Supporting Information for ***Journal of Materials Chemistry A***

Electrochemically prelithiated carbon anodes with regulated Na-ion intercalation behaviors for advanced sodium-ion energy storage devices

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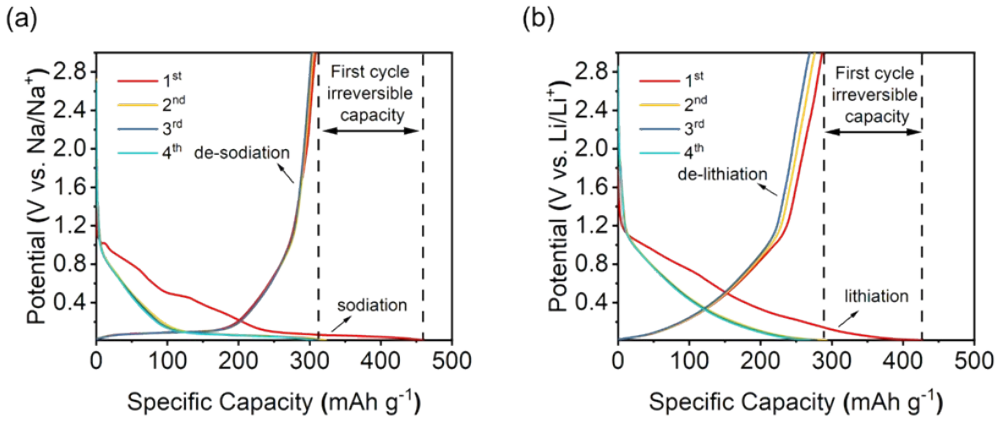

Figure S1. (a) Initial discharge/charge profiles of HC anode in a half cell with metal Na as the counter electrode. (b) Initial discharge/charge profiles of HC anode electrode with metal Li as the counter electrode.

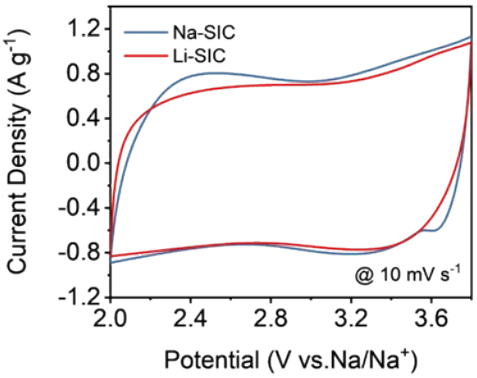

Figure S2. A comparison of CV curves of Li-SIC and Na-SIC based on the prelithiated HC anode and presodiated HC anode at a scan rate of 10 mV s^{-1}.

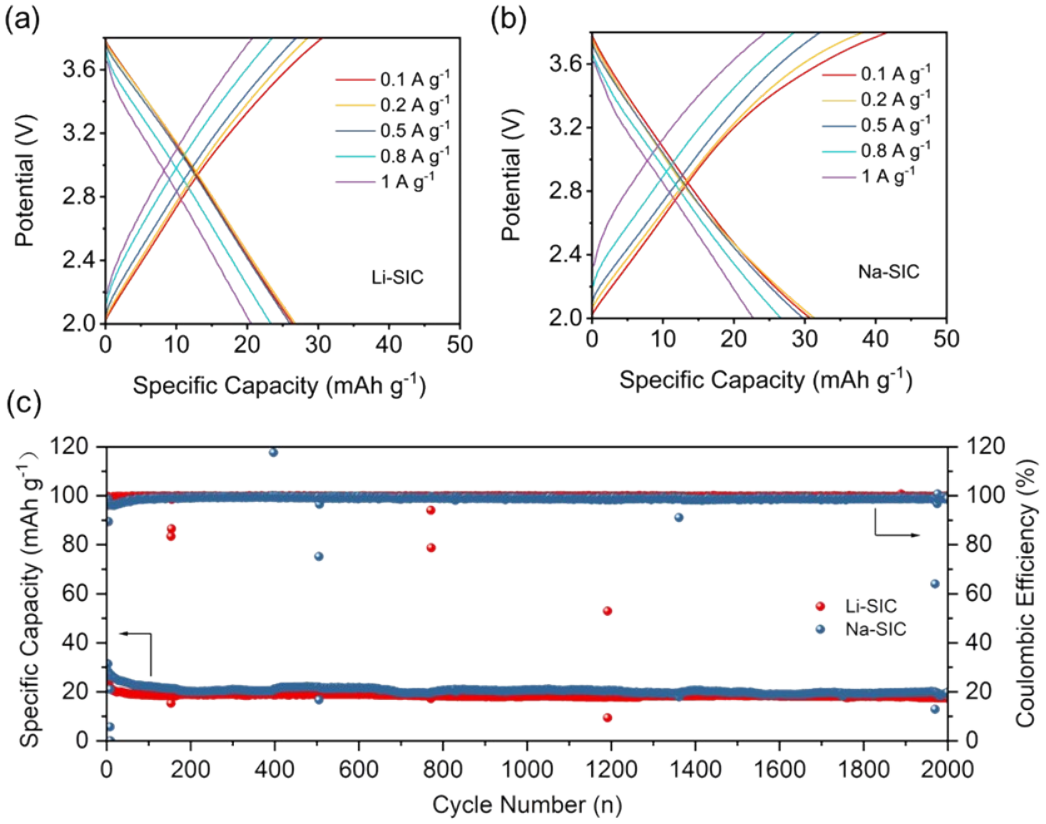

Figure S3. Charge/discharge curves at the different rates from 0.1 to 1 A g^{-1}: (a) Li-SIC and (b) Na-SIC. (c) The comparison of long-term cycling performance at a current rate of 100 mA g^{-1} for Li-SIC (red) and Na-SIC (blue).

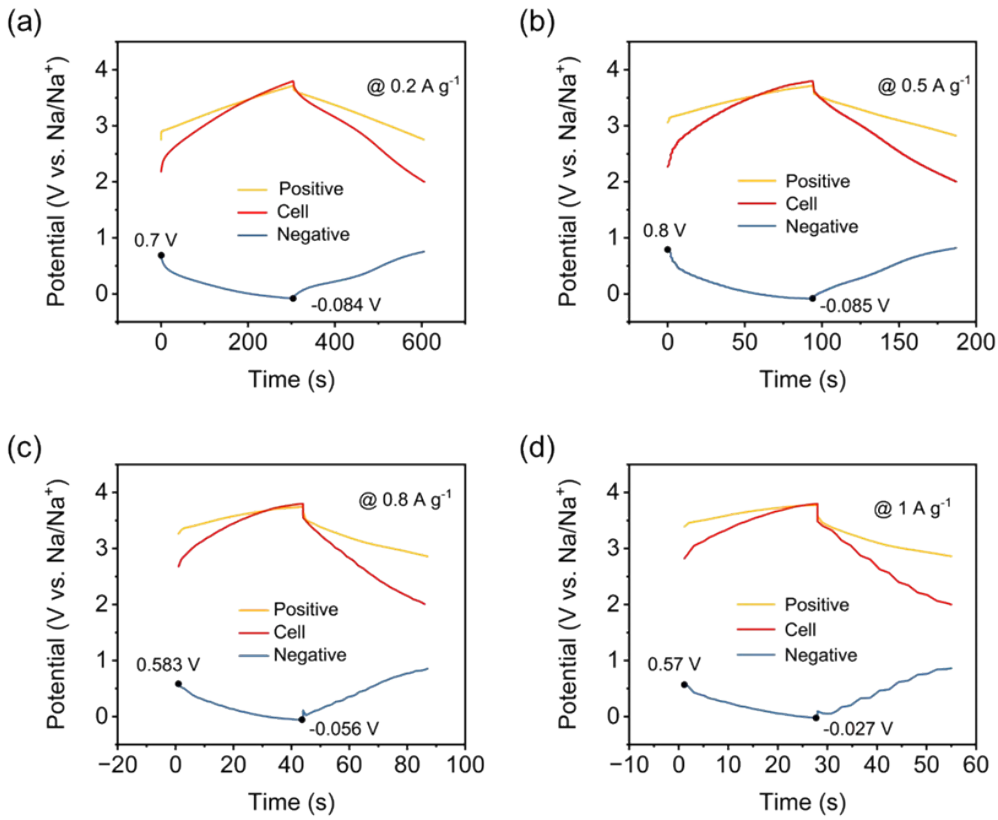

Figure S4. Galvanostatic charge/discharge profiles of Li-SIC recorded from a three-electrode Swagelok cell with a Na metal reference at the current densities of (a) 0.2 A g^{-1}, (b) 0.5 A g^{-1}, (c) 0.8 A g^{-1}, and (d) 1.0 A g^{-1}, respectively.

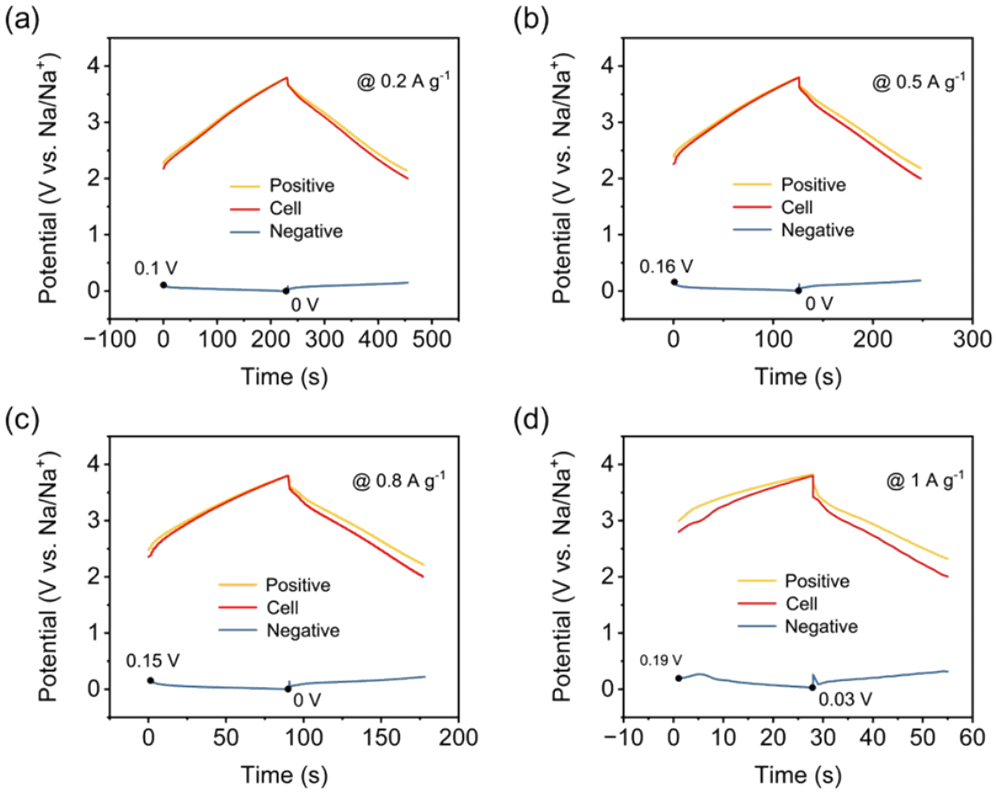

Figure S5. Galvanostatic charge/discharge profiles of Na-SIC recorded from a three-electrode Swagelok cell with a Na metal reference at the current densities of (a) 0.2 A g^{-1}, (b) 0.5 A g^{-1}, (c) 0.8 A g^{-1}, and (d) 1.0 A g^{-1}, respectively.

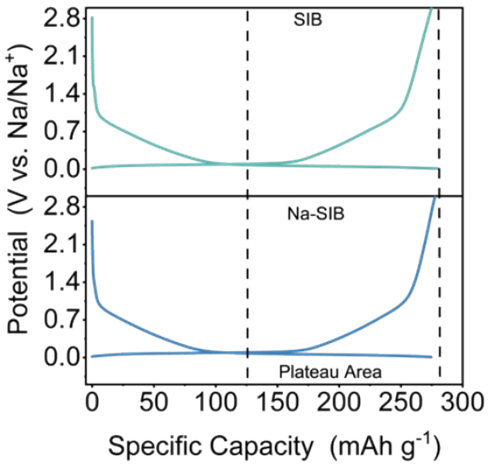

Figure S6. A comparison of charge/discharge profiles for HC anode based half cell before (upper) and after pre-sodiation (bottom) at a current density of 50 mA g^{-1}.

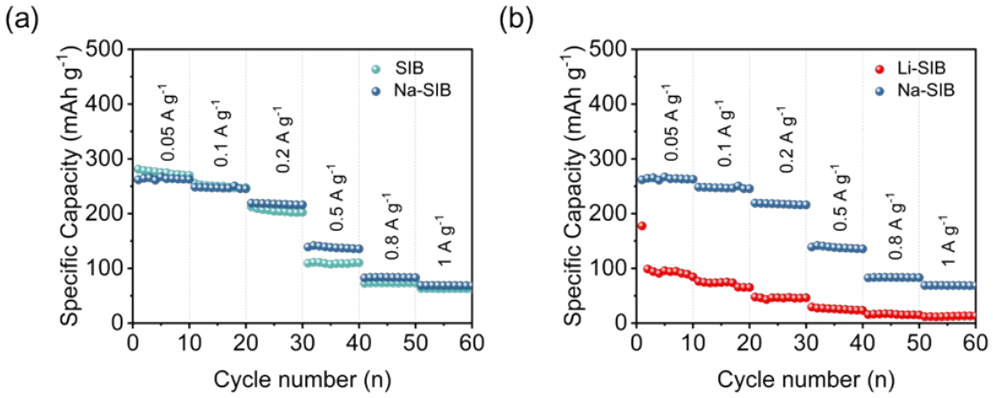

Figure S7. Rate capability at various current rates ranging from 0.05 to 1 A g^{-1} recorded in a half cell. (a) The comparison between HC anodes before (SIB) and after pre-sodiation (Na-SIB). (b) The comparison HC anodes after pre-lithiation (Li-SIB) and pre-sodiation (Na-SIB).

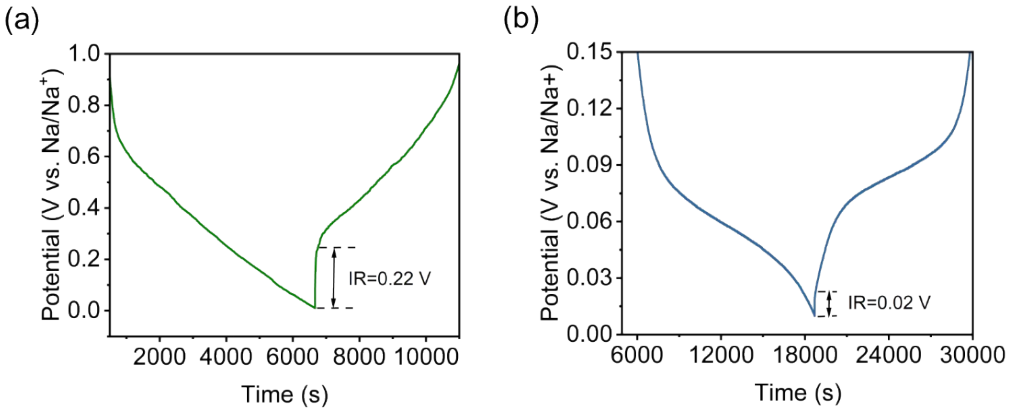

Figure S8. IR drops achieved from voltage profiles at a current density of 0.05 A g^{-1} in (a) Li-SIB and (b) Na-SIB.

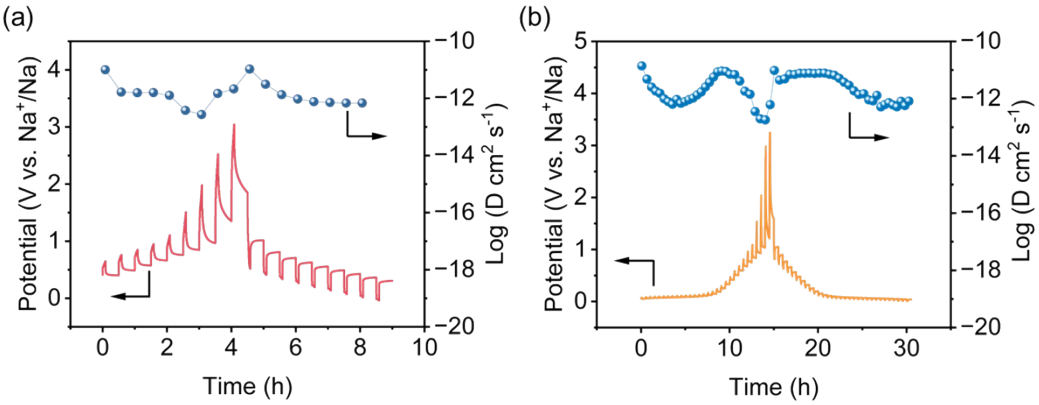

Figure S9. (a) Na^+ diffusion coefficient of prelithiated HC anode in a HC/Na half-cell evaluated from GITT curves. (b) Na^+ diffusion coefficient of presodiated HC anode in a HC/Na half-cell evaluated from GITT curves.

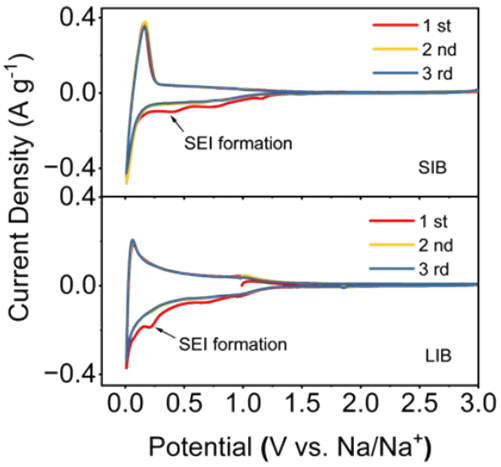

Figure S10. The comparison of CV curves of HC/Na (denoted as SIB) half cell (upper) and HC/Li (denoted as LIB) half cell (bottom) at a scan rate of 0.2 mV s^{-1}.

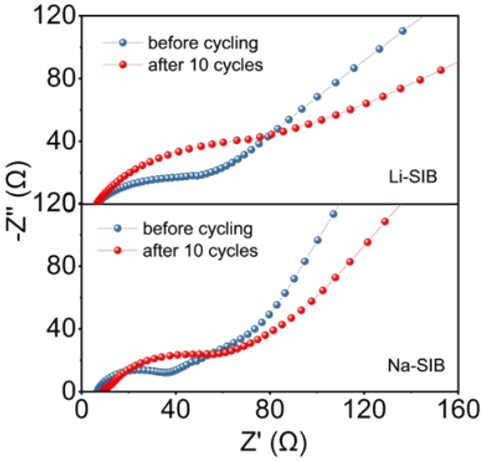

Figure S11. The comparison of EIS curves of prelithiated HC anode (denoted as Li-SIB) (upper) and presodiated HC anode (denoted as Na-SIB) (bottom) in the half cells with Na metal as the counter electrode.

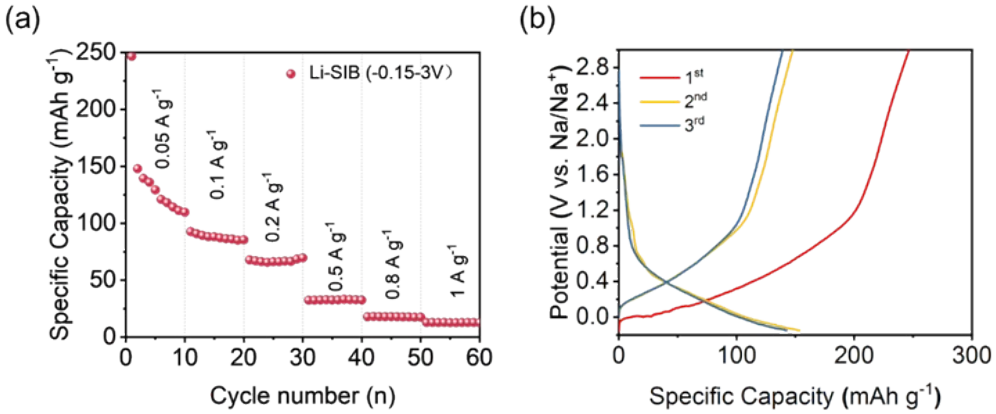

Figure S12. The electrochemical performance of Li-SIB with the expanded working potential from -0.15 to 3V (vs Na^+/Na). (a) The rate capability at the various current rates from 0.05 to 1 A g^{-1}. (b) Initial discharge/charge profiles.

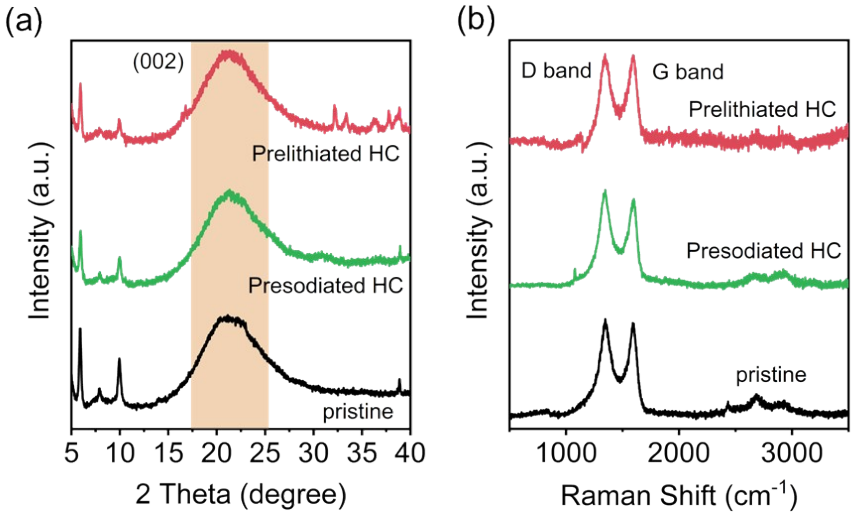

Figure S13. (a) XRD patterns of pristine HC, presodiated HC and prelithiated HC. (b) Raman spectra of pristine HC, presodiated HC and prelithiated HC. Note that these new peaks between 30-40° on ex-situ XRD pattern of prelithiated HC sample are mainly related to the Li_2CO_3, LiF, Li_2O and/or LiOH species from the SEI layer coated on prelthiated HC, and the broad peak around 38° is mainly due to the formation of LiF nanocrystals.

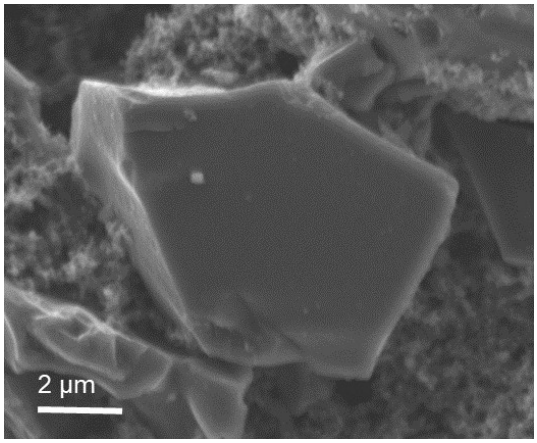

Figure S14. SEM image of pristine HC.

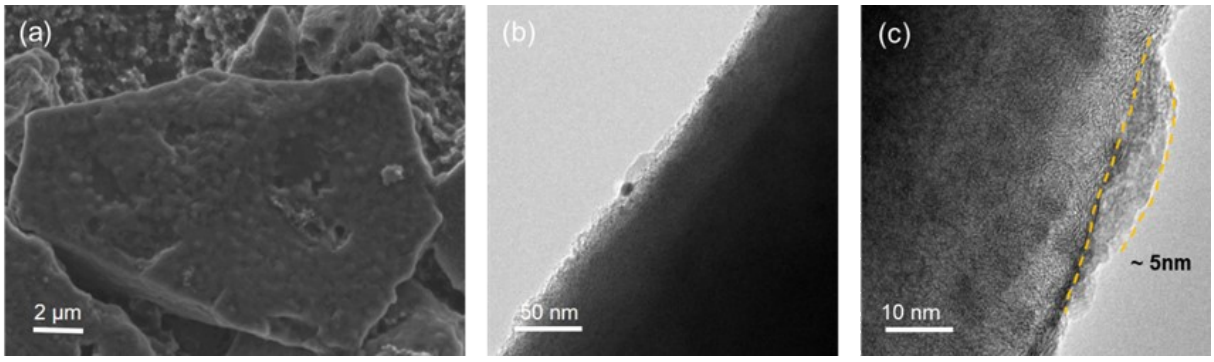

Figure S15. Morphological and structural characterization of presodiated HC anode after ten cycles in SIC: (a) SEM image, (b) TEM image, and (c) HRTEM image.

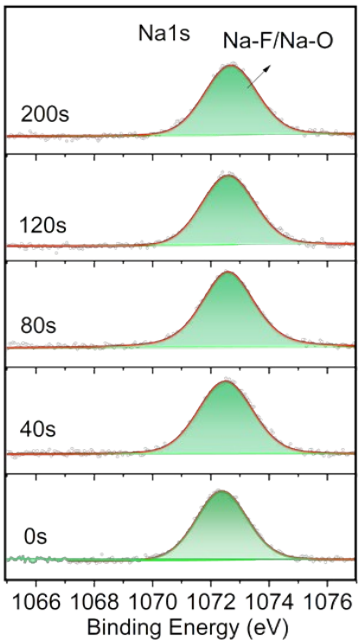

Figure S16. Na 1s XPS spectra for prelithiated HC anode after ten cycles in SICs.

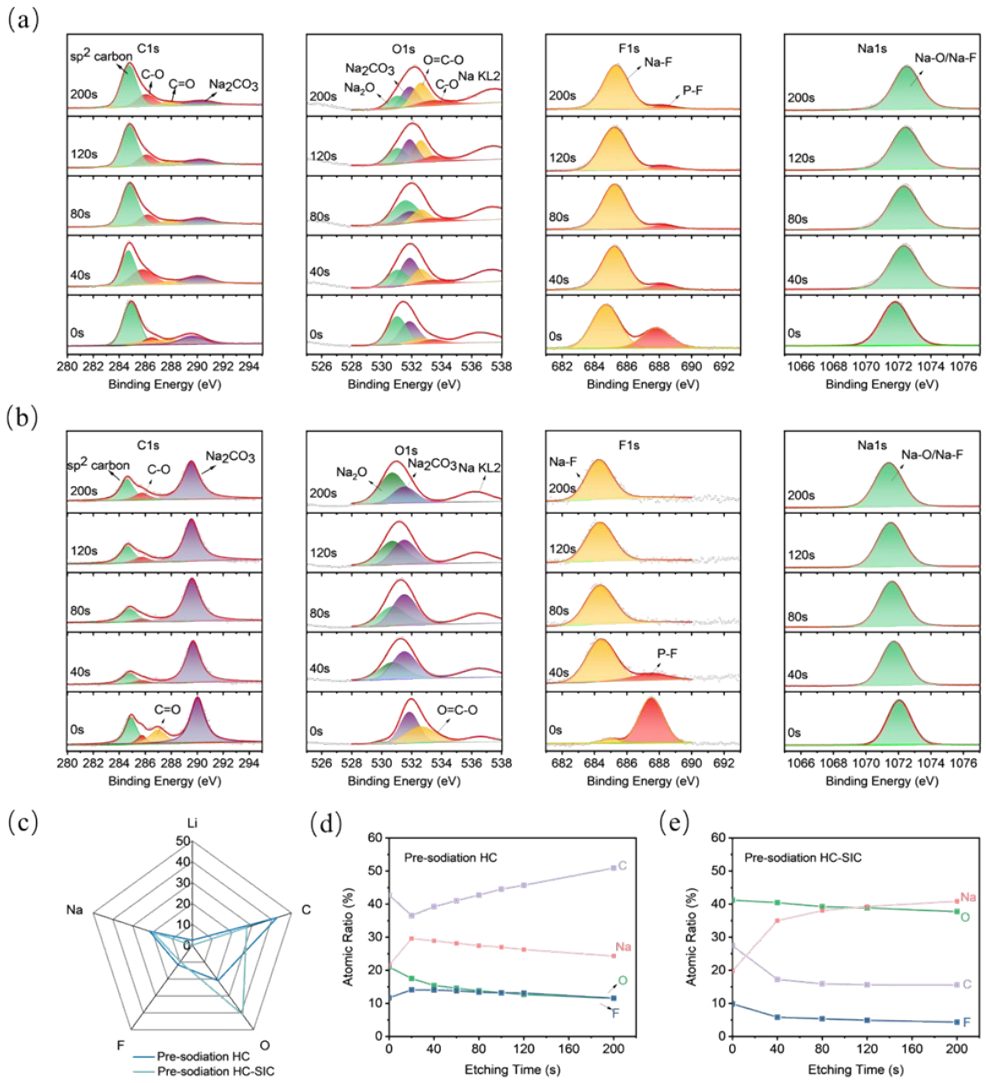

Figure S17. (a) C 1s, O 1s, F 1s and Na 1s XPS spectra of for the presodiated HC anode. (b) C 1s, O 1s, F 1s and Na 1s XPS spectra of for the presodiated HC anode after ten cycles in SIC. (c) Comparison of the elemental concentrations of the SEI on the surfaces of presodiated HC before and after ten cycles in SIC. Atomic compositions of Na, O, C, and F collected at different depths on presodiated HC anode (d) before and (e) after ten cycles in SIC.

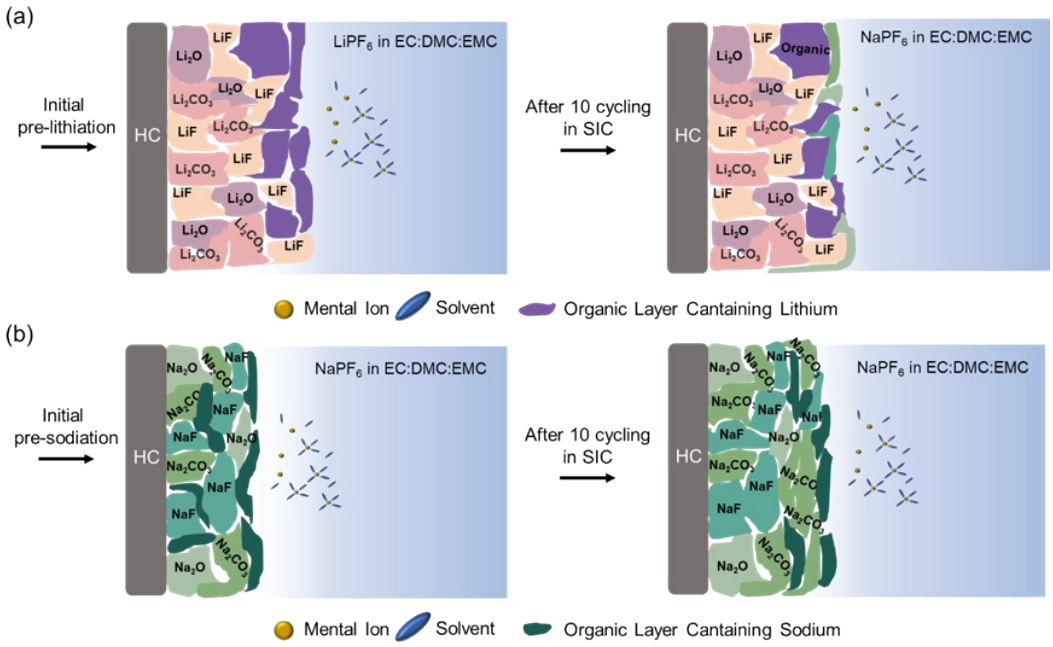

Figure S18. Schematic illustration of SEI layers formed on the surfaces of HC anodes: (a) Prelithiated HC anode before and after cycled in SIC. (b) Presodiated HC anode before and after cycled in SIC.

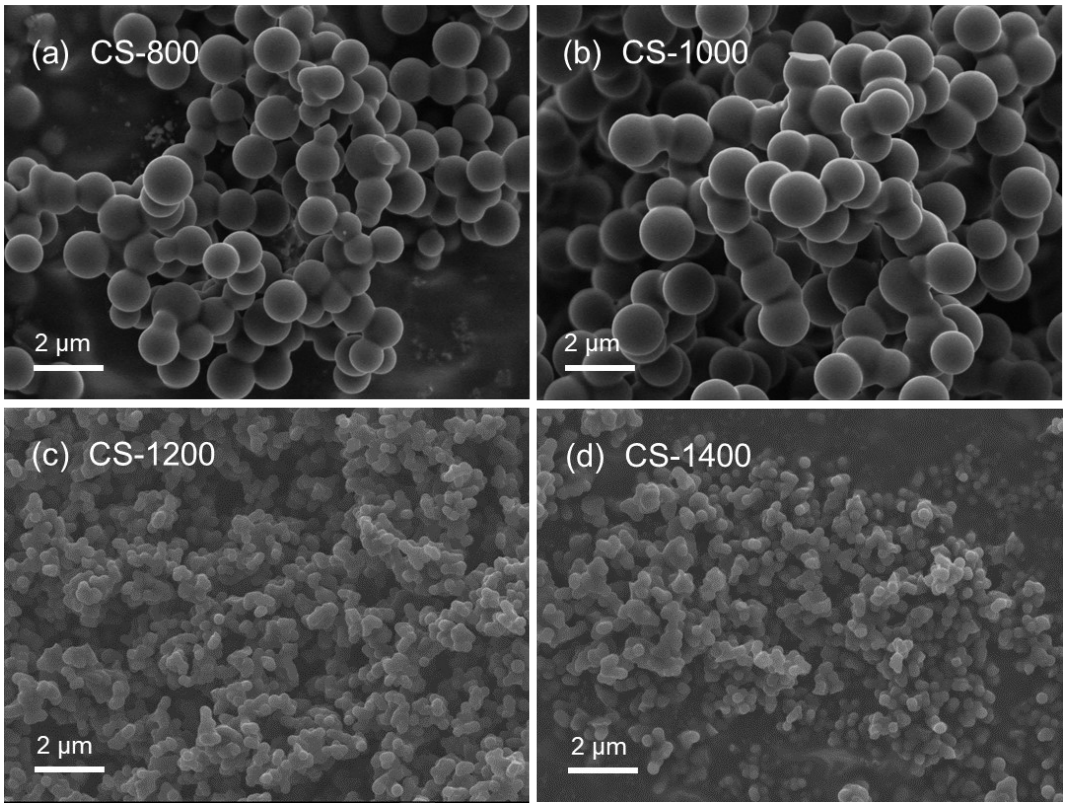

Figure S19. SEM images of home-made carbon spheres (CSs) pyrolyzed at different temperatures: (a) 800 °C, (b) 1000 °C, (c) 1200 °C and (d) 1400 °C.

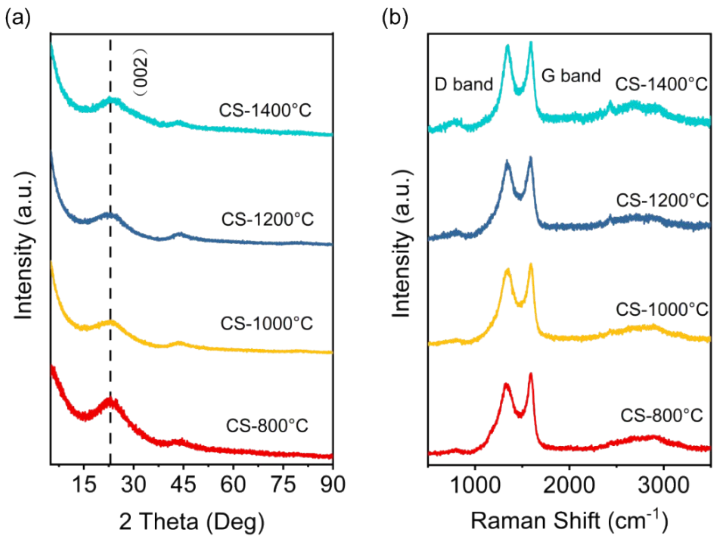

Figure S20. (a) XRD patterns and (b) Raman spectra of CSs samples pyrolyzed at different temperatures.

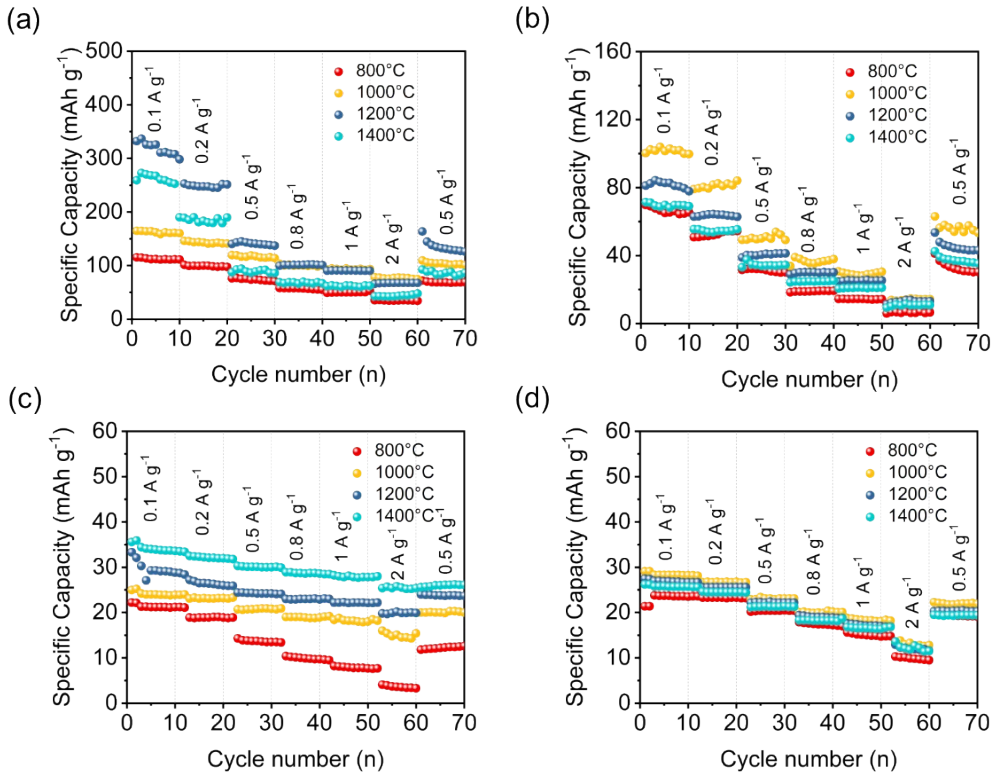

Figure S21. The rate capabilities of CSs pyrolyzed at different temperatures at various current rates from 0.1 to 2 A g^{-1} in (a) presodiated CSs/Na half cell, (b) prelithiated CSs/Li half cell, (c) presodiated CSs/AC full cell, and (d) prelithiated CSs/AC full cell.

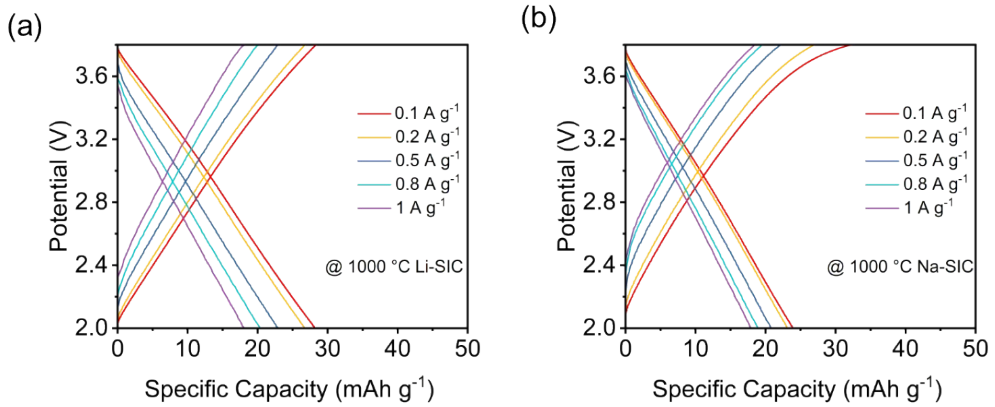

Figure S22. The charge/discharge profiles of SICs at various current densities from 0.1 to 1 A g^{-1} in (a) prelithiated CSs/AC SIC (denoted as Li-SIC) and (b) presodiated CSs/AC SIC (denoted as Na-SIC).

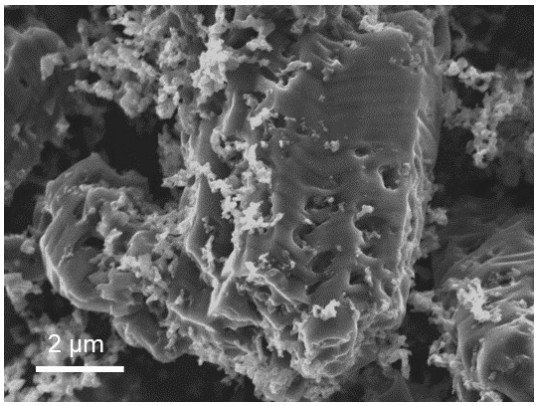

Figure S23. SEM image of SC.

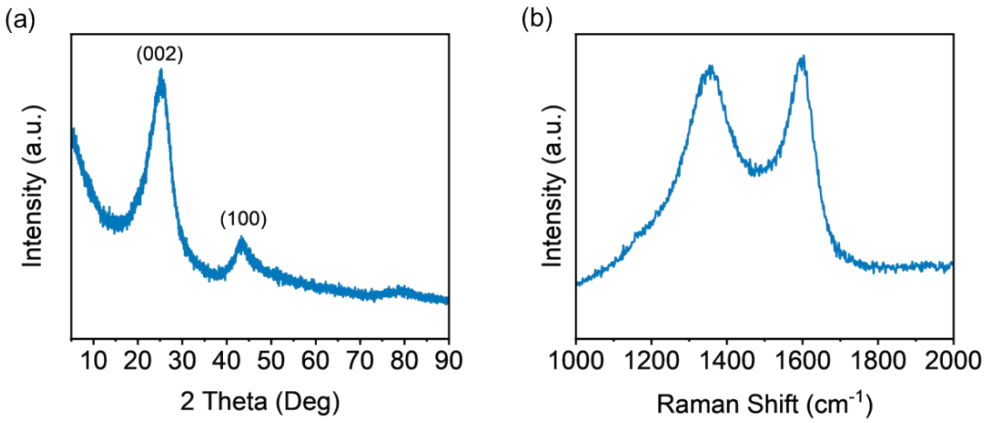

Figure S24. The XRD pattern (a) and Raman spectra (b) of SC sample.

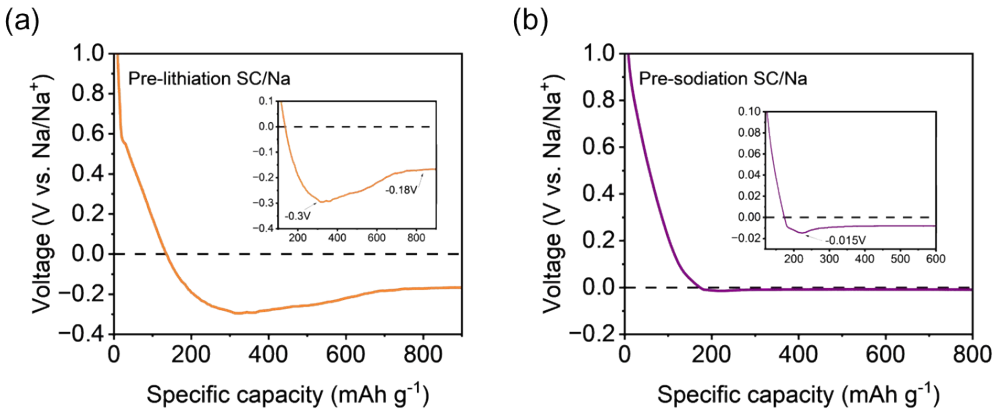

Figure S25. Voltage profiles of galvanostatic Na deposition on (a) prelithiated SC anode and (b) presodiated SC anode in half cell at 0.1 A g^{-1}.

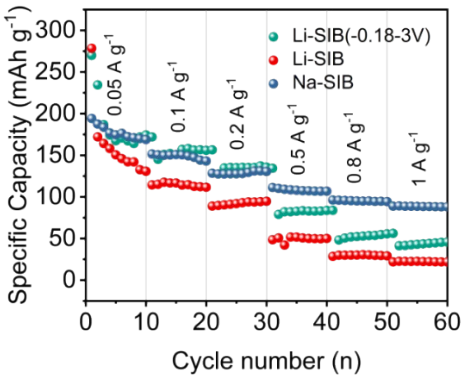

Figure S26. Rate capabilities at various current densities from 0.05 to 1 A g^{-1} for prelithiated SC anode with expanded working potential from -0.18 to 3V, prelithiated SC anode and presodiated SC anode in half cells with metal Na as the counter electrode.

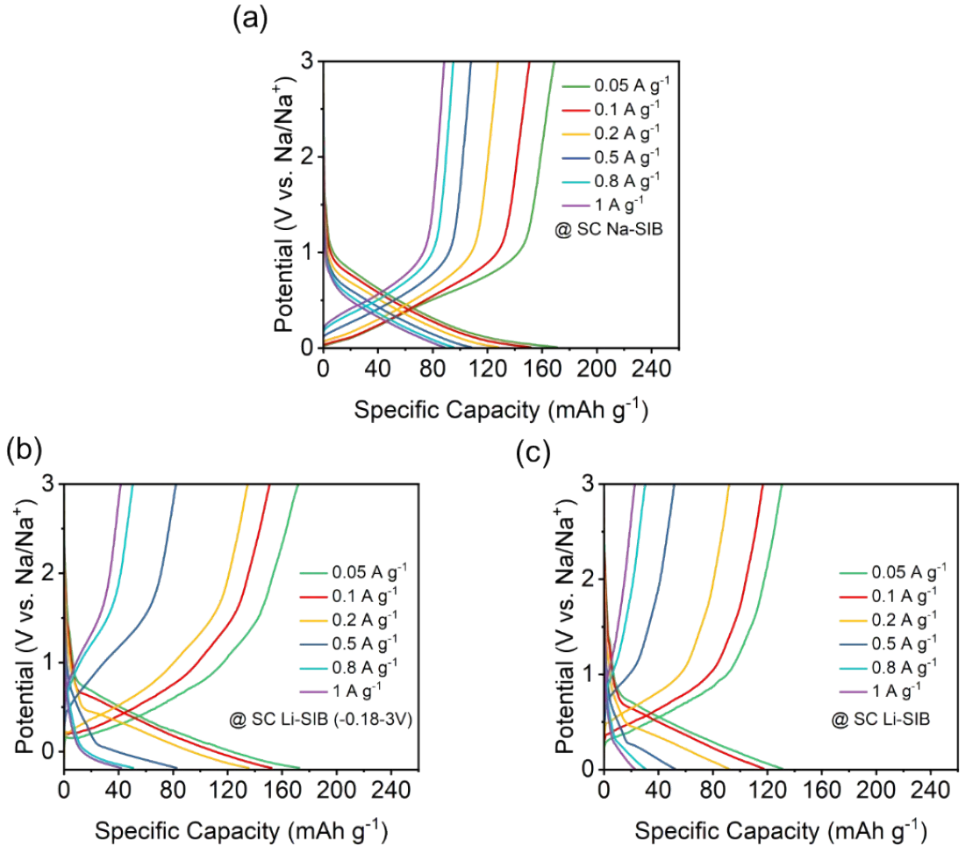

Figure S27. The charge/discharge profiles at various current densities from 0.1 to 1 A g^{-1} in half cells for (a) presodiated SC anode (denoted as Na-SIB), (b) prelithiated SC anode with the expanded working potential from -0.18 to 3 V vs Na^+/Na (denoted as Li-SIB (-0.18-3 V)) and (c) prelithiated SC anode (denoted as Li-SIB).

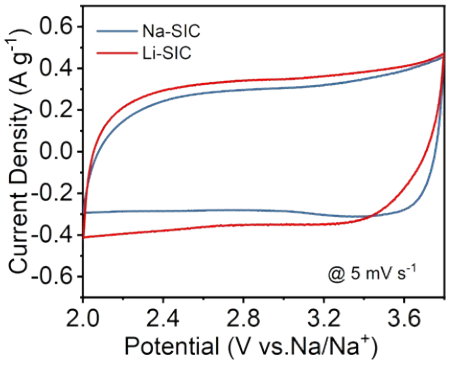

Figure S28. The CV curves of presodiated SC//AC SIC (denoted as Na-SIC) and prelithiated SC//AC SIC (denoted as Li-SIC) at a scan rate of 5 mV s^{-1}.

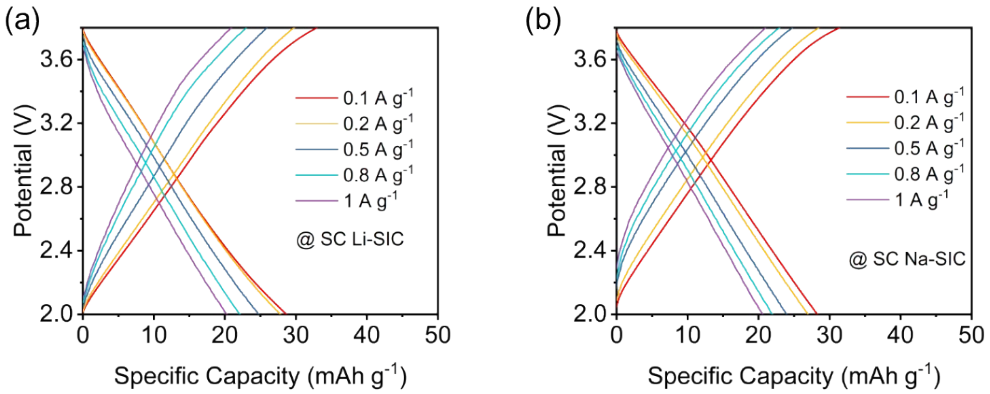

Figure S29. The charge/discharge profiles of (a) Li-SIC and (b) Na-SIC with SC anodes at the different current densities from 0.1 to 1 A g^{-1}.