Supporting Information

The crystal structures of Si and Si@cPAN were characterized by XRD. As shown in **Figure S1**, the diffraction peaks of the two materials correspond well to the crystal planes of crystalline silicon. Due to the low content and amorphous properties of the polymer, there is no additional diffraction peak in the diffraction pattern of Si@cPAN, which proves that the coated Si@cPAN still maintains the same crystal structure as Si.

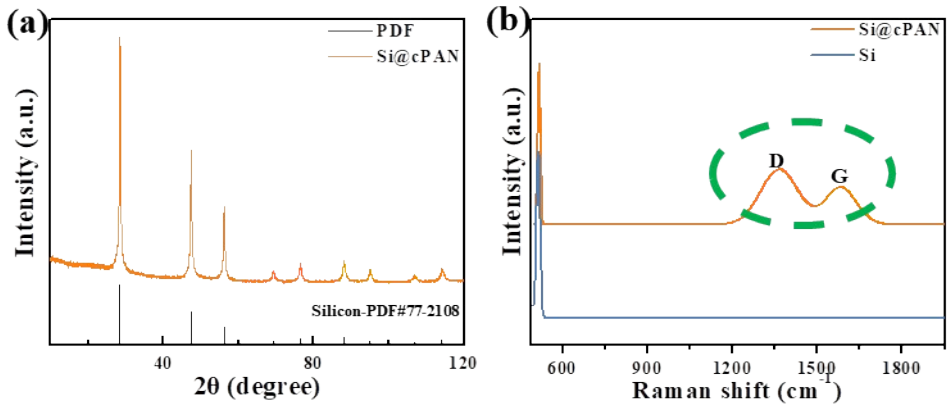

Figure S1. (a) XRD patterns of Si@cPAN. (b) Raman spectra of Si and Si@cPAN.

The cycling performance of Si@cPAN@CMC electrodes treated at different temperatures is shown in Figure S2a. As discussed in Figure 1, PAN undergoes structural and property transformations with the temperature increasing. Si@PAN-300@CMC electrode exhibits a stable cycle due to the favorable elasticity of PAN treated at 300°C, however, it exhibits a low capacity after activation due to the poor conductivity and not contributing to capacity of PAN treated at 300°C. PAN treated at 500°C, due to its excellent conductivity and less non-capacity components, leads to a higher capacity for Si@PAN-500@CMC electrode but suffers from rapid decay

because of the reduced elasticity. In contrast, Si@cPAN-400@CMC with a balanced elasticity and conductivity demonstrated superior capacity and cycling stability even at a current density of 2 A g^{-1}.

Cyclic voltammetry (CV) was used to characterize the electrochemical reaction and kinetics of the materials. As shown in **Figure S2**. Five cycles were performed at a 0.1 mV s^{-1} within the range of 0−1.2 V without activation. The sharp reduction peak near 0.01 V in the first cathode scan is due to the lithiation of crystalline silicon. Two oxidation peaks observed at 0.37 V and 0.53 V during initial and subsequent anode scans were attributed to delithiation of the Li-Si alloy. On the following cathode scan, a new reduction peak appeared at 0.18 V, which was attributed to reversible lithiation of amorphous silicon.

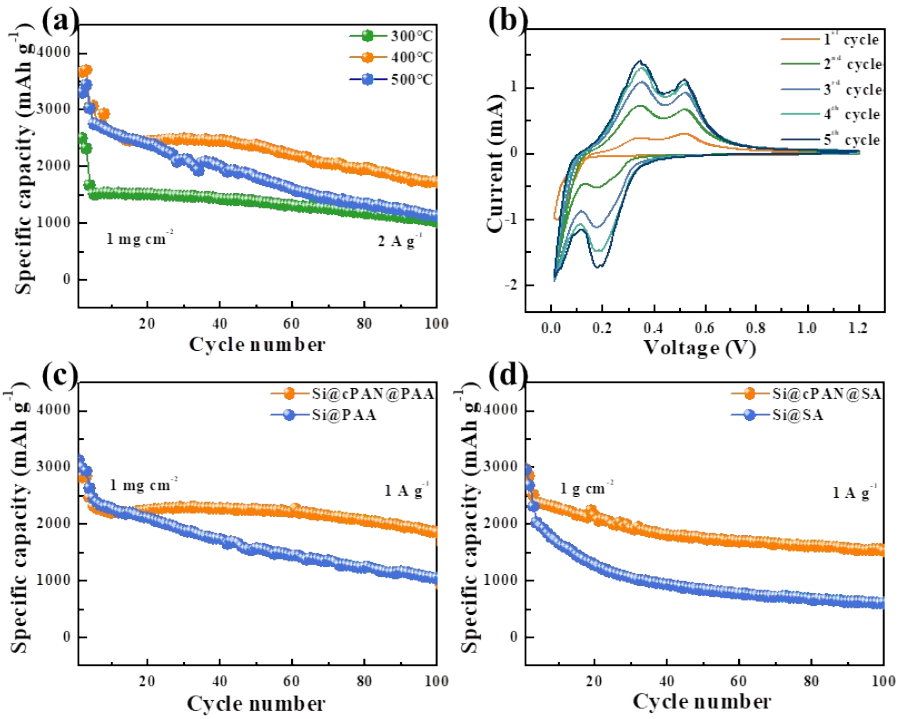

Figure S2. (a) Cycling performance of Si@cPAN@CMC electrodes treated at 300°C, 400°C, 500°C at 2 A g^{-1} for 100 cycles. (b) CV curves of Si@cPAN@CMC electrode for different cycles. Cycling performance of (c) Si@PAA and Si@cPAN@PAA

electrodes, (d) Si@SA and Si@cPAN@SA electrodes at 1 A g^{-1} for 100 cycles.

Thermal Gravimetric (TG) results presented in Figure S3a reveals that Si content constitutes 95.7% in Si@cPAN, signifying that cPAN content comprises 4.3% in Si@cPAN. Consequently, the Si@cPAN@CMC electrode by mixing Si@cPAN, carbon black, and CMC with a mass ratio of 8.4:1:0.6, which can ensure that the Si content occupies 80% within the electrode, and the cycling performance is shown in Figure S3b. And in order to further demonstrate the effect of cPAN coating, the electrochemical performance of Si@cPAN electrode without any binders is exhibited as Figure S4. To ensure the same Si contents in Si@CMC and Si@cPAN@CMC electrodes,

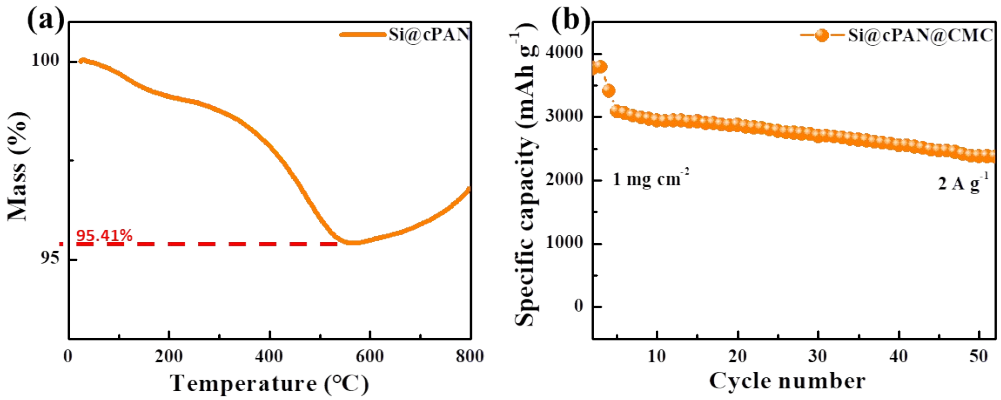

Figure S3. (a) TG curve of Si@cPAN particles. (b) Cycling performance of the Si@cPAN@CMC electrode by mixing Si@cPAN, carbon black, and CMC with a mass ratio of 8.4:1:0.6 .

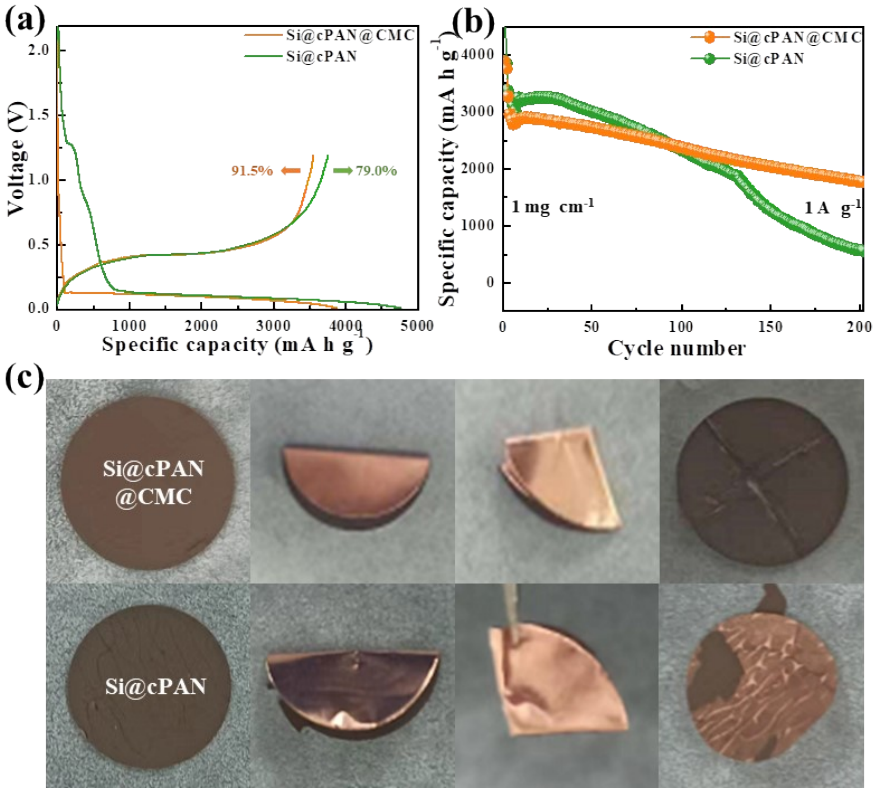

Figure S4. (a) The charge-discharge profiles and (b) cycling performances of Si@cPAN@CMC and Si@cPAN electrodes for 200 cycles. (c) Electrode photo of Si@cPAN@CMC and Si@PAN electrodes with a mass loading of 3.0 mg cm^{-2}

Table S1. Fitting parameters of EIS curves

	Si@CMC R_{SEI}/Ω	Si@CMC R_{ct}/Ω	Si@cPAN@CMC R_{SEI}/Ω	Si@cPAN@CMC R_{ct}/Ω
10 cycles	64.61	46.31	14.82	11.48
20 cycles	71.57	53.33	25.90	28.27
50 cycles	89.72	80.46	39.31	31.37

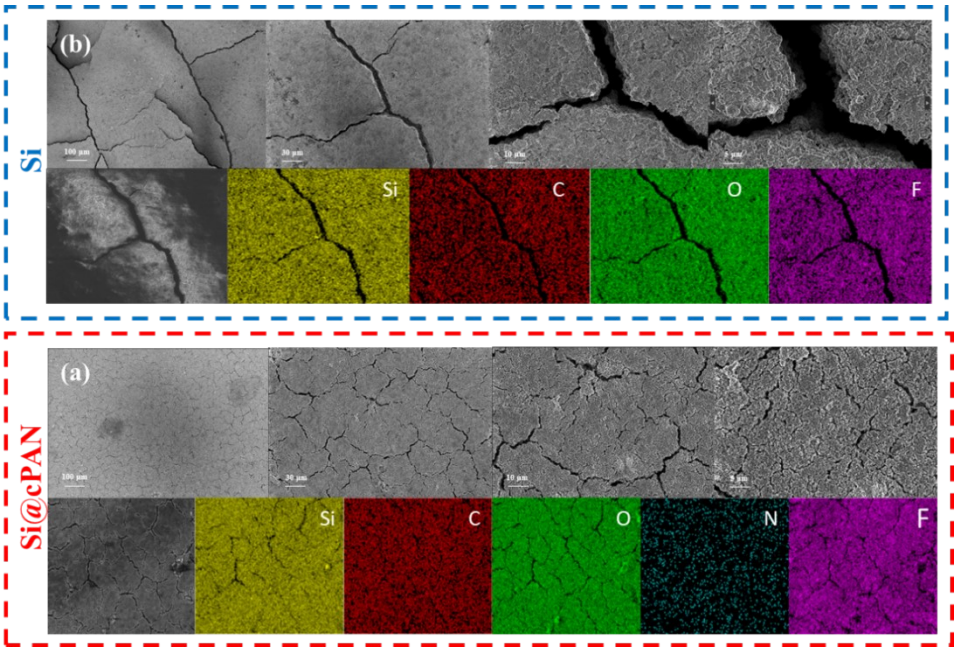

Figure S5. SEM images and corresponding EDS mappings of (a) Si@CMC and (b) Si@cPAN@CMC electrodes after 50 cycles at 1 A g^{-1}

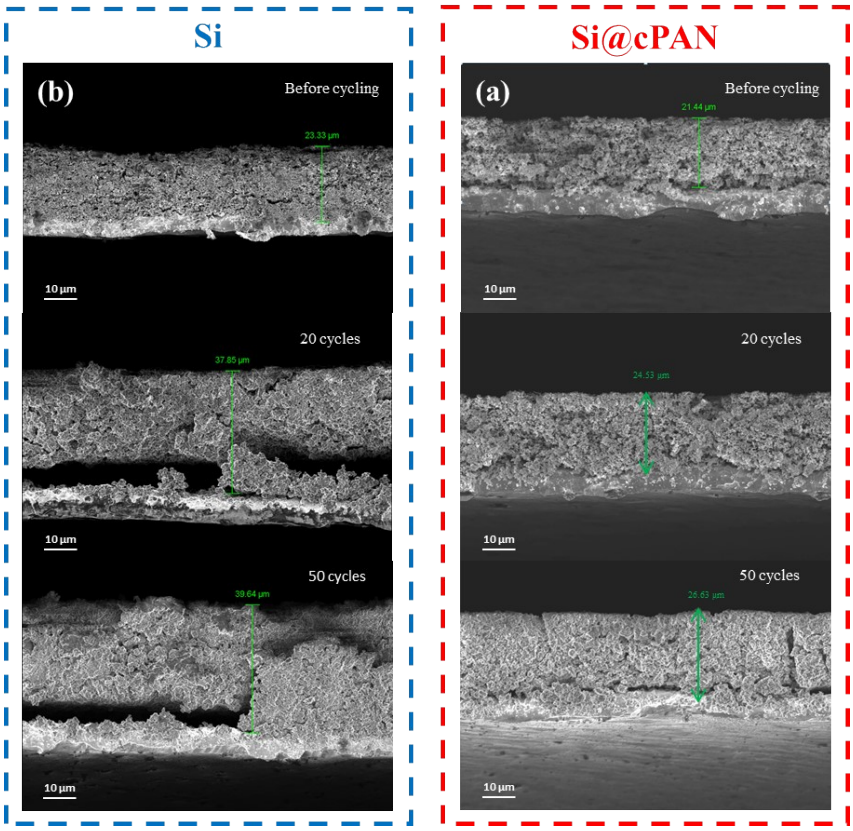

Figure S6. Cross-sectional SEM images of (a)Si@cPAN@CMC electrodes (b) Si@CMC electrodes before cycling and after 20 and 50 cycles at 1 A g^{-1}

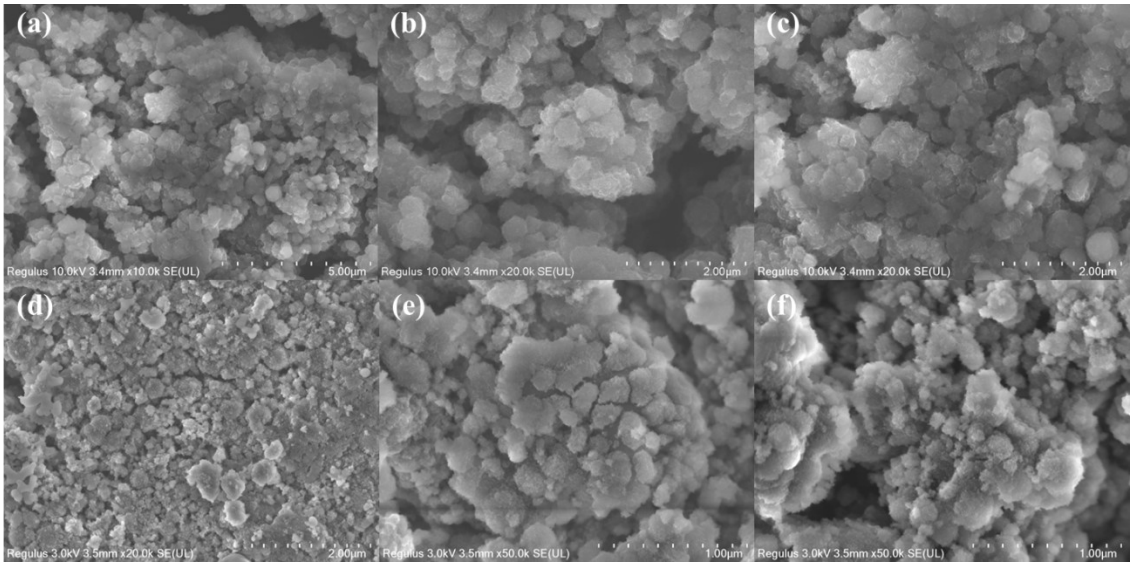

Figure S7. SEM images of (a-c) Si@cPAN@CMC and (d-f) Si@CMC particles after 50 cycles at 1 A g^{-1}

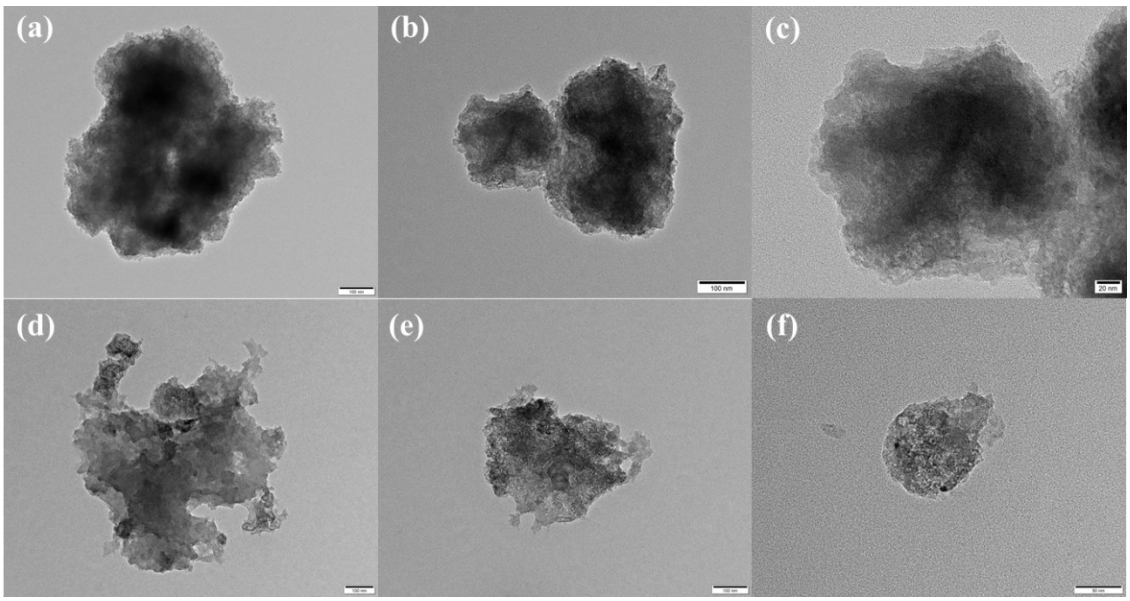

Figure S8. TEM images of (a-c) Si@cPAN@CMC and (d-f) Si@CMC particles after 50 cycles at 1 A g^{-1}

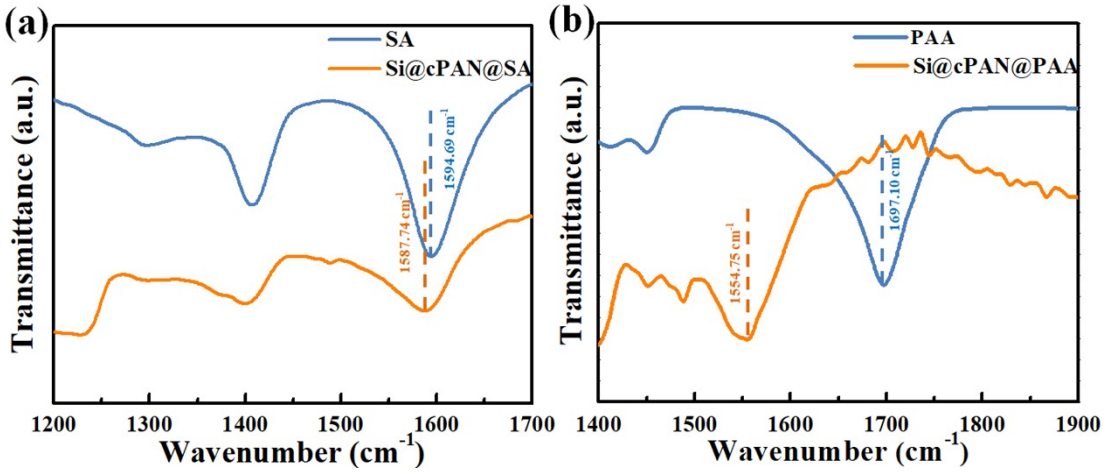

Figure S9. FTIR spectra of (a) SA and Si@cPAN@SA, (b) PAA and Si@cPAN@PAA electrode.