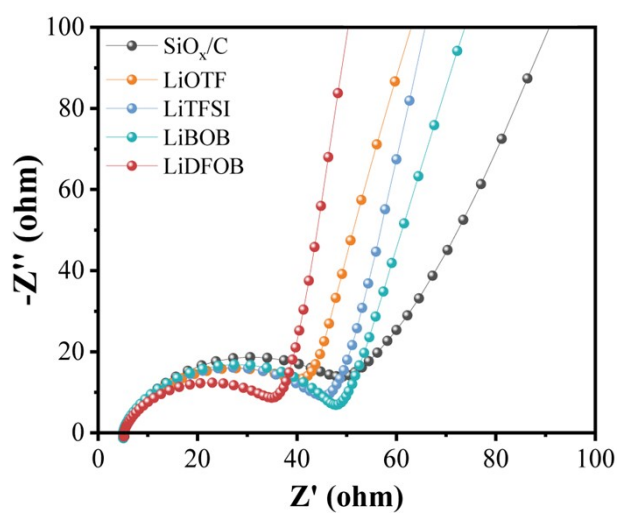


Fig. S8 EIS results of SiO_x/C, LiOTF, LiTFSI, LiBOB, and LiDFOB electrodes before cycling.

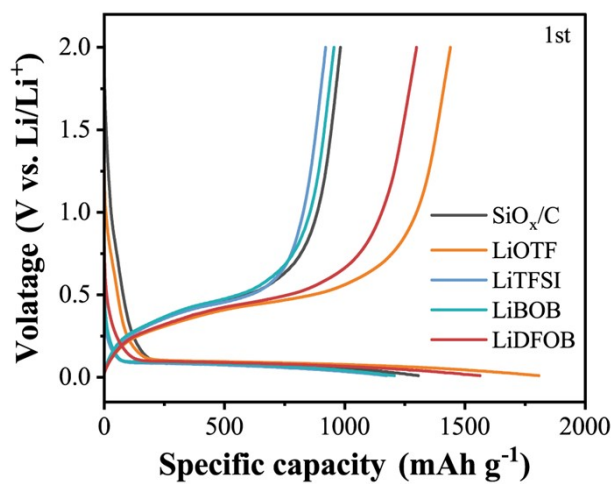


Fig. S9 Charge-discharge curves of SiO_x/C, LiOTF, LiTFSI, LiBOB, and LiDFOB electrodes.

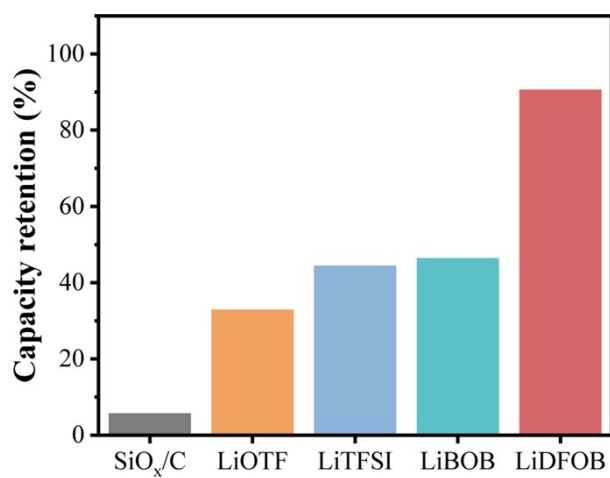


Fig. S10 Capacity retention of SiO_x/C, LiOTF, LiTFSI, LiBOB, and LiDFOB electrodes at 0.5 A g⁻¹.