PEO-Li₂₁Si₅ as a pre-lithiation and structural protection layer for lithium-ion

batteries

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Supplementary Figure 1. Fragmentation process of the $Li_{21}Si_5$ particles.



Supplementary Figure 2. Optical Picture of the $Li_{21}Si_5$ porder.



Supplementary Figure 3. Reaction between $Li_{21}Si_5$ and polar solutions.



Supplementary Figure 4. XRD data of PEO- $Li_{21}Si_5$ patterns exposure in ambient air (40% humidity).



Supplementary Figure 5. XPS full spectrum of PEO-Li $_{21}$ Si₅ powder.



Supplementary Figure 6. FTIR spectra of PEO and PEO-Li $_{21}$ Si₅.





Supplementary Figure 8. First charge/discharge curves of LFP||Si@C/Li₂₁Si₅ full-cell at 0.02 C.



Supplementary Figure 9. long-term cycling test of LFP $||Si@C/Li_{21}Si_5|$ full cell at 2 C.



Supplementary Figure 10. Application of $Li_{21}Si_5$ in carbon-based collectors. a, photograph of carbon nanotube macroscopic film (CMF). b, schematic diagram of CMF as a collector. c, irreversible lithium embedding phenomenon of CMF as an anode collector. d, charging curve of LFP||CMF cell and LFP||Li cell. e, first charge/discharge curve of LFP||CMF cell. f, second turn discharge curve of LFP||CMF cell. g, charging curve of LFP||CMF/Li_{21}Si_5 cell and LFP||Li cell. h, first charge/discharge curve of LFP||CMF/Li_{21}Si_5 cell. i, second turn discharge curve of LFP||CMF/Li_{21}Si_5 cell.