

Supporting Information

Molecular Design of a Multifunctional Binder via Grafting and Cross-linking for High Performance Silicon Anodes

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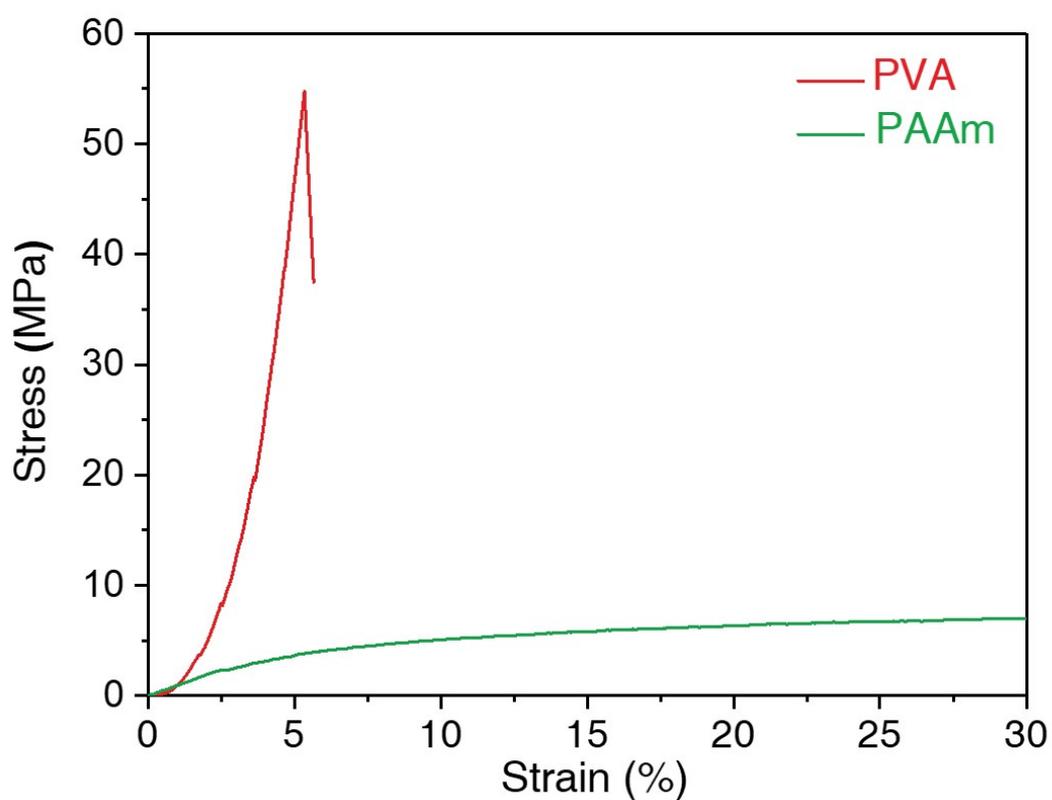


Fig. S1 Tensile test of PVA and PAAm hydrogel sheets. The PAAm hydrogel can be stretched without breaking under the action of a tensile force, indicating that PAAm has excellent elasticity, while PVA bears a large force under the effect of tensile force, and breaks after a short deformation, indicating that PVA hydrogel is rigid compared to PAAm hydrogel.

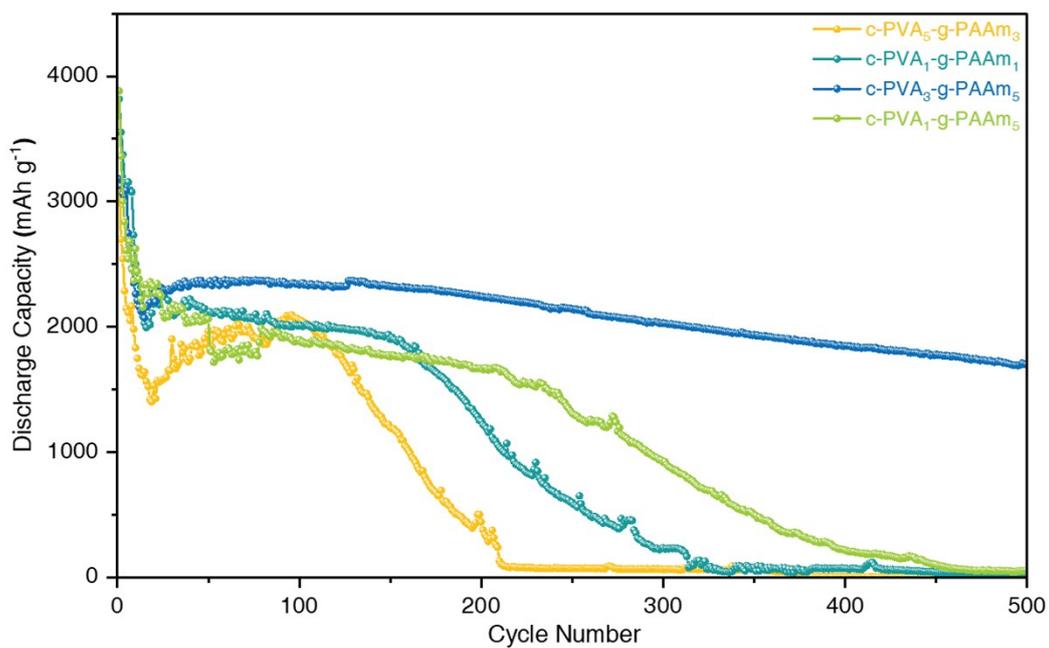


Fig. S2 Long-term cycling behavior of Si-c-PVA-g-PAA electrodes with different graft ratios at 0.2 C. It can be seen that the optimal performance has been obtained with the PVA to PAAm ratio at 3:5. Subsequent experiments have been conducted with this optimized ratio.

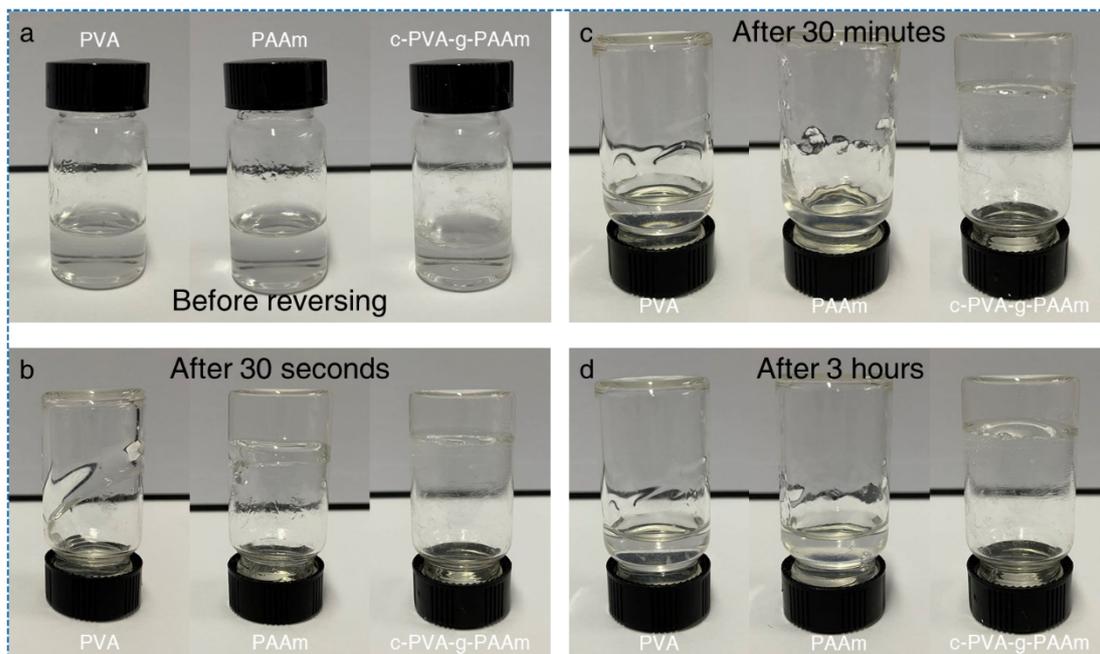


Fig. S3 Digital photos showing the viscosity variation of the as-prepared hydrogels with time at the room temperature. (a) Before reversing, (b) 30 seconds after reversing, (c) 30 minutes after reversing, (d) 3 hours after reversing. Specifically, the PVA and PAAm hydrogels flow down after 30 s and after 30 min, respectively, indicating their low viscosity and thus the ineffective binding with the active materials. As opposed, the graft and cross-linking procedures considerably enhances the solution viscosity, with the c-PVA-g-PAAm hydrogel remaining on the top of the bottle even after 3 h of reversing.

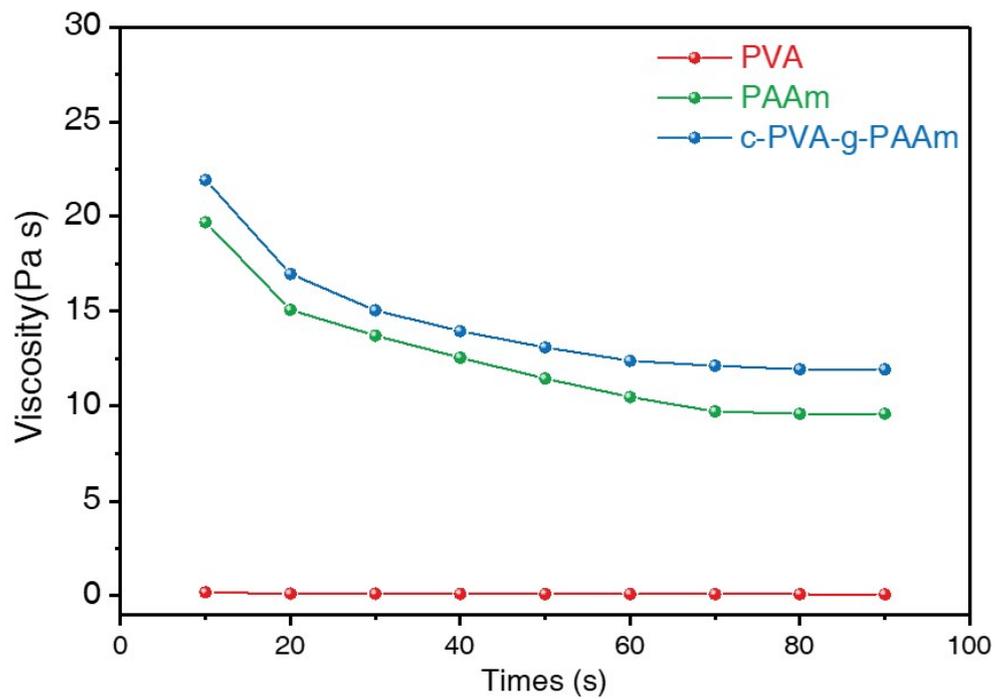


Fig. S4 The viscosity variation of the as-prepared hydrogels conducted on a rotational rheometer at room temperature.

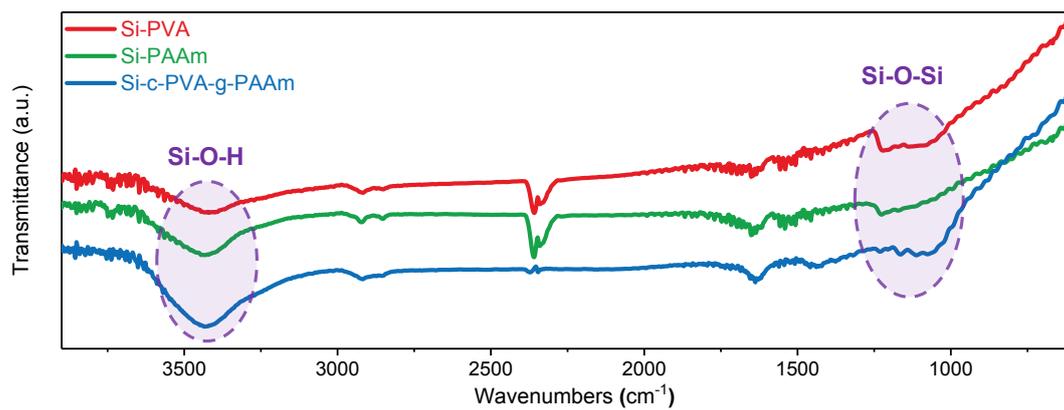


Fig. S5 FTIR spectra of the fresh Si-PVA, Si-PAAm and Si-c-PVA-g-PAAm electrodes.

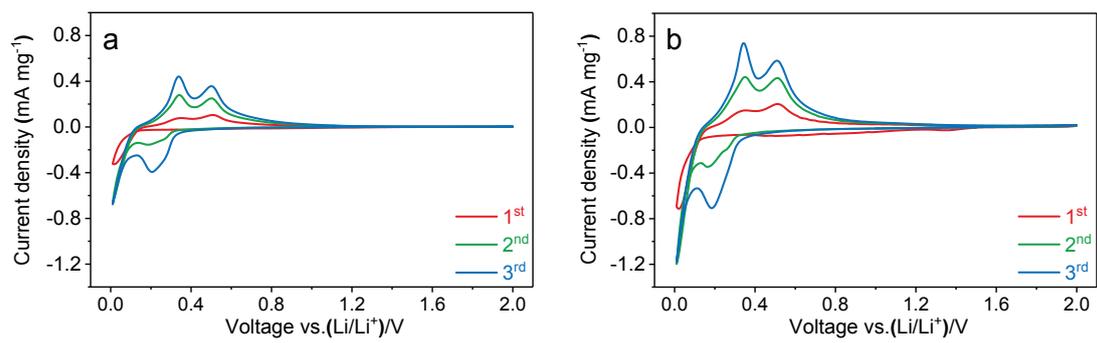


Fig. S6 CV curves of the (a) Si-PVA and (b) Si-PAAm electrodes.

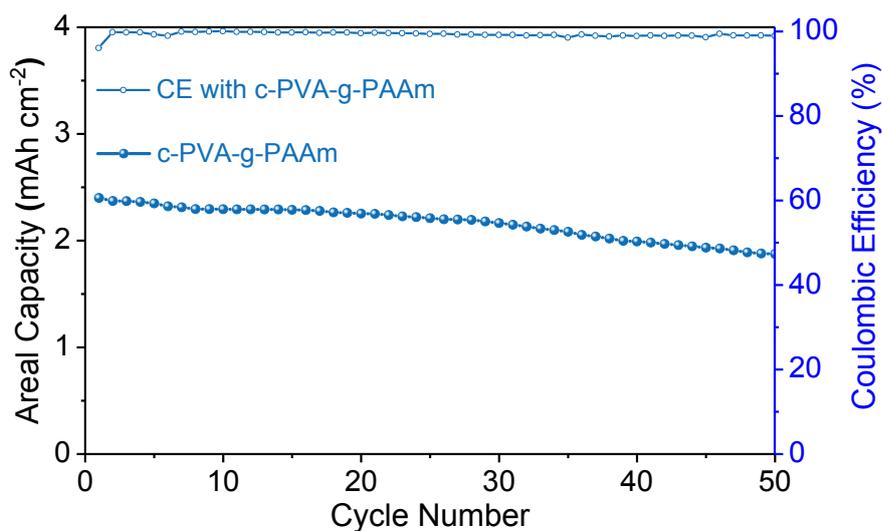


Fig. S7 Cycling performance of the Si-c-PVA-g-PAAm/LiFePO₄ full cell at a current density of 1 mA cm⁻². The loading of Si-c-PVA-g-PAAm and LiFePO₄ on the electrodes is 1.2 and 14.92 mg cm⁻², respectively. Prior to this, the Si-c-PVA-g-PAAm electrode was firstly pre-lithiated at a relatively low current density of 0.2 mA cm⁻² for three cycles to alleviate the effect of low initially Coulombic efficiency of the full cell.

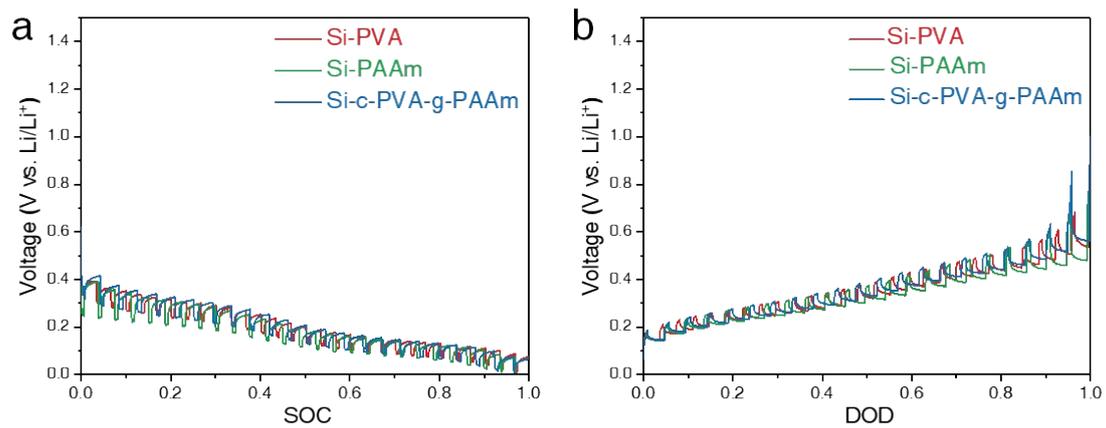


Fig. S8 GITT curves of (a) Si-PVA and (b) Si-PAAm electrodes.

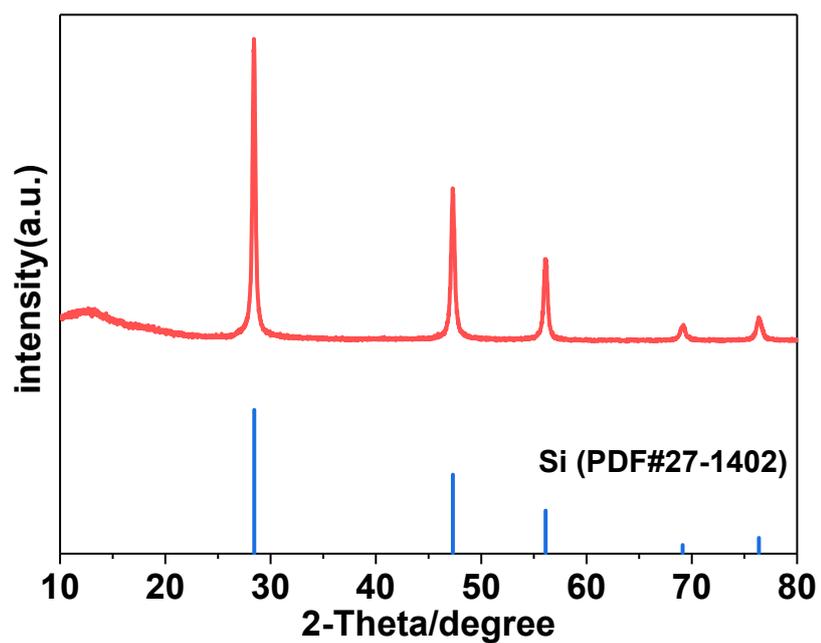


Fig. S9 XRD patterns of Si particles. Crystal phase structures of Si particles were characterized by XRD. **Fig. S8** displays the characteristic peaks of Si, which confirmed that the Si particles are pure phase.

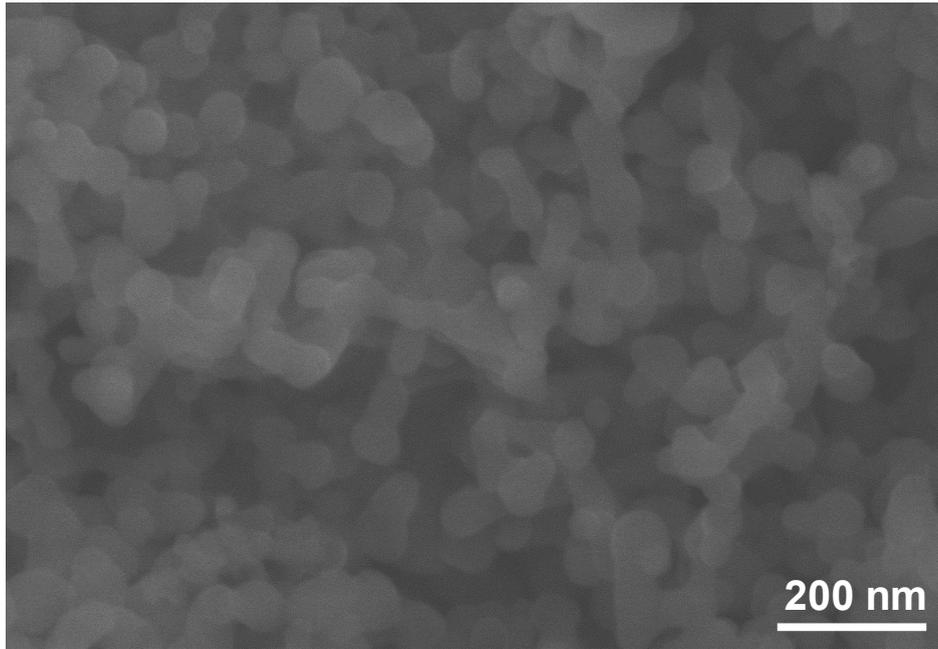


Fig. S10 SEM image of Si particle, showing the homogeneous distribution of the nanosized Si particles.

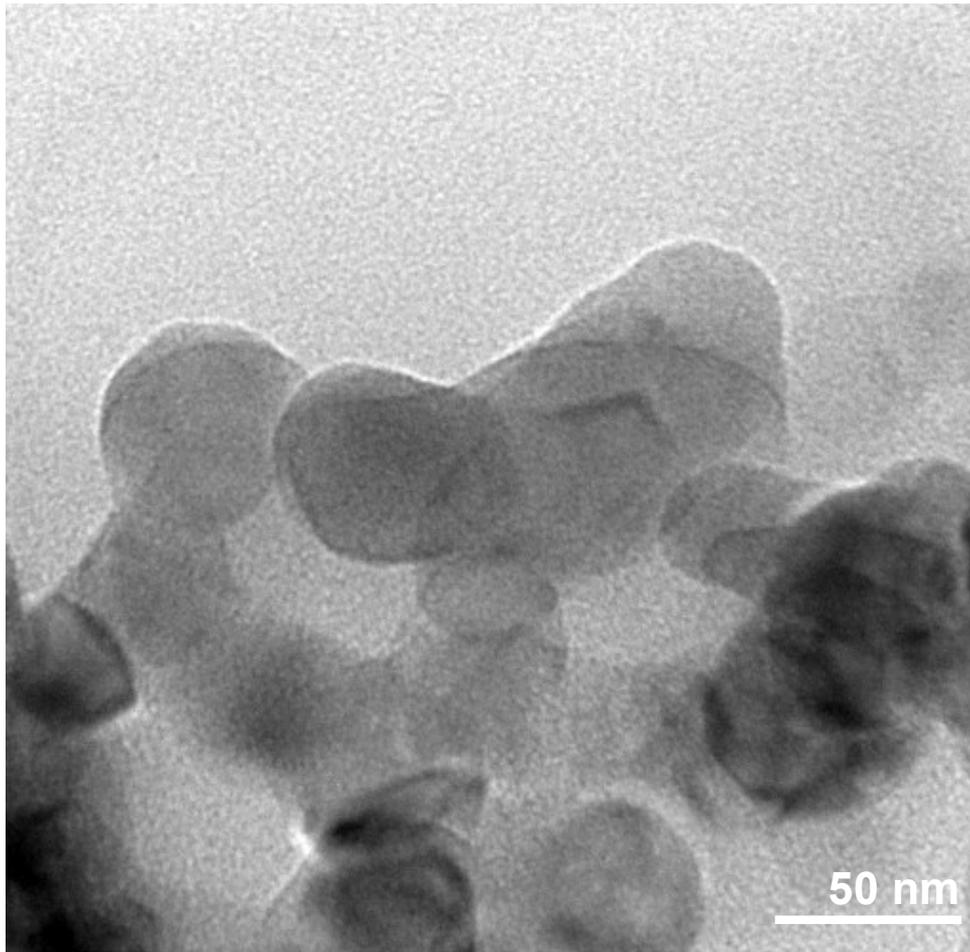


Fig. S11 TEM image of Si particles. The average diameter of the particles locates between 50 to 60 nm.

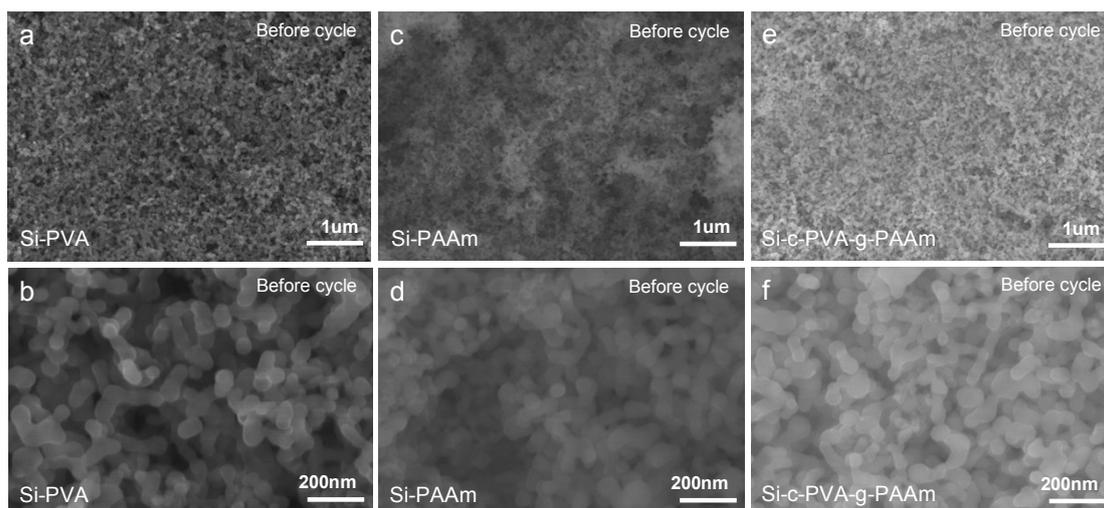


Fig. S12 SEM images of pristine Si electrodes with the (a,b) PVA, (c,d) PAAm and (e,f) c-PVA-g-PAAm binder, where the three electrodes show similar morphology and particle distribution before cycling, despite a relatively rougher surface of the Si-PAAm electrode.

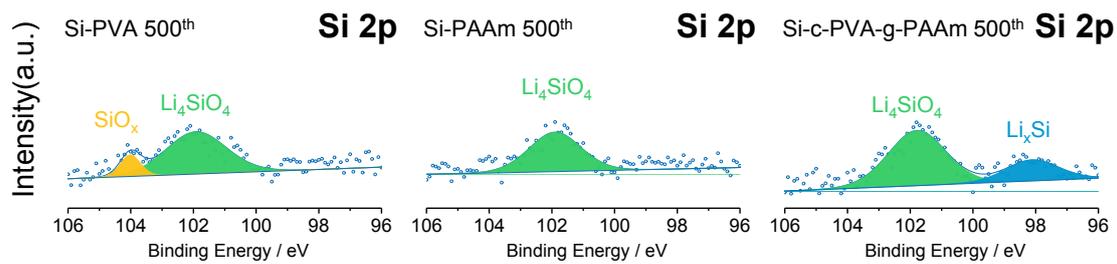


Fig. S13 XPS spectra of the Si 2p peaks of the Si-PVA, Si-PAAm and Si-c-PVA-g-PAAm electrodes after 500 cycles.

Table S1 Impedance parameters of electrodes Si-PVA, Si-PAAm and Si-c-PVA-g-PAAm electrodes after different cycles.

Si electrode	$R_{SEI} (\Omega) 8^{th}$	$R_{CT} (\Omega) 8^{th}$	$R_{SEI} (\Omega) 500^{th}$	$R_{CT} (\Omega) 500^{th}$
PVA	5.2	4.3	74.3	65.7
PAAm	8.7	37.3	69.8	64.2
c-PVA-g-PAAm	9.6	16.4	33.2	26.8