

Supporting Information

Chitosan-g-Poly(Acrylic Acid) Copolymer and Its Sodium Salt as Stabilized Aqueous Binders for Silicon Anodes in Lithium-Ion Batteries

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No. of pages: 10

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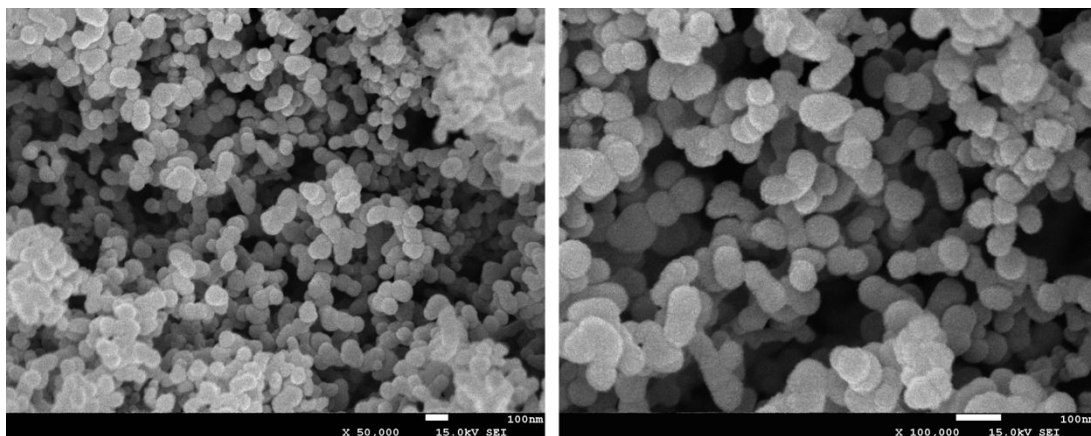


Figure S1. SEM images of Si nanoparticles.

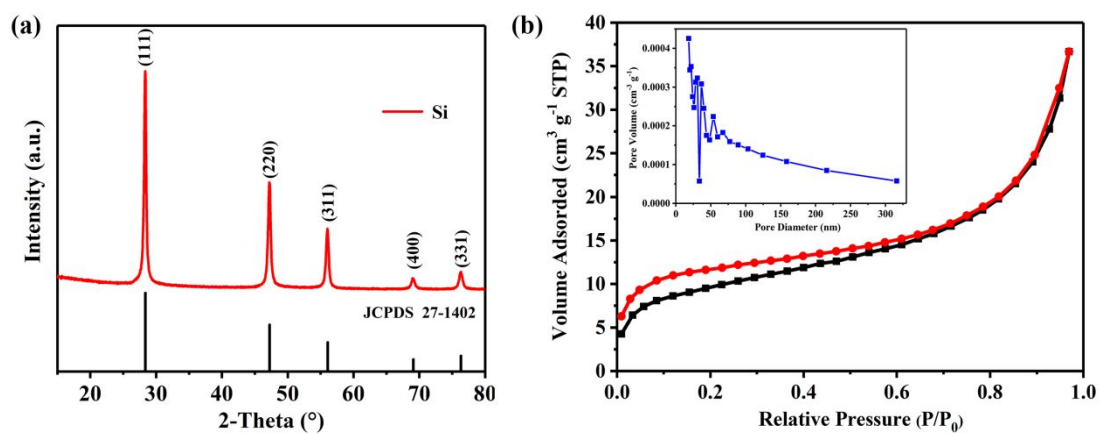


Figure S2. XRD pattern (a) and nitrogen adsorption-desorption isotherm and the (inset) pore size distribution (b) of Si nanoparticles.

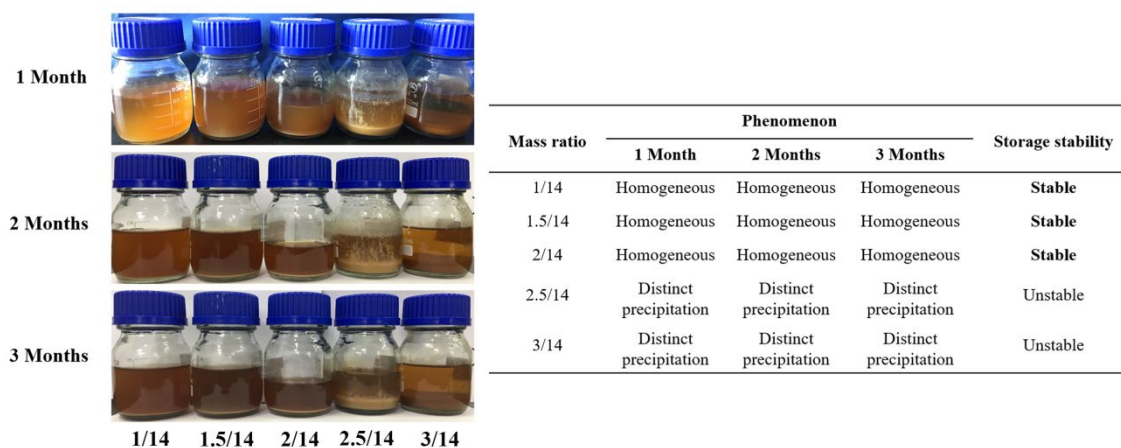


Figure S3. Storage stability of CS-PAA prepared under different CS/AA weight ratios.

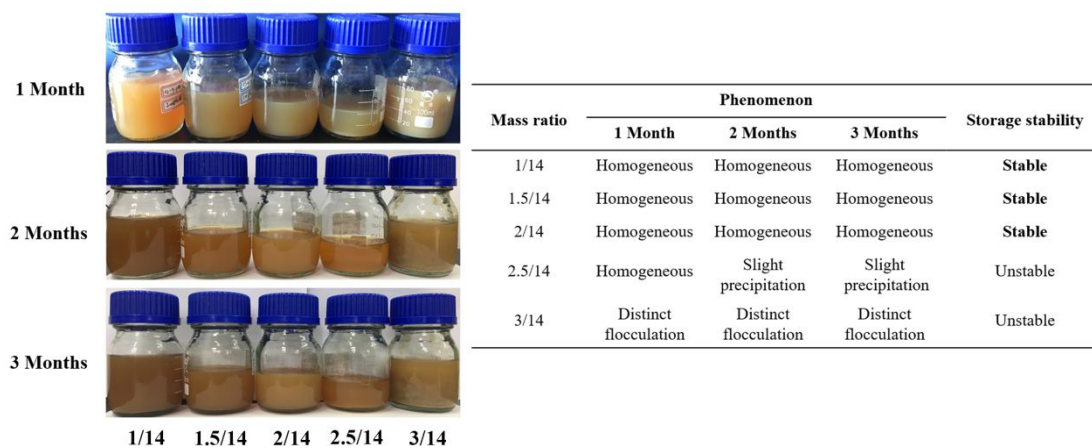


Figure S4. Storage stability of CS-PAANa prepared under different CS/AA weight ratios.

Table S1. Solubility of CS-PAA and CS-PAANa films in different solvents under 25 °C and 50 °C.

| Sample | Soak for 18 hours (25 °C) | Solubility | Soak for 18 hours (50 °C) | Solubility |
|----------------------|------------------------------|------------|------------------------------|------------|
| CS-PAA membrane | Acetic acid solution (3 wt%) | Insoluble | Acetic acid solution (3 wt%) | Insoluble |
| | DMSO | | DMSO | |
| | NMP | | NMP | |
| | MeOH | | MeOH | |
| CS-PAANa membrane | Acetic acid solution (3 wt%) | Insoluble | Acetic acid solution (3 wt%) | Insoluble |
| | DMSO | | DMSO | |
| | NMP | | NMP | |
| | MeOH | | MeOH | |

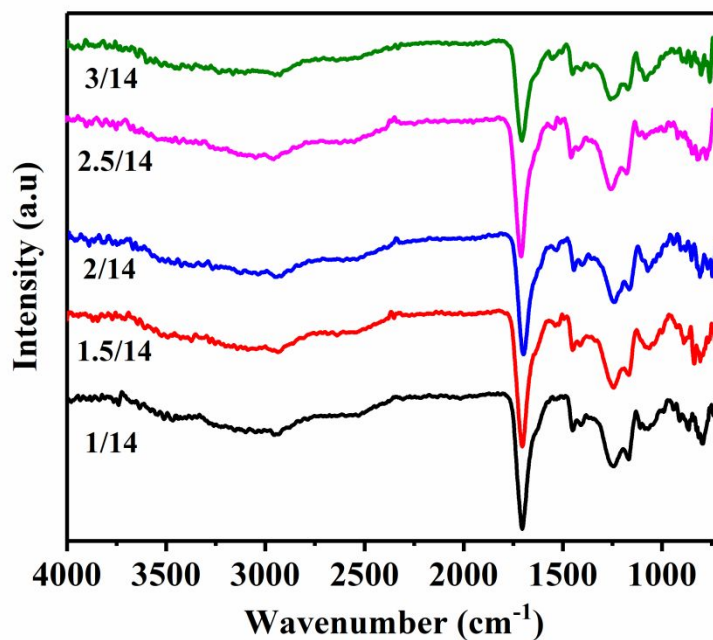


Figure S5. FT-IR spectra of CS-PAA binder films with different CS/AA weight ratios.

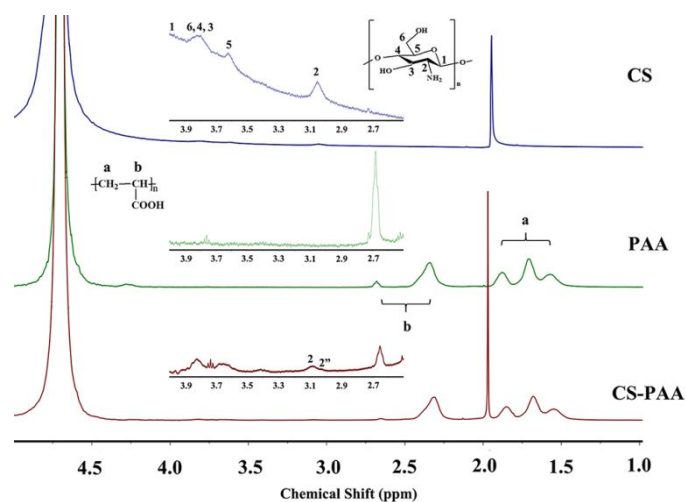


Figure S6. ^1H -NMR spectra of CS, PAA, and CS-PAA binders.

The ^1H -NMR spectra of CS, PAA, and CS-PAA binders are shown in **Figure S6**, Proton signals at 3.0-3.1 ppm (C-2), 3.5-3.7 ppm (C-5), 3.7-3.9 ppm (C-6, 4, 3) and 3.9-4.0 ppm (C-1) are observed, which conform to the ^1H -NMR spectrum of CS as a reference.^[1-3] The proton signals at 1.4-1.9 (a) and 2.2-2.8 (b) ppm in PAA are attributed to the protons of the acrylic unit.^[4] For CS-PAA, all proton signals for CS and PAA are observed after graft polymerization. Meanwhile, a shifted proton signal (~ 3.0 ppm) is detected, corresponding to the reaction of PAA onto C2-NH₂ of CS.^[5,6]

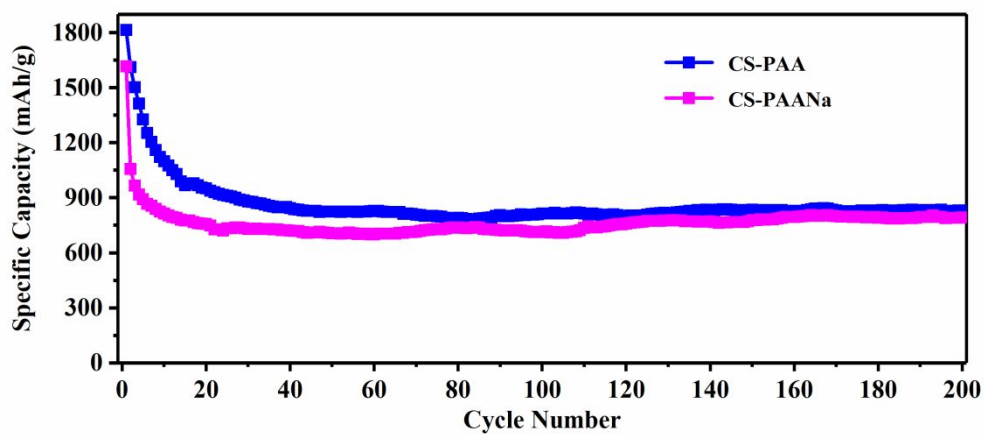


Figure S7. Cycling performance of electrodes at 2.7 A/g (mass loading is ~ 0.3 mg).

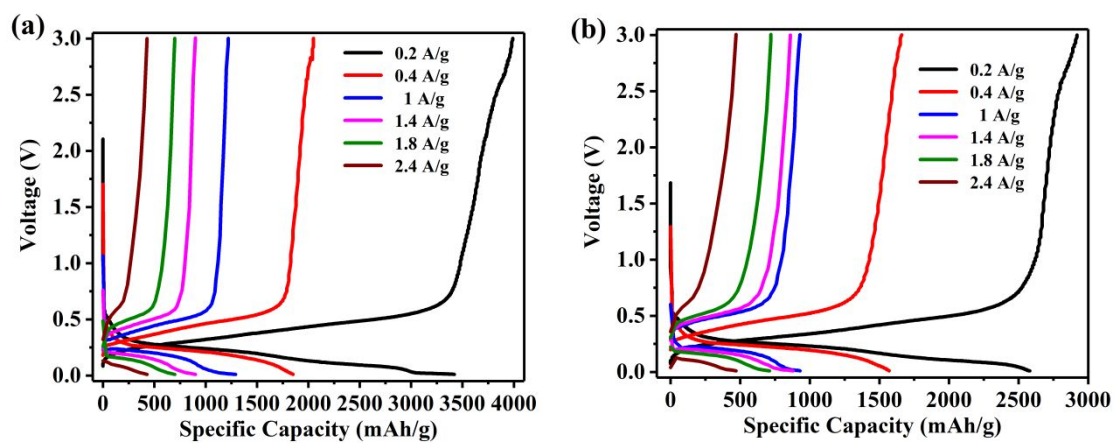


Figure S8. Galvanostatic voltage profiles of (a) Si/CS-PAA and (b) Si/CS-PAANa electrodes (mass loading is ~ 1 mg).

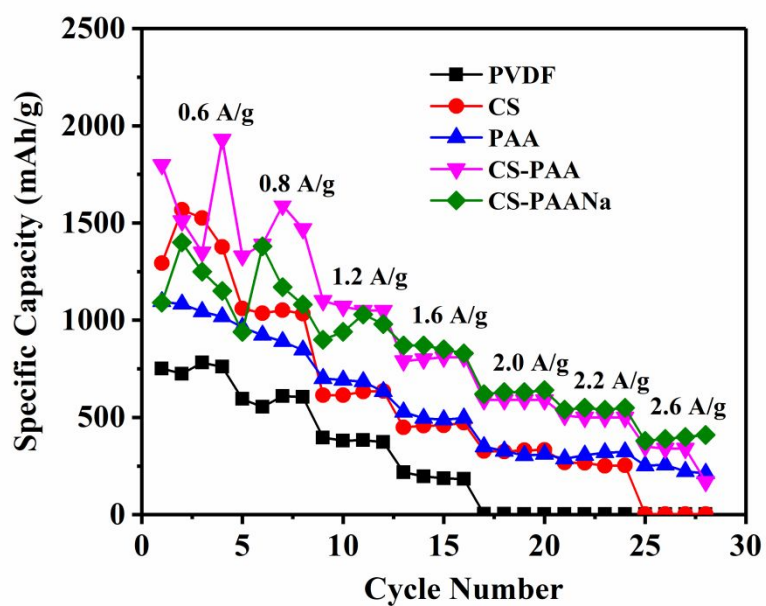


Figure S9. Rate performance of Si/PVDF, Si/CS, Si/PAA, Si/CS-PAA, and Si/CS-PAANa

electrodes (mass loading is ~1 mg).

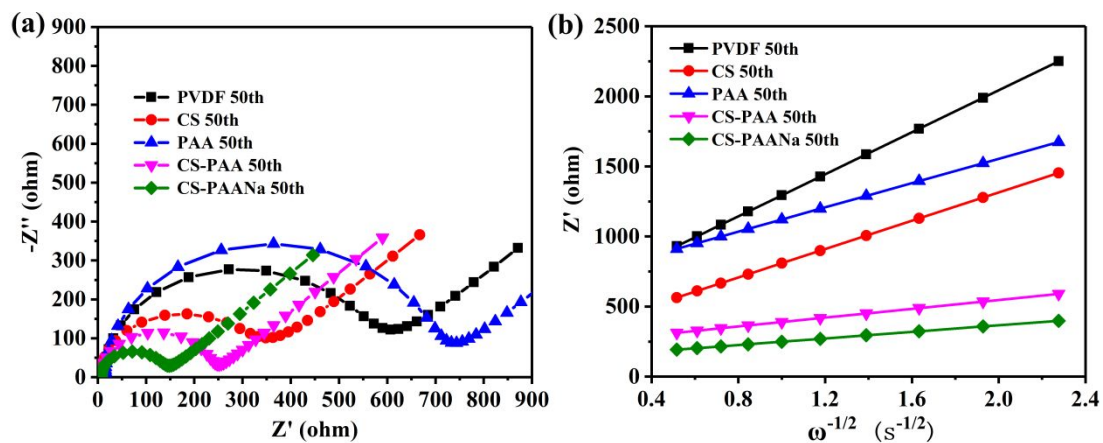


Figure S10. Electrochemical impedance spectra (a) and the plots of Z_{re} vs $\omega^{-1/2}$ for Si/PVDF, Si/CS, Si/PAA, Si/CS-PAA and Si/CS-PAANa electrodes after 50 cycles (b).

Table S2. Impedance parameters of Si/PVDF, Si/CS, Si/PAA, Si/CS-PAA and Si/CS-PAANa

electrodes after 50 cycles.

| Sample | R_s (Ω) | R_{ct} (Ω) | Warburg coefficient | D ($\times 10^{-17}$ $\text{cm}^2 \text{s}^{-1}$) |
|---------------|--------------------|-----------------------|---------------------|---|
| PVDF 50th | 13.33 | 534.40 | 119.37 | 0.30 |
| CS 50th | 5.87 | 303.00 | 80.50 | 0.99 |
| PAA 50th | 15.29 | 674.2 | 68.95 | 1.57 |
| CS-PAA 50th | 6.28 | 225.90 | 25.10 | 32.51 |
| CS-PAANa 50th | 6.08 | 126.80 | 18.65 | 79.25 |

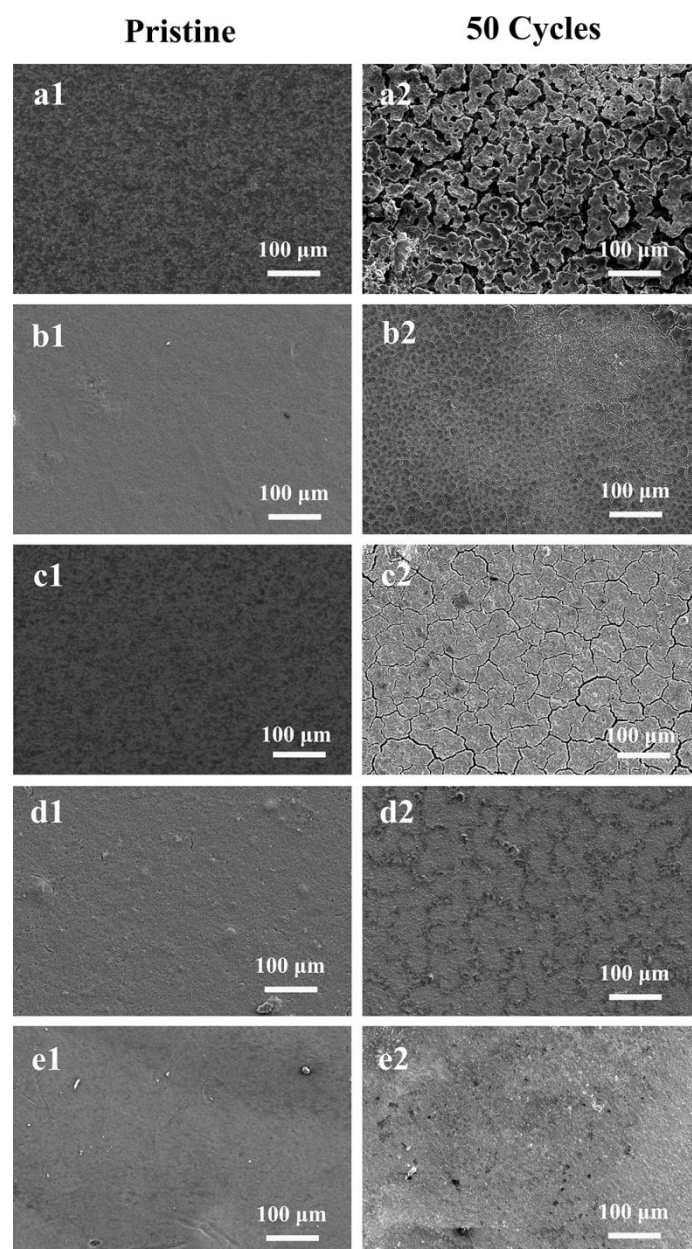


Figure S11. SEM images of (a) Si/PVDF, (b) Si/CS, (c) Si/PAA, (d) Si/CS-PAA and (e) Si/CS-PAA Na electrode surfaces at low magnification.

Reference

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