Enhanced Na⁺ pseudocapacitance in P-S co-doping carbon anode: raising from the surface modification by sulfur and phosphorus with C-S-P coupling

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Figure S1. the digital picture of the synthetic process of PSC



Figure S2. the resultant of CS_2 and RP after different temperature for 24 h (before washing).



Figure S3. (a), (b), (c) FESEM images; (d), (e), (f) TEM images of the PSC obtained at 400, 500, 600 °C. (g) XRD patterns, (h) N2 adsorption desorption isotherms and (i) the P and S contents of the PSC obtained at 400, 500, 600 °C.



Figure S4. (a)XRD pattern and (b) Raman spectra of SC.



Figure S5. The survey XPS spectrum of PSC obtained at 500°C



Figure S6. (a) Voltage profiles of SC at 100 mA g^{-1} , (b) cycling performance and coulombic efficiency of SC at 100 mA g^{-1} , (c) cycling performance and coulombic efficiency of PSC at 100 mA g^{-1} , (d) cycling performance and coulombic efficiency of SC, PSC at 500 mA g^{-1} .

Table S1. The atomic ratios of P to S in the raw materials and the practical atomic ratios of P to S doped in the PSCs (obtained at 500°C for 24h).

RP(mg):CS ₂ (μl)	100:100	100:150	100:250	100:300
P/S(atomic ratio) In the raw materials	1:1	1:1.5	1:2.5	1:3
P/S(atomic ratio) In the PSC	0.85	0.66	0.47	0.48



Figure S7. (a), (b), (c) the charge and discharge voltage profiles, (d) the long cycle performances of PSCs obtained at 500°C with the atomic ratios of P to S in the raw materials is 1:1, 1:1.5, 1:3.

The atomic ratio of P to S doped in PSC can be controlled by adjusting the atomic ratio of P to S in the raw materials. Specifically, 100mg RP and different volumes (100, 150, 250 and 300 μ l) of CS₂ are sealed in the quartz tube then sintered at 500 °C for 24 h. The practical P/S atomic ratios in PSCs are shown in Table S1. The charge and discharge curves of these PSCs are obtained at 100 mA g⁻¹ as shown in Figure S7, it is

found that the PSCs deliver the initial discharge/charge capacities of 1079.8/442, 988.4/452.0, 972.6/519.9 and 859.2/486.2 mAh g⁻¹ with the initial coulombic efficiency of 40.9%, 45.7%, 53.4% and 56.6%, corresponding the atomic ratios P to S of 0.85, 0.66, 0.47 and 0.48 in the PSCs, respectively. Obviously, the capacity of PSC increases with the P/S atomic ratio decreases. When the P/S atomic ratio lowers to the minimum value of 0.47 (corresponding to 100 mg RP with 250 μ l CS₂ in the raw materials), PSC can reach the highest capacity of 519.9 mAh g⁻¹ and excellent cycling (Figure S7d).



Figure S8. The SEM images of PSC electrode before and after 1000 cycles at 1A g⁻¹



Figure S9. The Nyquist plots of the PSC after various cycles at 50mA g⁻¹



Figure S10. the optimized structure of (a) single S-doped carbon, (b) single P-doped carbon after adsorbed with sodium ion and the corresponding differential charge density (DCD) maps. (For the structure, Gray, yellow, pink, red represents C, S, P, Na ions,

respectively; For DCD, yellow and cyan isosurfaces represent the charge accumulation and depletion in the system, respectively.)



Figure S11. The XRD patterns of PSC tested at different charge-discharge states.



Figure S12. (a) the XRD pattern, (b) SEM, (c) TEM images of the NVP@C.



Figure S13. (a) the cycling performance of NVP@C at 100mA g⁻¹, (b) the chargedischarge curves of NVP@C at 100 mA g⁻¹.