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Supporting Information

Designing Pinecone-like and Hierarchical Manganese Cobalt Sulfides for Advanced Supercapacitor Electrode

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Figure S1. TEM images of MnS (a) and CoS (b); b and c are manganese and sulfur EDX-elemental mapping images of MnS, e and f are cobalt and sulfur EDX-elemental mapping images of CoS.



Figure S2. (a) XPS curves of MnS and Mn-glycerate; (a) XPS curves of CoS and Co-glycerate.



Figure S3. EIS curves of MnS, CoS and MIX.



Figure S4. SEM (a) and TEM (b) photos of Mn-Co-glycerate.



Figure S5. TEM photos of MnCoS1 (a), MnCoS2 (b), MnCoS3 (c), MnCoS4 (d) and MnCoS5 (e).



Figure S6. (a, b, c, d) TEM, manganese, cobalt, sulfur EDX-elemental mapping images of one sphere in Mn-Co-S3. EDX curve (e) of image a. The Inserted table presents the weight percentage and atomic percentage of manganese, cobalt and sulfur elements in image a.



Figure S7. (a, b, c, d) TEM, manganese, cobalt, sulfur EDX-elemental mapping images of the extruded portion from the sphere of Mn-Co-S5. EDX curve (e) of image a. The inserted table presents the weight percentage and atomic percentage of manganese, cobalt and sulfur elements in image a.



Figure S8. CV curves of MnCoS1 (a), MnCoS2 (b), MnCoS3 (c), MnCoS4 (d) and MnCoS5 (e) at different scan rates.



Figure S9. GCD curves of MnCoS1 (a), MnCoS2 (b), MnCoS3 (c), MnCoS4 (d) and MnCoS5 (e) at different current densities.



Figure S10. N₂ adsorption desorption (a) and pore size distribution of all MnCoS samples (b).



Figure S11. SEM (a, b) and TEM (c, d) pictures of MnCoS4 after 5500 cycles at 10A/g, and EDX-elemental mapping images of (d).



Figure S12. XPS curves for Mn 2p (a) and Co 2p (b) of original MnCoS4 and MnCoS4 after 5000 cycles at 10 A/g.



Figure S13. CV (a) and GCD (b) curves of Ni foam and carbon cloth at 50 mV/s, CV (c) and EIS (d) curves of Ni foam, carbon cloth, Ni foam@MCS4 and carbon cloth@MCS4.

For clarification, in Figure S13 Ni foam@MCS4 is the exact same MnCoS4 electrode in the main manuscript and MnCoS4 powders were coated on the surface of carbon cloth without further pressing at 10 Mpa to fabricate carbon cloth@MCS4 which is different from the preparation process as Ni foam@MCS4, because all the carbon cloth@MCS4 would totally turn into fine powders if it were pressed at 10 Mpa. The compressive strength of carbon cloth is far worse than Ni foam.

Figure S13(a) shows that at 50 mV/s Ni foam and carbon cloth both reveal corresponding capacitive behavior to some extent, which could also been noticed in Figure S13(b) from their GCD curves. Taking Ni foam@MCS4 and carbon cloth@MCS4 into consideration, the electrochemical effect of Ni foam and carbon cloth could be neglected for their insignificant influence which has been demonstrated in Figure S13(c). The specific capacitance and oxidation behaviors of Ni foam and

carbon cloth during charge and discharge process contribute almost nothing to the electrochemical performance of Ni foam@MCS4 and carbon cloth@MCS4. In Figure S13(d), the charge transfer resistance of Ni foam and carbon cloth are very low, indicating fairly good electro-conductivity. As is discussed in the main manuscript, MCS4 sample (Ni foam@MCS4) has a very low equivalent series resistance of ~0.4 ohms while the equivalent series resistance of carbon cloth@MCS4 is about 7 ohms which reveals that the conductivity of carbon cloth@MCS4 is inferior to Ni foam@MCS4. This phenomenon could be attributed from loose connection between carbon cloth and MnCoS4 powders because of lacking high pressure operation during electrode preparation process. In a brief summary, the influence of Ni foam and carbon cloth to electrochemical performance of as-prepared electrode materials could be neglected and Ni foam is more suitable than carbon cloth to be used as current collector in this work.