

## Supporting Information

### **Hierarchical Integrated 3D Hollow MnS@MoS<sub>2</sub> Microcube via Template-Controlled Synthesis for Asymmetric Supercapacitors**

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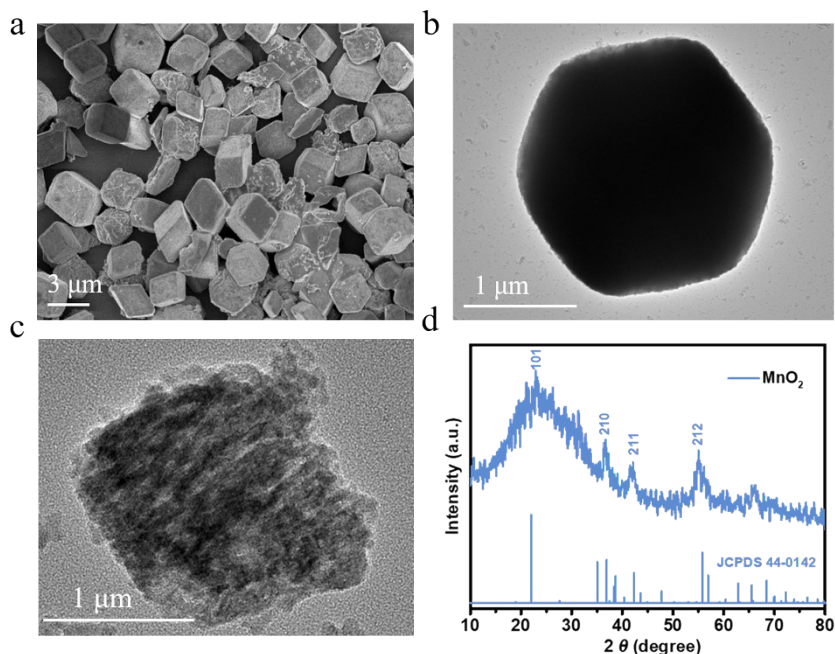


Figure S1. (a, b) SEM and TEM images of MnCO<sub>3</sub>. (c, d) TEM image and XRD pattern of hollow MnO<sub>2</sub>.

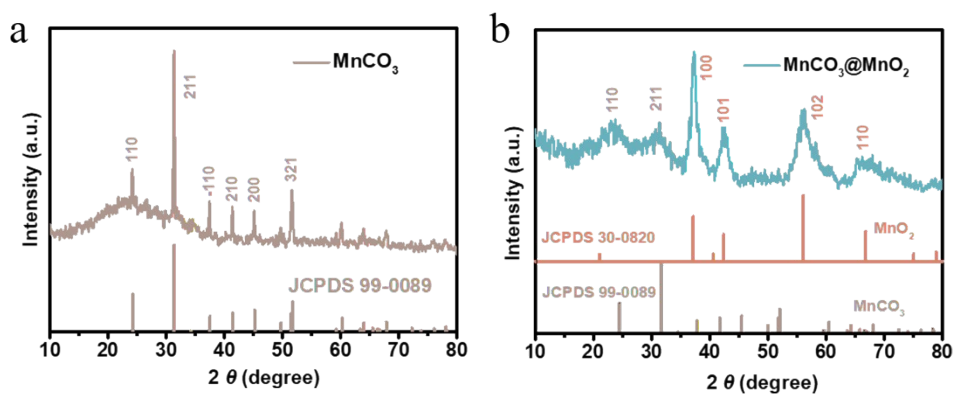


Figure S2. The XRD pattern of (a) MnCO<sub>3</sub> and (b) MnCO<sub>3</sub>@MnO<sub>2</sub>.

The peak at 23° and 33° in the XRD patterns (Figure S2b) are matched to the (110) and (211) crystal planes of MnCO<sub>3</sub> (JCPDS 99-0089), which confirmed the existence of intermediates. This XRD pattern (Figure S2) indicated the MnCO<sub>3</sub> transformed into the MnCO<sub>3</sub>@MnO<sub>2</sub> after the calcination and the as-synthesized composite was the basis of H<sup>+</sup> etching into hollow-cube through.

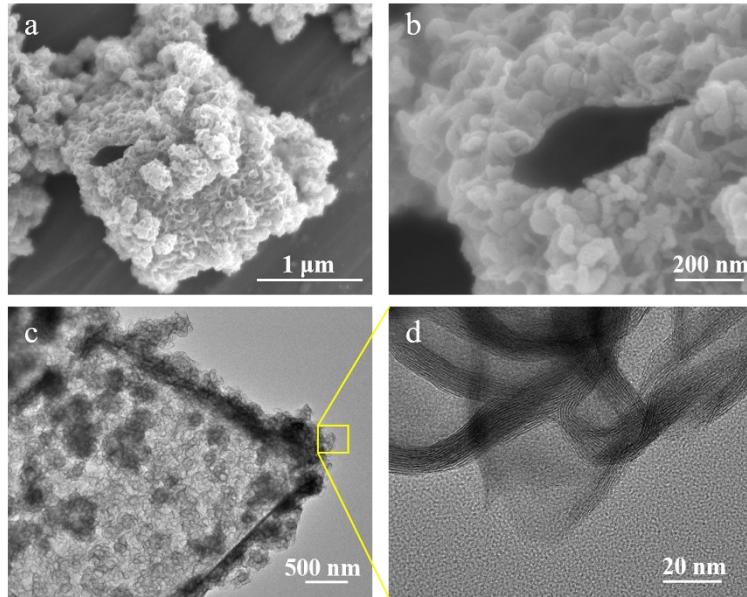


Figure S3. The SEM and TEM images of MnS@MoS<sub>2</sub>.

As shown in Figure 1d and Figure S3c, the TEM images of MnS@MoS<sub>2</sub> proved the existence of hollow structure. The hollow MnS microcube was wrapped by the MoS<sub>2</sub> nanosheets. Besides, the structural features of MoS<sub>2</sub> nanosheets can be seen from the enlarged TEM image of edges (Figure S3d).

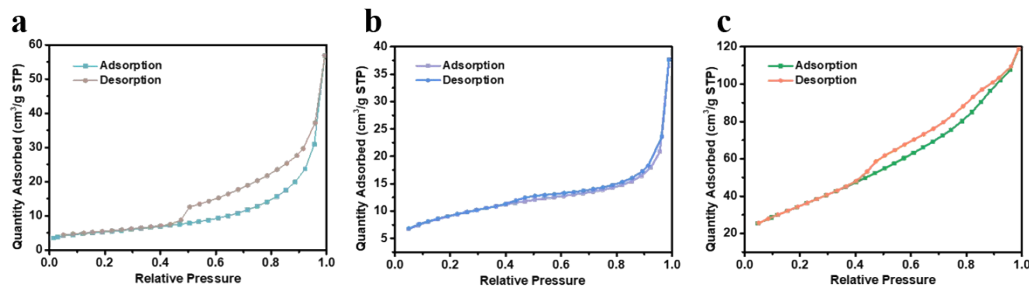


Figure S4. The N<sub>2</sub> ab-/desorption isotherm curves of (a) MnCO<sub>3</sub>, (b) MnO<sub>2</sub> and (c) MnS@MoS<sub>2</sub> composite.

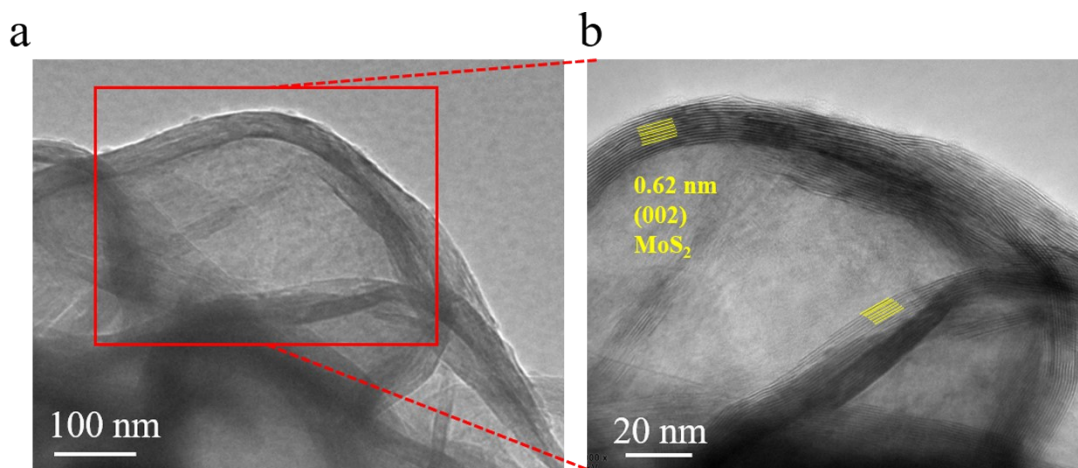


Figure S5. (a) TEM images of edge MoS<sub>2</sub>. (b) Enlarged TEM image of a.

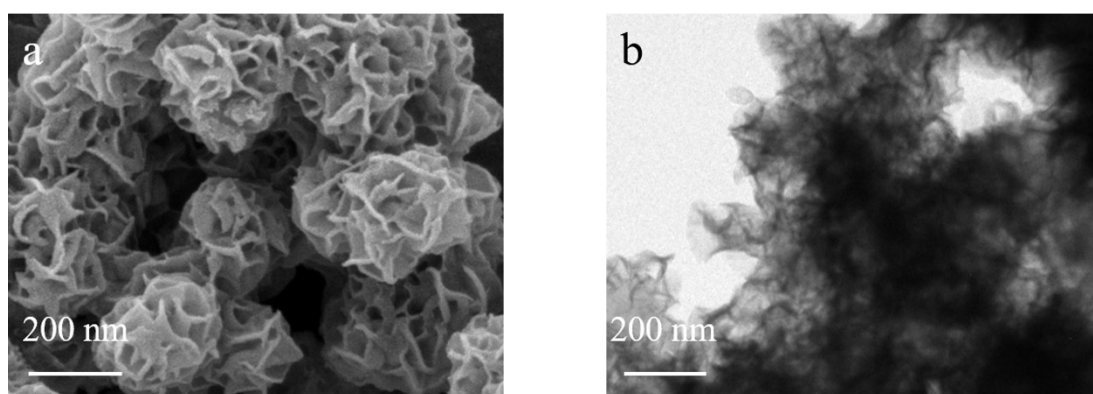


Figure S6. (a, b) SEM and TEM images of pure MoS<sub>2</sub>.

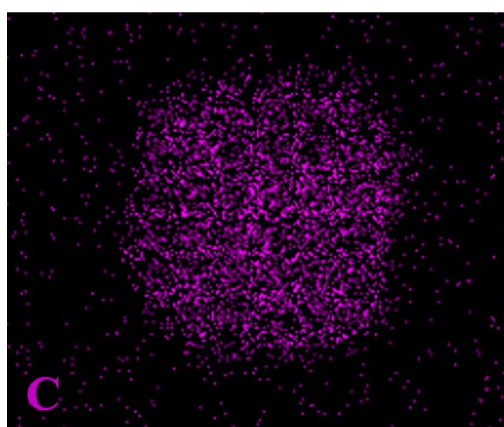


Figure S7. EDS mapping of C elements.

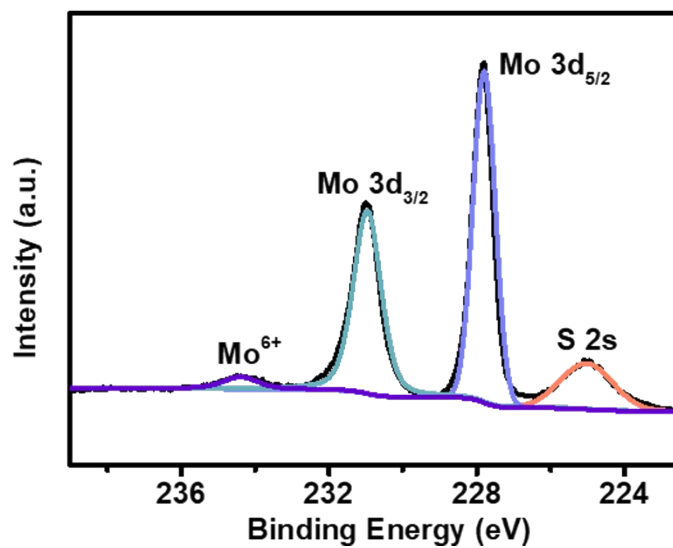


Figure S8. The high-resolution Mo XPS spectra of MoS<sub>2</sub>.

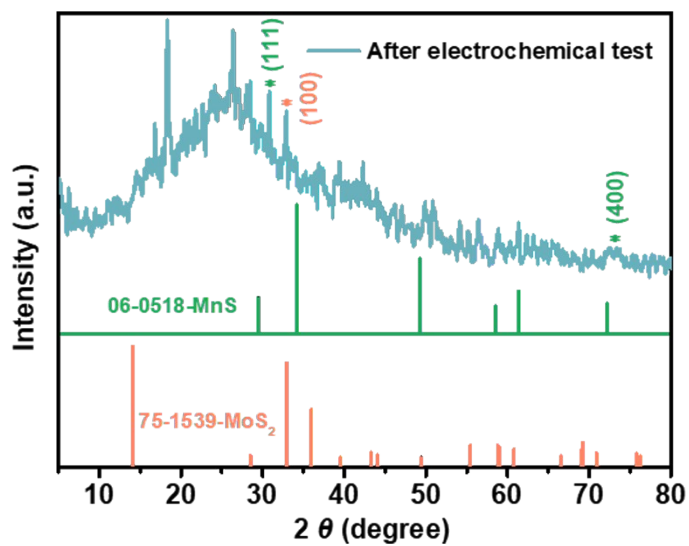


Figure S9. The XRD pattern of MnS@MoS<sub>2</sub> after electrochemical test.

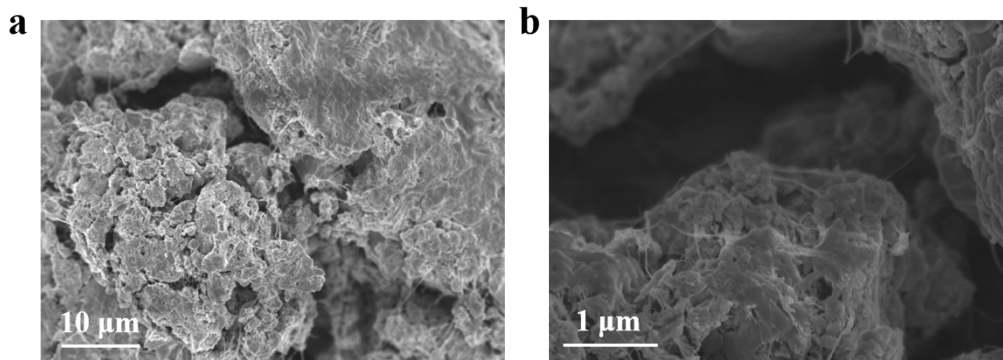


Figure S10. The SEM images of MnS@MoS<sub>2</sub> after electrochemical test.

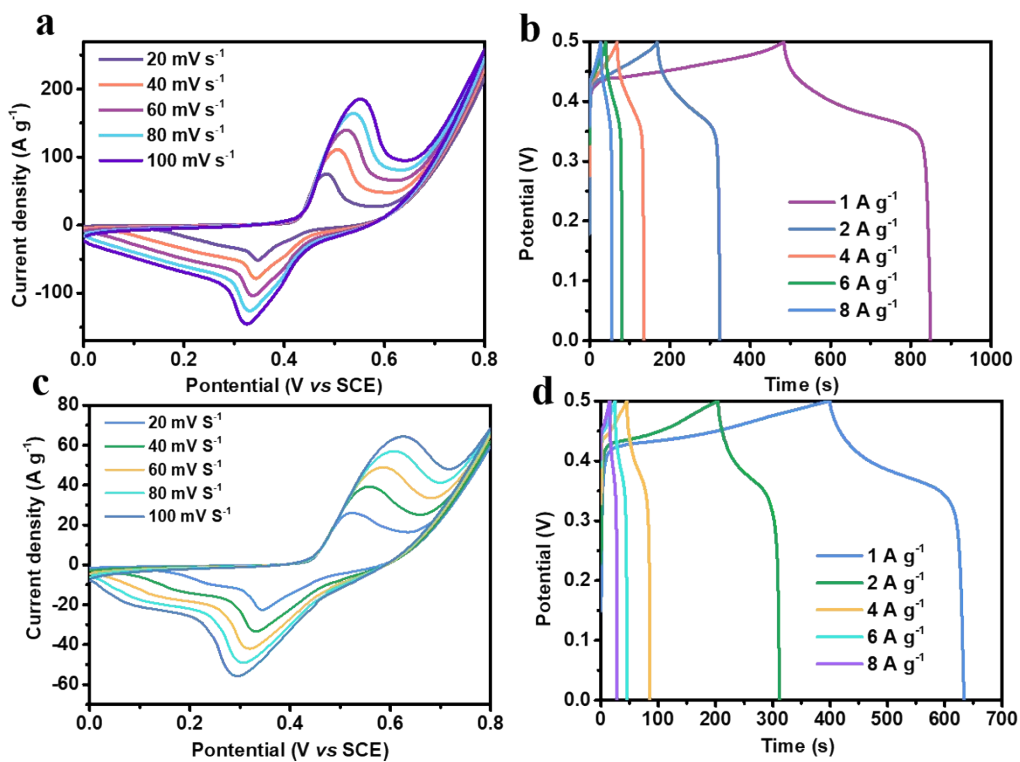


Figure S11. (a) CV curves of MoS<sub>2</sub> at various scan rates. (b) GCD curves of MoS<sub>2</sub> at various current densities. (c) CV curves of MnS at various scan rates. (d) GCD curves of MoS<sub>2</sub> at various current densities.

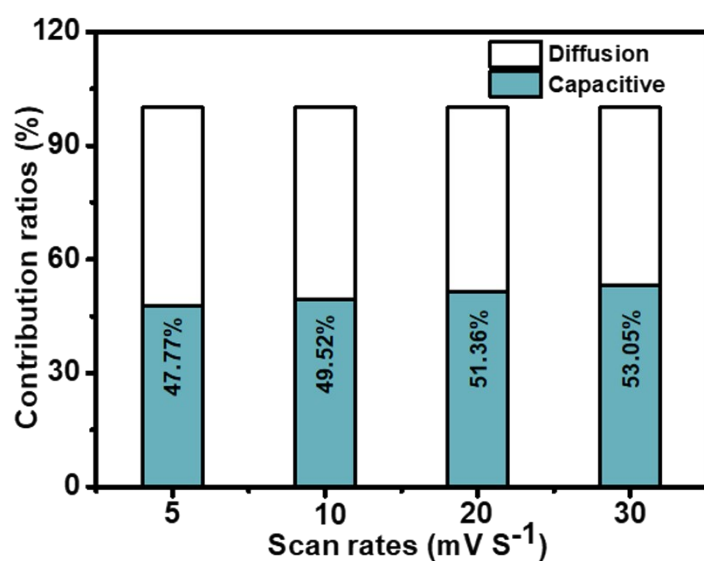


Figure S12. The capacitive contributions of MnS@MoS<sub>2</sub> at 5-30 mV s<sup>-1</sup>.

The contribution of different charge storage mechanism can be further characterized by the data at the scan rates of 5-30 mV s<sup>-1</sup>. The contribution of surface capacitive process increased with the scan rates increasing.

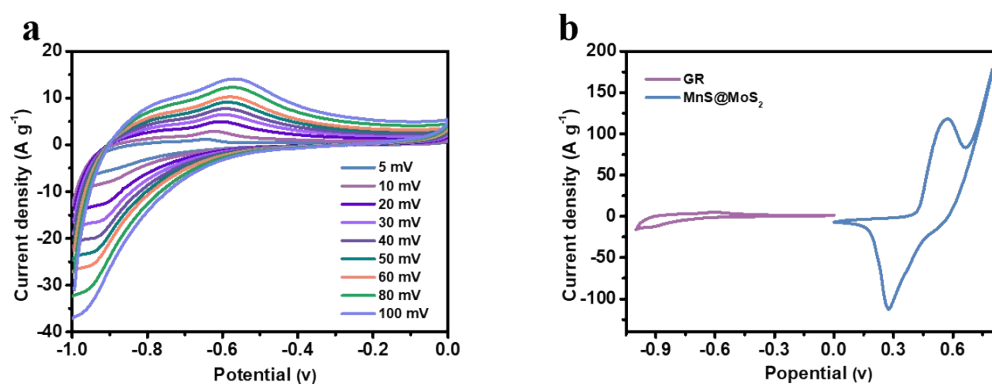


Figure S13. (a) CV curves of the GR at 5–100 mV s<sup>-1</sup>. (b) CV curves collected for GR and MnS@MoS<sub>2</sub> electrodes at a scan rate of 50 mV s<sup>-1</sup>.

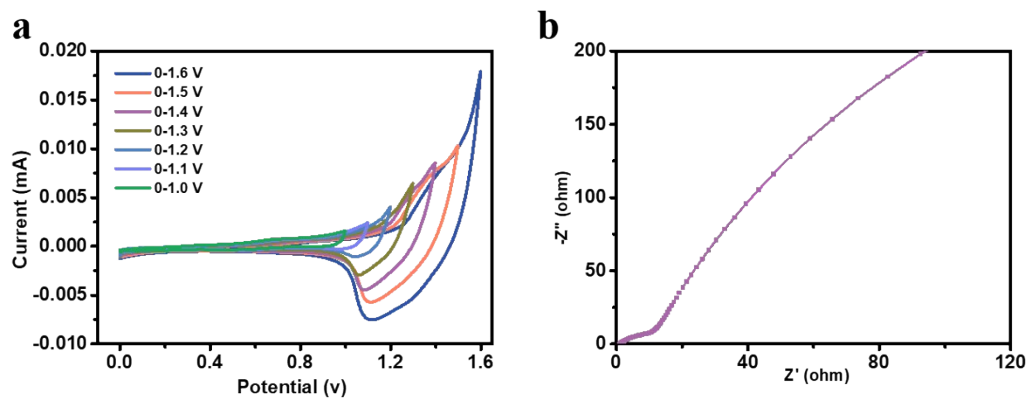


Figure S14. (a) CV curves of the MnS@MoS<sub>2</sub>//GR at various potential windows of 20 mV s<sup>-1</sup>. (b) EIS of Mn@MoS<sub>2</sub>//GR device.

Table S1 Comparison with previous reported asymmetric supercapacitors device

Electrode material	Energy density	Power density
Ni-Co-S@MnS	20.5	2000
MnO <sub>2</sub> @ppy	23	1800
MnS microfiber	42.7	1200
MnS nanocrystal	24.9	5946
This work	65/29	1600/6400