

## Supporting Information

### Multiregion Janus-Featured Cobalt Phosphide/Cobalt Composite for Highly Reversible Room-Temperature Sodium-Sulfur Batteries

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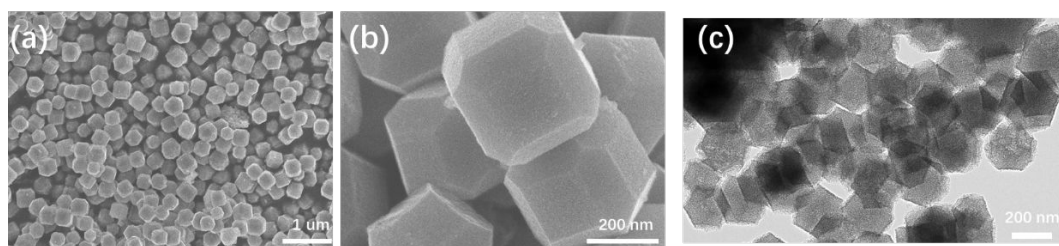
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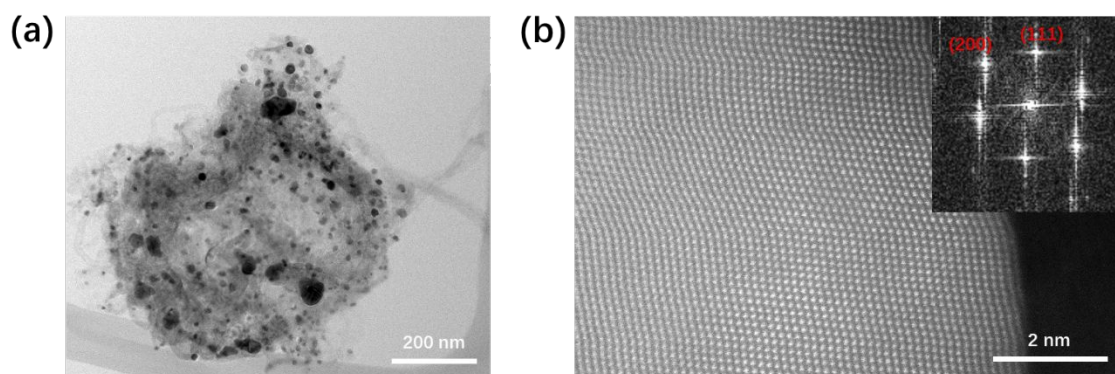
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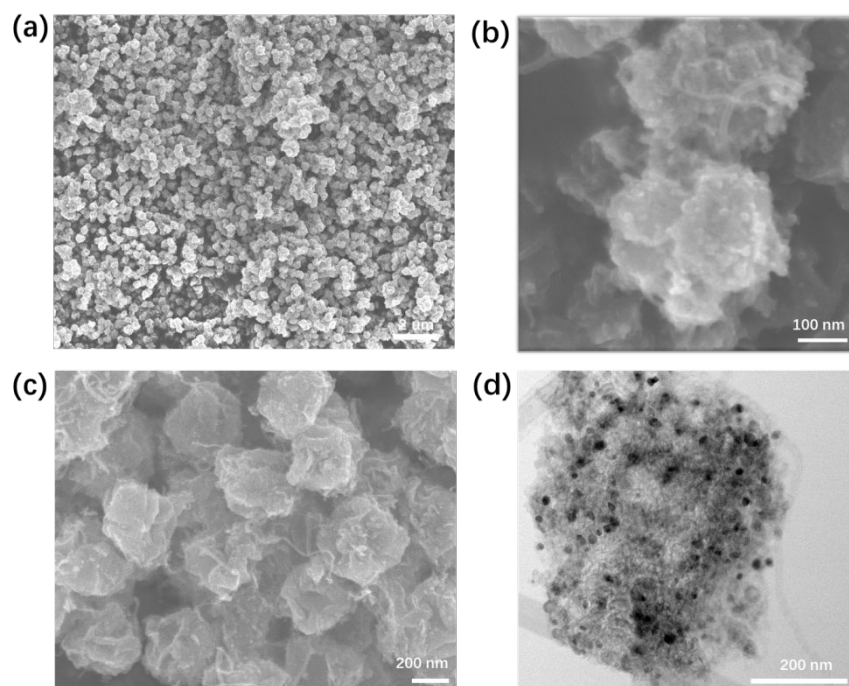
## Supplementary Figures



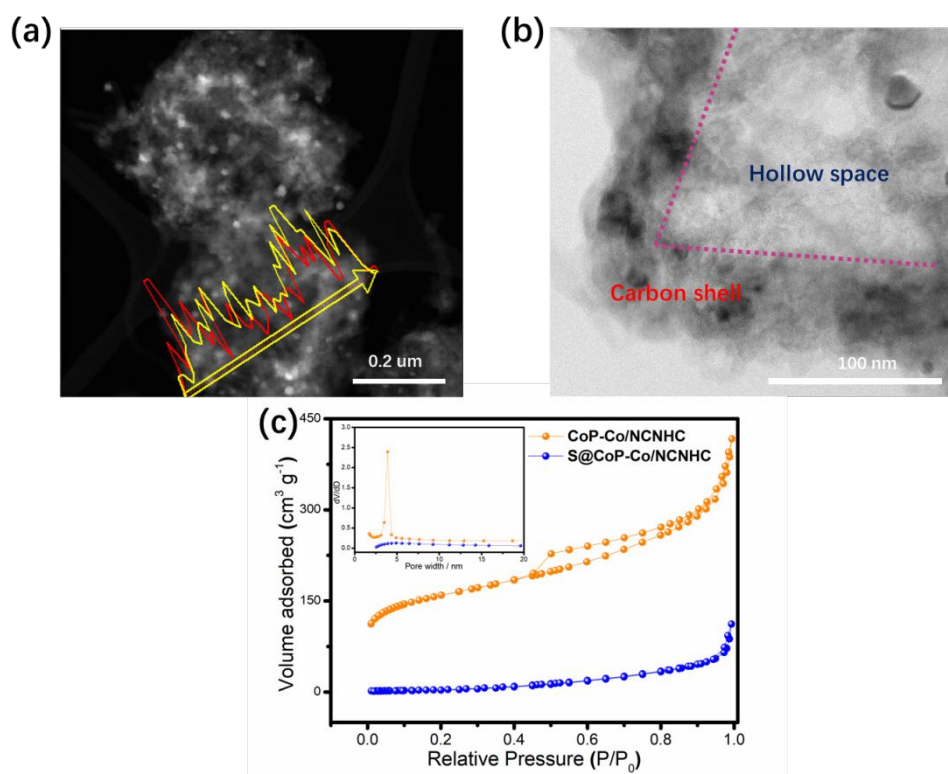
**Figure S1.** (a) SEM, (b) magnified SEM, and (c) TEM images of pure ZIF8.



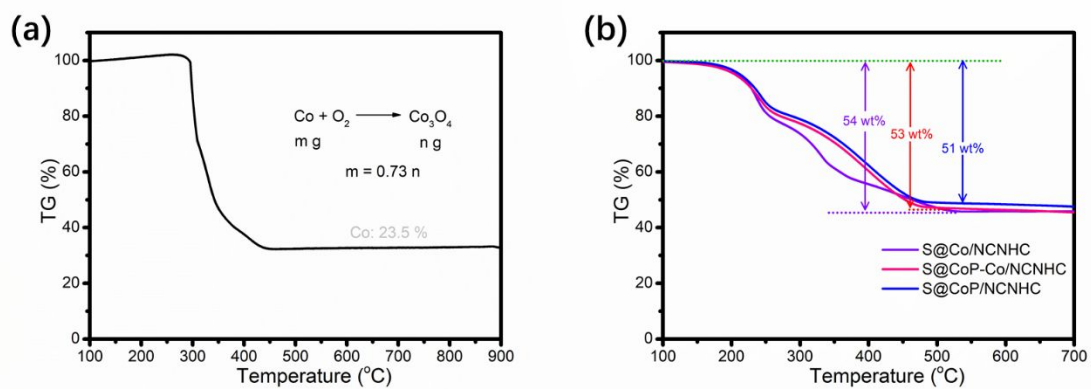
**Figure S2.** (a) STEM and (b) HAADF-STEM images of the Co/NCNHC composite. Inset in (b): FFT of the selected area.



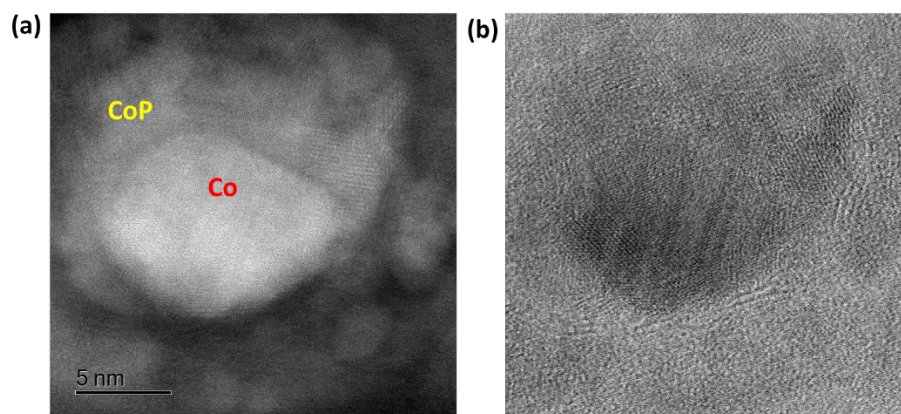
**Figure S3.** (a and b) SEM images of  $\text{Co}_3\text{O}_4\text{-Co/NCNHC}$ . (c) SEM and (d) STEM images of the  $\text{CoP-Co/NCNHC}$  composite.



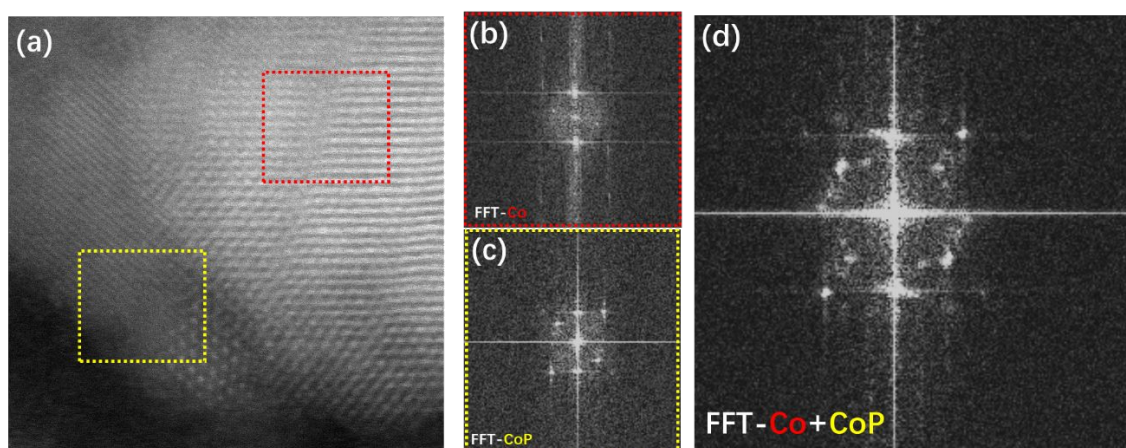
**Figure S4.** (a) Line scanning EDS spectra of S@CoP-Co/NCNHC composite (red and yellow lines refer to sulfur and carbon, respectively). (b) Enlarged STEM image of S@CoP-Co/NCNHC composite. (c) N<sub>2</sub> absorption/desorption isotherms and pore size distribution (inset) for the CoP-Co/NCNHC matrix and S@CoP-Co/NCNHC composite.



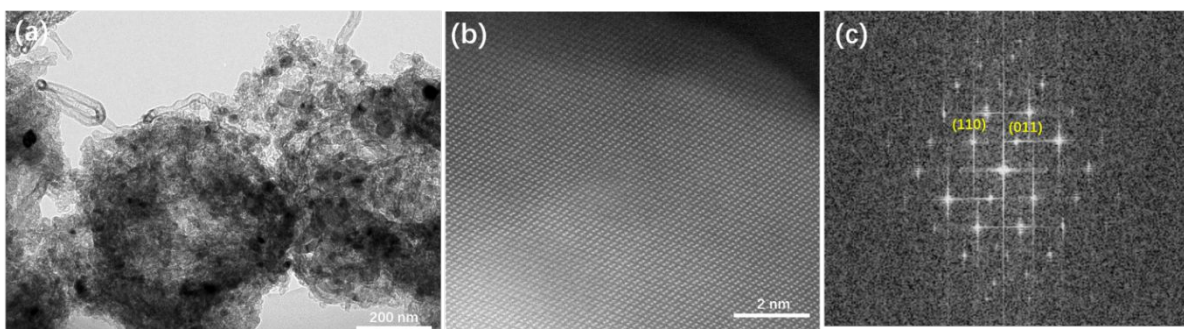
**Figure S5.** Thermogravimetry (TG) curves of (a) the Co/NCNHC in air at a heating rate of  $5^\circ\text{C min}^{-1}$  and (b) the S@CoP-Co/NCNHC, S@Co/NCNHC, and S@CoP/NCNHC in Ar at a heating rate of  $5^\circ\text{C min}^{-1}$ .



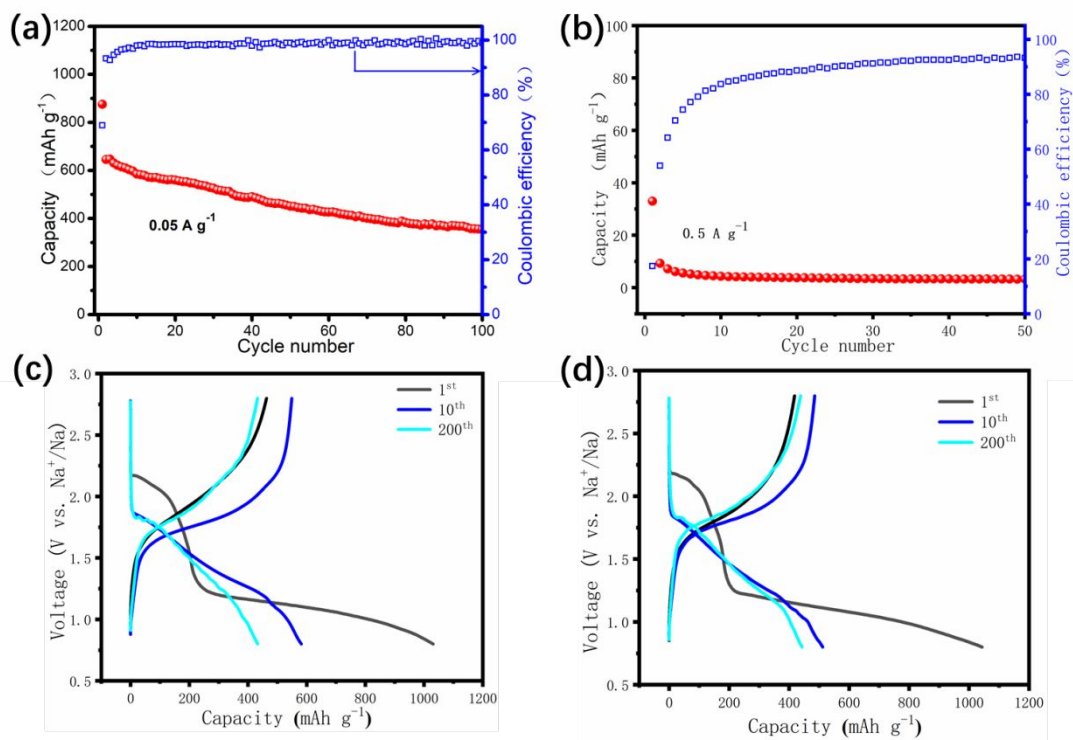
**Figure S6.** (a) HAADF- and (b)ABF-STEM images of an individual multiregion Janus-featured CoP-Co composite of the S@CoP-Co/NCNHC composite.



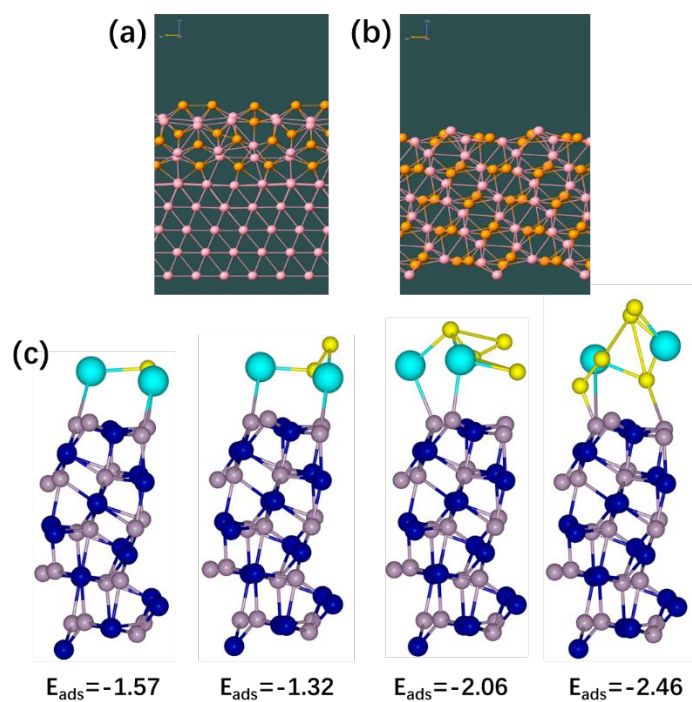
**Figure S7.** (a) HAADF-STEM image of the CoP-Co heterostructure. Corresponding FFT patterns of (b) Co, (c) CoP, and (d) Co+CoP



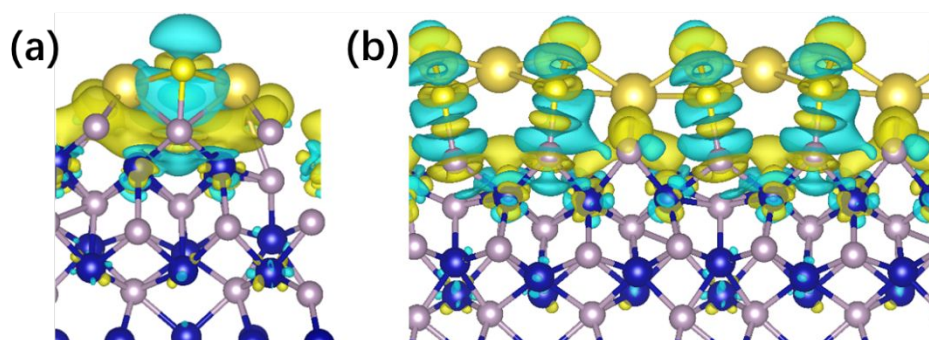
**Figure S8.** (a) STEM and (b) HAADF-STEM images, and (c) FFT pattern of the S@CoP/NCNHC composite.



**Figure S9.** Cycling performance of (a) S@NCNHC at 0.05 A g<sup>-1</sup> and (b) CoP-Co/NCNHC. Charge/discharge curves of (c) S@Co/NCNHC and (d) S@CoP/NCNHC at 0.1 A g<sup>-1</sup>.



**Figure S10.** Optimized adsorption configurations of (a) CoP-Co and (b) CoP surface. (c) Atomic conformations of Na<sub>2</sub>S<sub>x</sub> species adsorbing on CoP surface with corresponding adsorption energy.



**Figure S11.** Charge density difference of (a)  $\text{Na}_2\text{S}$  and (b)  $\text{Na}_2\text{S}_4$  adsorbing on CoP-Co surface as defined by:  $\Delta\rho = \Delta\rho_{\text{CoP-Co-Na}_2\text{S}_x} - \Delta\rho_{\text{CoP-Co}} - \Delta\rho_{\text{Na}_2\text{S}_x}$ . The yellow and green area represents accumulation and depletion, respectively.



**Figure S12.** FESEM image of the cross-section for S@CoP-Co/NCNHC electrode.

## Supplementary Tables

**Table S1** | Comparison with previous room-temperature Na-S batteries in electrochemical properties

Active material	Loadin g sulfur	Electrolyte	Current collector	Working current	Cycle number	Retained discharge capacity	Ref
S@CoP-Co/NCNH C	53%	1M NaClO <sub>4</sub> in EC:PC with 3%FEC	Al foil	1A g <sup>-1</sup>	700	448mAh g <sup>-1</sup>	This work
S@C	35%	1M NaPF <sub>6</sub> with 0.25M NaNO <sub>3</sub> in TEGDME	SS discs	1.6A g <sup>-1</sup>	1500	300mAh g <sup>-1</sup>	
c-PANS	31%	0.8M NaClO <sub>4</sub> in EC:DEC	Al foil	0.22A g <sup>-1</sup>	500	180mAh g <sup>-1</sup>	9
TeS@pPAN	47.7%	1M NaClO <sub>4</sub> in EC:DEC with 10%FEC	Al foil	0.5A g <sup>-1</sup>	600	970mAh g <sup>-1</sup>	13
S/Ni-MOF-2D	48.6%	1M NaClO <sub>4</sub> in EC:PC with 5%FEC	Al foil	1.6A g <sup>-1</sup>	1000	374mAh g <sup>-1</sup>	19
rGO/VO <sub>2</sub> /S	40%	1M NaClO <sub>4</sub> in TEGDME	Al foil	0.32A g <sup>-1</sup>	600	300mAh g <sup>-1</sup>	21
SeS@pPAN	44.6%	1M NaTFSI in TEGDME with 5%FEC	Al foil	0.4A g <sup>-1</sup>	200	600mAh g <sup>-1</sup>	42
S@CNT/NPC	35%	1M NaClO <sub>4</sub> in EC:DEC	Al foil	0.8A g <sup>-1</sup>	500	410mAh g <sup>-1</sup>	45
S@HCS/MoS <sub>2</sub>	44%	1M NaClO <sub>4</sub> in TEGDME	Al foil	1.6A g <sup>-1</sup>	1000	220mAh g <sup>-1</sup>	46
S/(CNT@MPC	40%	1M NaClO <sub>4</sub> in EC:PC	Al foil	0.16A g <sup>-1</sup>	20	1000mAh g <sup>-1</sup>	6