

Supplementary material

Dual-Stabilization Strategy Enabled High-Entropy Sulfide@Carbon Spheres for High-Performance Sodium Storage

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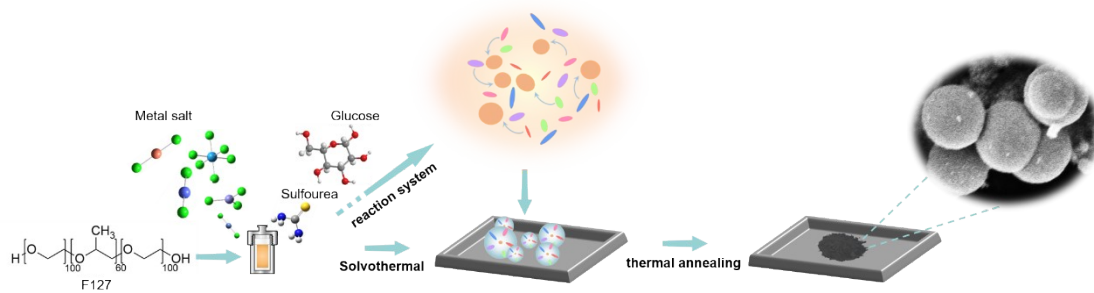


Figure S1. Schematic diagram of the preparation process for the (FeCoNiCuMo)₂S₂/C.

Formation Mechanism of High-Entropy Sulfide/Carbon Composite (FeCoNiCuMo)₂/C

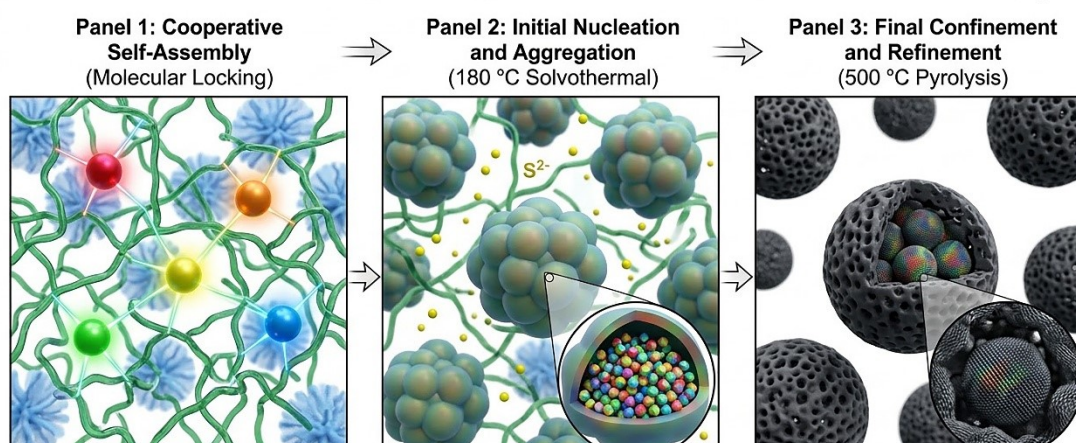


Figure S2. Schematic illustration of the multi-stage formation mechanism for (FeCoNiCuMo)₂/C high-entropy sulfide spheres. Panel 1: Cooperative Self-Assembly (Molecular Locking). The process initiates with the formation of a stable organic–inorganic supramolecular network through hydrogen bonding between F127 surfactant micelles and glucose molecules. Simultaneously, the five transition metal cations (Fe²⁺, Co²⁺, Ni²⁺, Cu²⁺, and Mo⁵⁺) are coordinated and “locked” onto the multi-dentate ligands of the polymer chains. This dual-interaction mechanism ensures an atomic-scale homogeneous distribution of the multi-element precursors, effectively preventing premature elemental segregation at the molecular level. **Panel 2: Initial Nucleation and Crystallization (180 °C Solvothermal Stage).** During the solvothermal reaction, the slow decomposition of thiourea provides a controlled release of S²⁻ ions. The high configurational entropy of the five-metal system (1.607R) suppresses the formation of individual binary sulfides, favoring **synchronous nucleation** of entropy-stabilized grains. As evidenced by Figure S12, the product at this stage consists of spherical micro-aggregates that already exhibit discernible diffraction peaks, confirming that the initial crystallization of the high-entropy sulfide core is successfully initiated during the solvothermal process. **Panel 3: Mesoscale Carbon Confinement and Structural Refinement (500 °C Pyrolysis).** Upon subsequent pyrolysis, the cross-linked polymer matrix is converted into a rigid and continuous carbon framework. This carbon scaffold provides intense spatial confinement that serves as a physical “cage”, effectively preventing the further aggregation or long-range diffusion of the pre-formed sulfide nuclei. This mechanism strictly limits the final nanoparticle size to approximately 1 nm while facilitating the final solid-state crystallization of the dual-phase high-entropy framework, ensuring long-term structural and interfacial stability

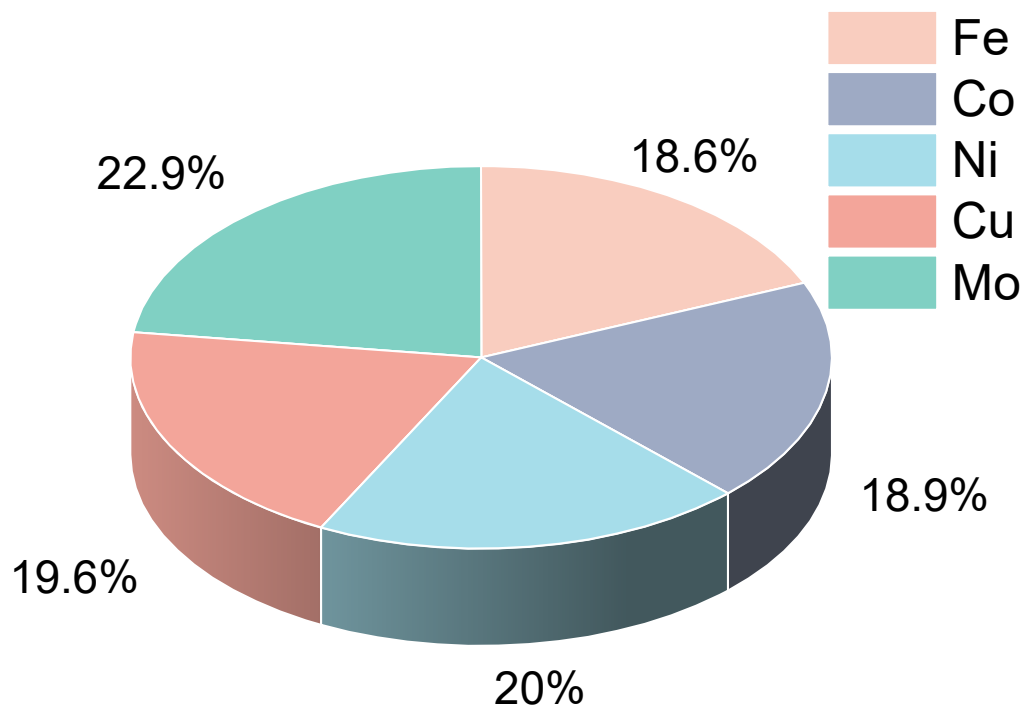


Figure S3. The ICP-OES of $(\text{FeCoNiCuMo})\text{S}_2/\text{C}$

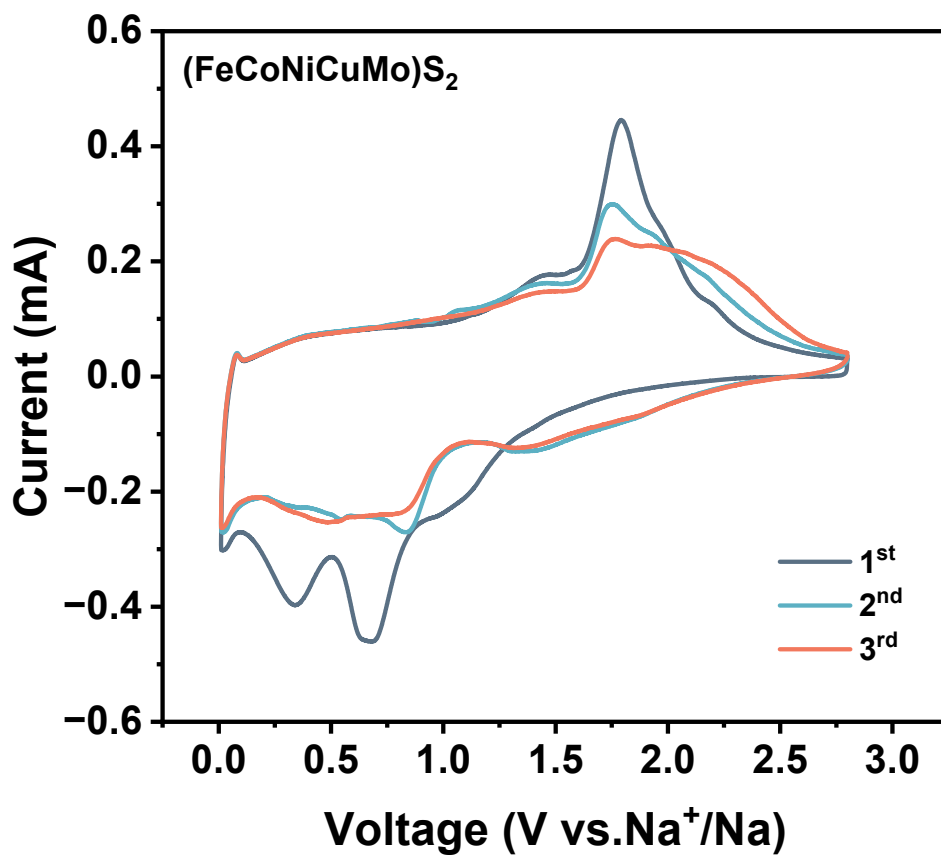


Figure S4. CV curves of $(\text{FeCoNiCuMo})\text{S}_2$.

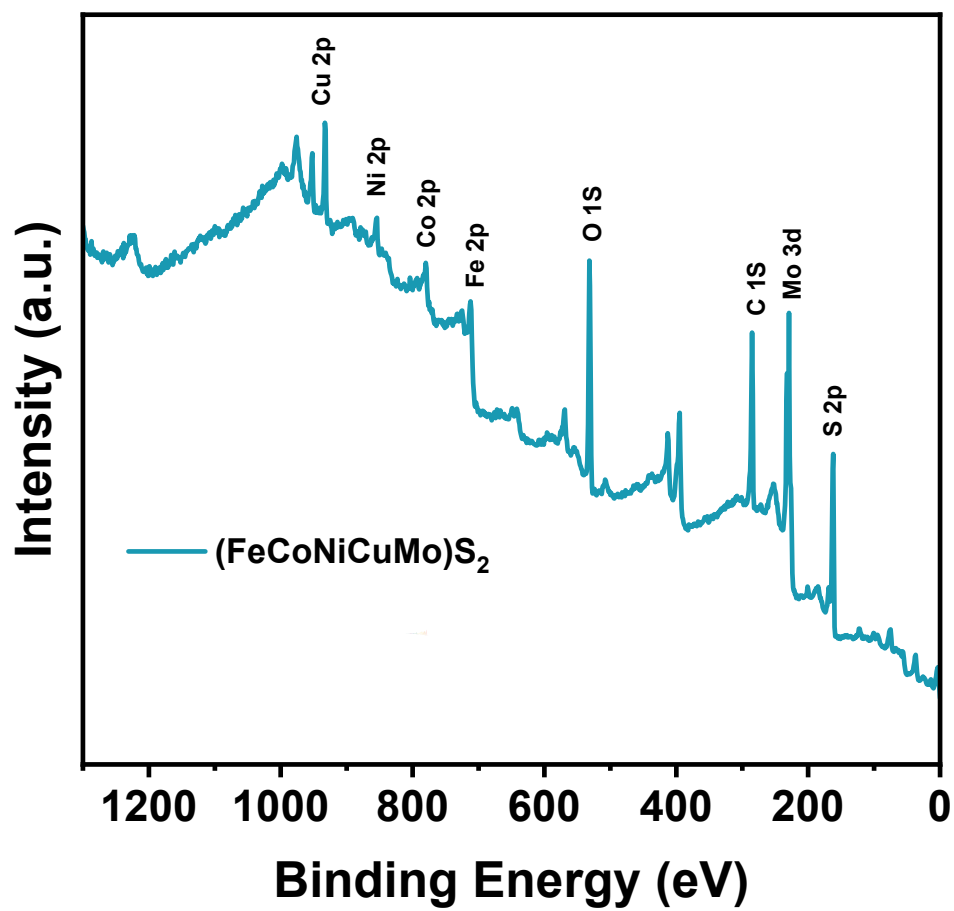


Figure S5. CV curves of $(\text{FeCoNiCuMo})\text{S}_2/\text{C}$.

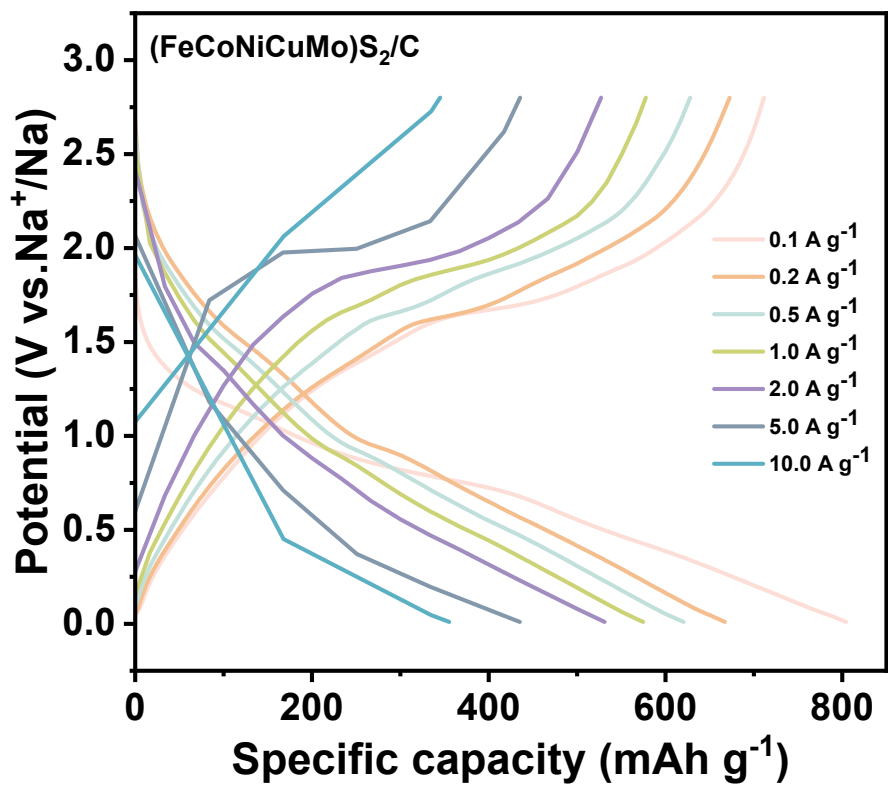


Figure S6. Rate capabilities of $(\text{FeCoNiCuMo})\text{S}_2/\text{C}$.

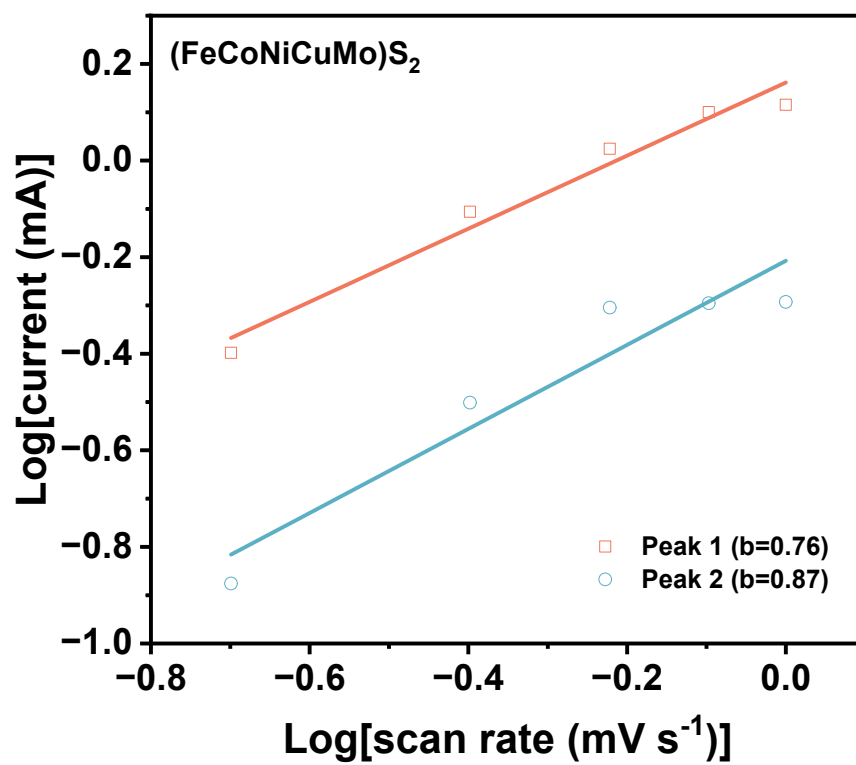


Figure S7. The linear correlation between the peak current (log *i*) and the sweep rate (log *v*) of

(FeCoNiCuMo)S₂.

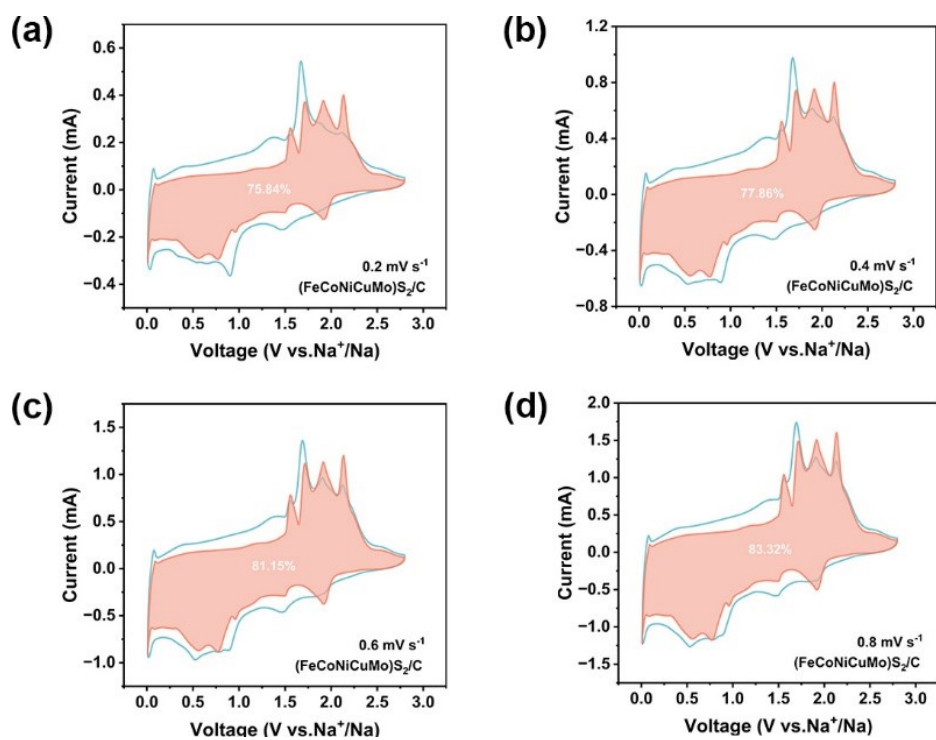


Figure S8. Capacitive contribution (shaded area) of the (FeCoNiCuMo)₂/C electrode at scan rates of: (a) 0.2, (b) 0.4, (c) 0.6, and (d) 0.8 mV s⁻¹. Shaded areas represent capacitive contributions, with corresponding percentages labeled on each subfigure.

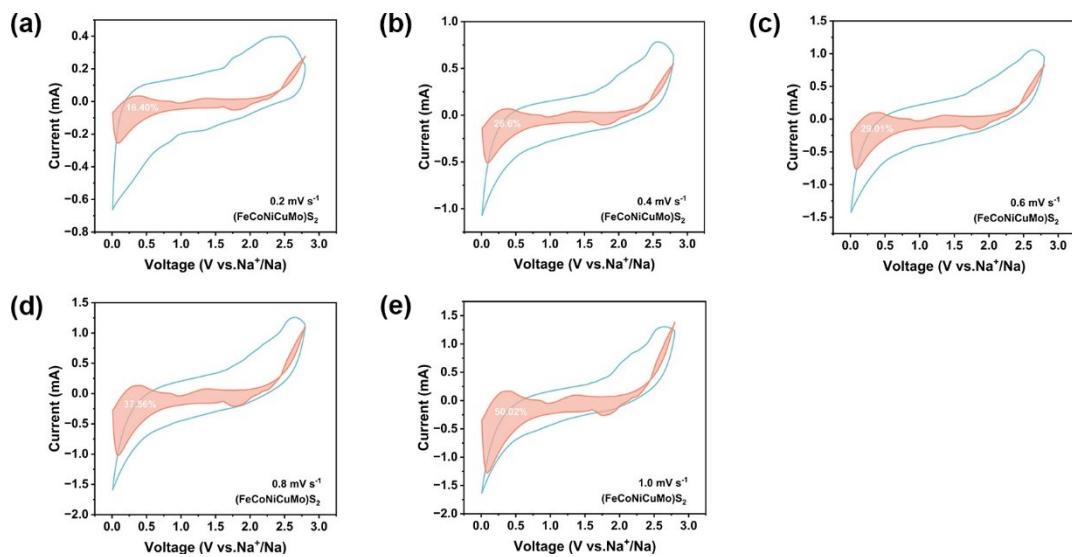


Figure S9. Capacitive contribution (shaded area) of the (FeCoNiCuMo)₂ electrode at scan rates

of: (a) 0.2, (b) 0.4, (c) 0.6, and (d) 0.8 mV s⁻¹ (e) 1.0 mV s⁻¹.

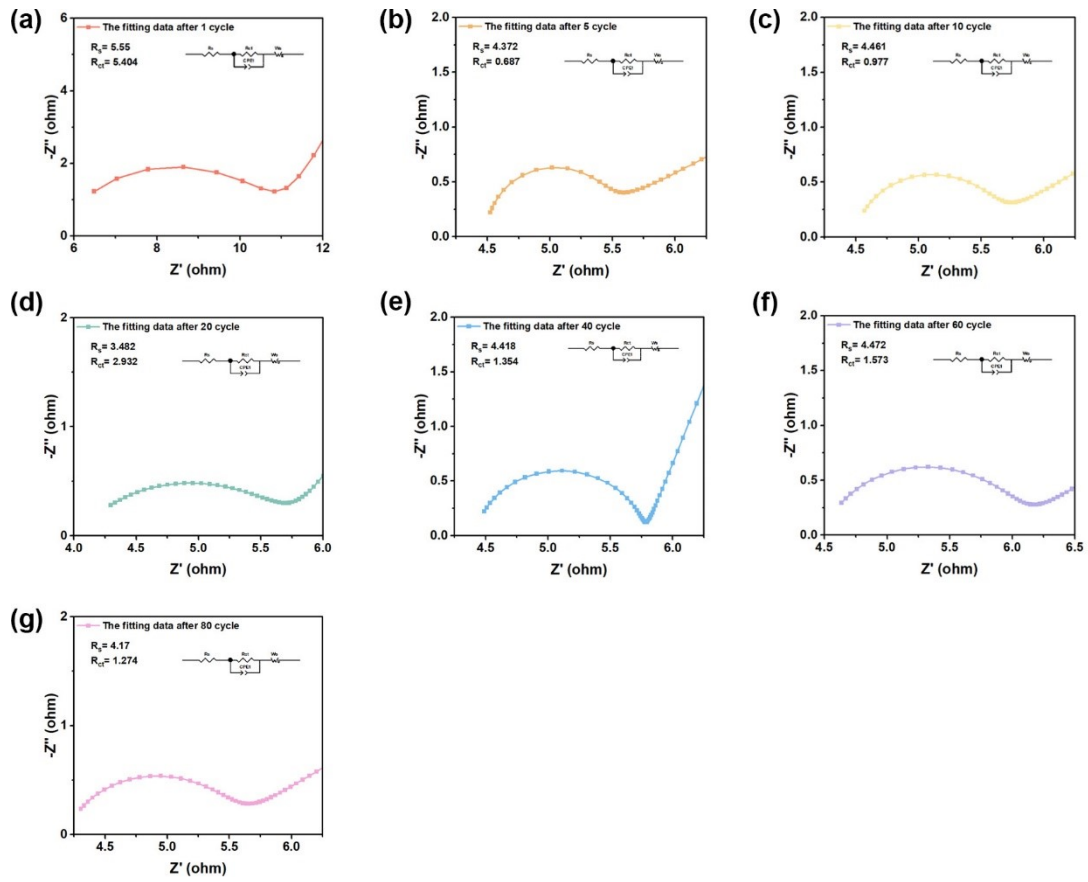


Figure S10. Electrochemical impedance spectroscopy (EIS) of the $(\text{FeCoNiCuMo})\text{S}_2/\text{C}$ electrode and the corresponding equivalent circuit model (inset), showing the evolution of the spectra after different numbers of charge-discharge cycles: (a) 1, (b) 5, (c) 10, (d) 20, (e) 40, (f) 60, and (g) 80 cycles.

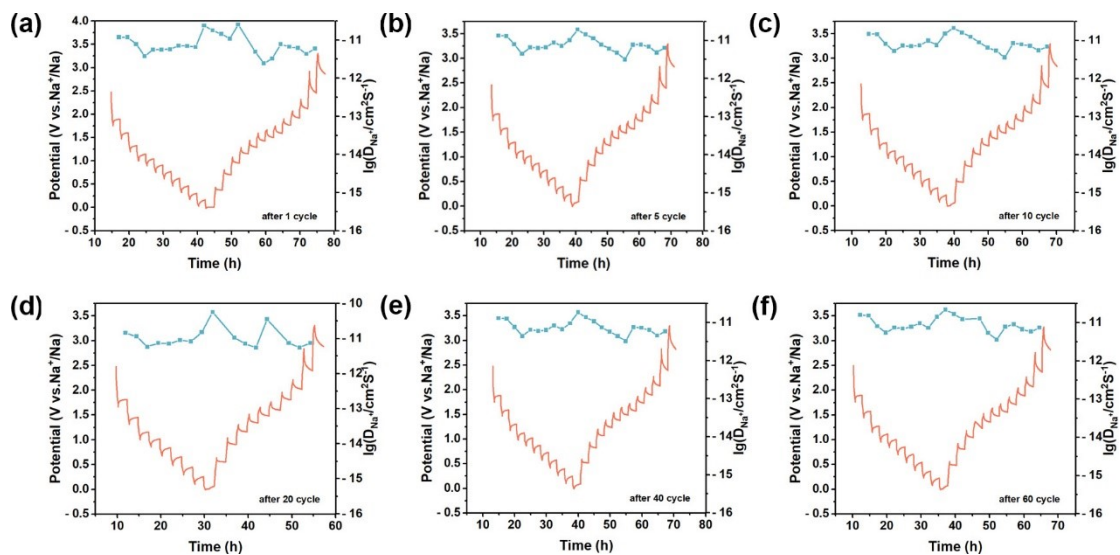


Figure S11. Potential response profiles from GITT measurements for the $(\text{FeCoNiCuMo})\text{S}_2/\text{C}$ electrode, showing the evolution of polarization and hysteresis upon cycling: (a) 1, (b) 5, (c) 10, (d) 20, (e) 40, and (f) 60 cycles.

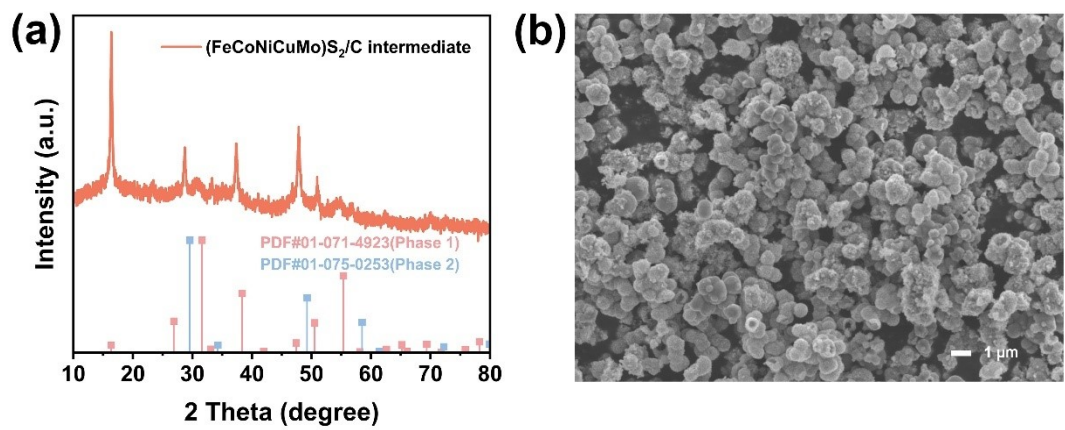


Figure S12. (a) XRD pattern and (b) SEM image of the (FeCoNiCuMo)₂S₂/C intermediate obtained after the solvothermal reaction.

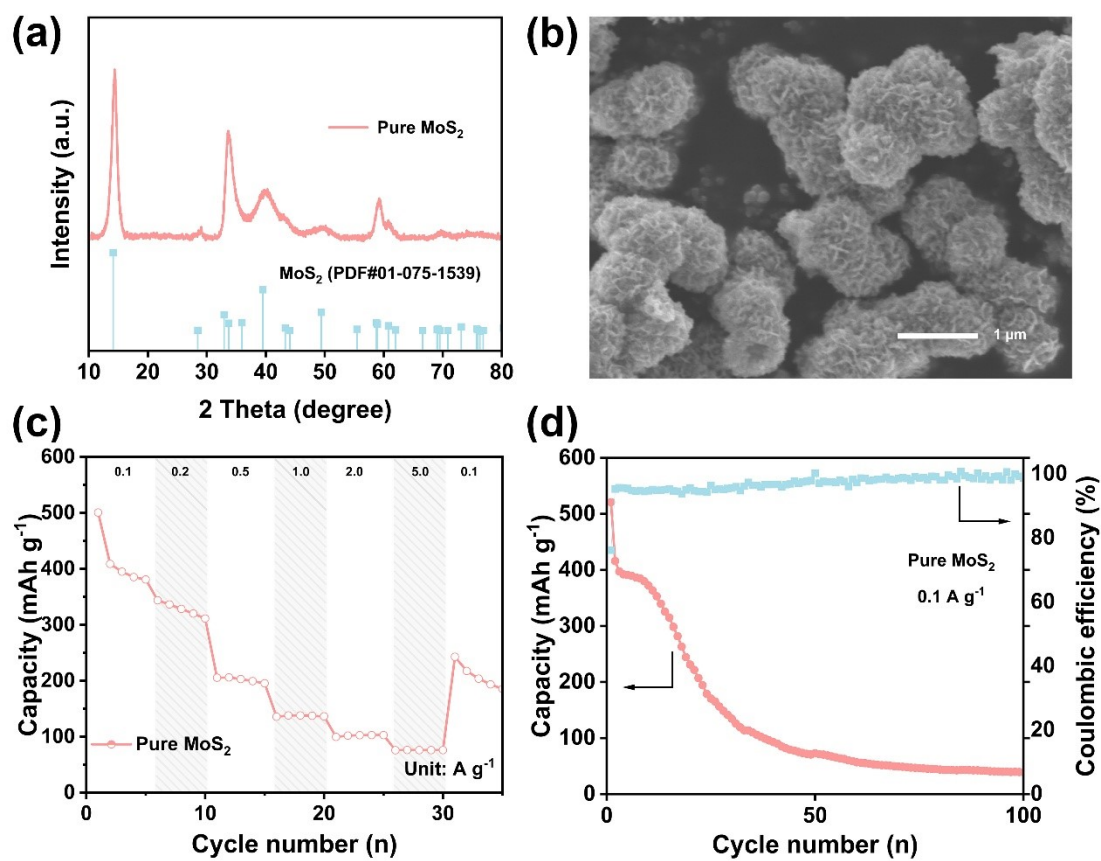


Figure S13. (a) XRD pattern and (b) SEM image of the pure MoS₂. (c) Rate capability and (d) cycling performance at 0.1 A g⁻¹ of the pure MoS₂ electrode.

Table S1. The sodium storage performance of similar metal sulfides materials reported in the literature.

Samples	Cyclic stability			Rate capacity	Ref.
	Current density (A g ⁻¹)	Capacity (mAh g ⁻¹)	Cycle number		
(FeCoNiCuMo) S ₂ /C	0.1	757	80	711,672,627,577,527, 437 and 345 mAh g ⁻¹ at 0.1, 0.2, 0.5, 1.0, 2.0,5.0 and 10.0 A g ⁻¹ , respectively	This Work
	1	468	600		
SnSbMnBiTe	0.1	381.7	80	437.4, 416.5, 386.2, 381.0 and 351.0 mAh g ⁻¹ , at 0.2, 0.5,1.0, 2.0 and 5.0 A g ⁻¹ , respectively	[1]
SnCo@C	0.1	276.2	120	-	[2]
Bi ₈₀ Sn ₅₀ Sb ₅₀ Cu ₈₀ Al ₈₀ @PCFs	5	235	500	202, 190,185 and 180 mAh g ⁻¹ at 10, 15, 20and 25.0 A g ⁻¹ , respectively	[3]
NiS ₂ /NG	0.1	676.0	100	615.1, 594.6, 569.7, 555.8, and 545.0 mAh g ⁻¹ at 0.1, 0.2, 0.5, 1, 1.5, and 2.0 A g ⁻¹ , respectively	[4]
WZSC (WS _{2-x} / ZnS@C)	1	329.1	500	468.2, 411.1, 352.5, 296.4, 251.4, 206.4, 180.7, and 181.9 mAh g ⁻¹ at 0.1, 0.2, 0.5, 1, 2, 5, 10, 20 A g ⁻¹ , respectively	[5]

CC-ZnS/CNT	0.1	730	200	634, 603, 522, 410, 317, and 295 mAh g ⁻¹ at 0.1, 0.2, 0.5, 1, 1.5, and 2 A g ⁻¹ , respectively	[6]
M-FeS@C+CNT.	1	366	860	505, 486, 471, 464, 454, 438, 418, 403, 380, and 332 mAh g ⁻¹ at 0.05, 0.1, 0.2, 0.3, 0.5, 1, 2, 3, 5, and 10 A g ⁻¹ , respectively	[7]
NiS ₂ @C@C	0.1	580.8	100	694, 671, 670, 631, 555, and 448 mAh g ⁻¹ at 0.05, 0.1, 0.2, 0.4, 0.8, and 1.6 A g ⁻¹ , respectively	[8]
Co ₃ S ₄ @PANI	0.2	252.5	100	295.5, 218.6, 189.3, 184.1 mAh g ⁻¹ at 200, 1000, 2000, 4000 mA g ⁻¹ , respectively	[9]
CuS-RGO-2	0.1	392.9	50	509.1, 383.3, and 370.2 mAh g ⁻¹ at 100, 250, and 500 mA g ⁻¹ , respectively	[10]

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