

## Supporting Information

### **Heteroarchitecturing a Novel Three-Dimensional Hierarchical MoO<sub>2</sub>/MoS<sub>2</sub>/Carbon Electrode Material for High-Energy and Long-Life Lithium Storage**

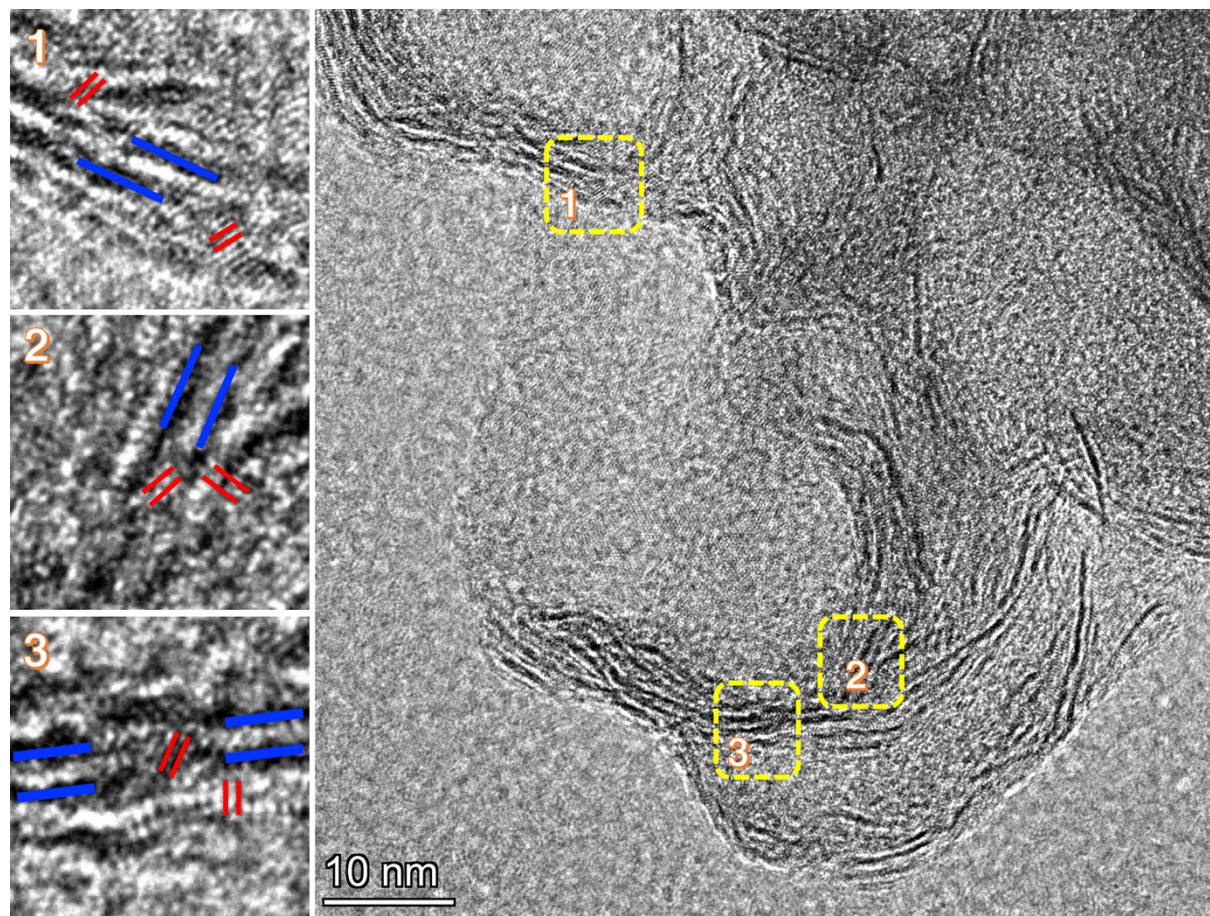
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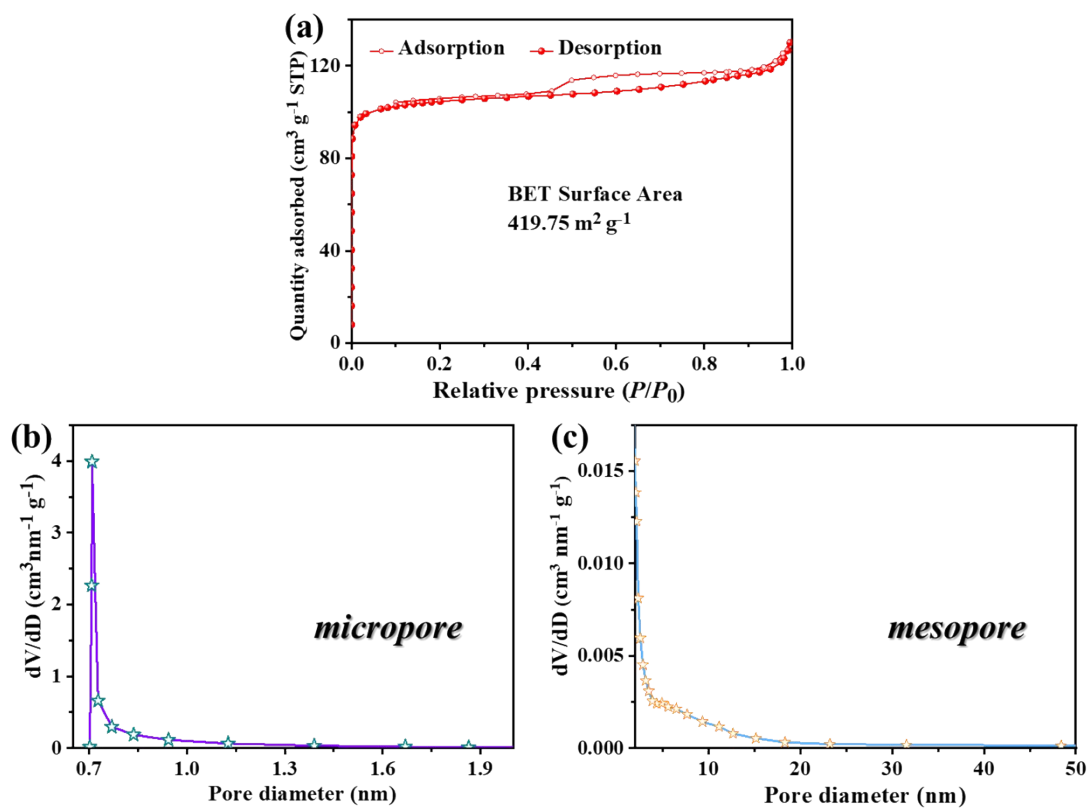
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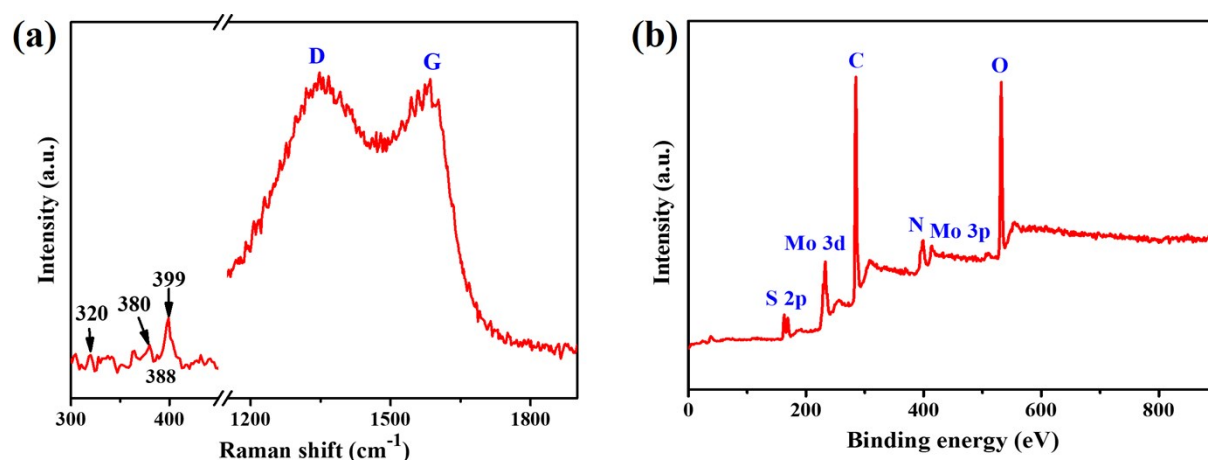
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**Figure S1** HRTEM image of MoO<sub>2</sub>/MoS<sub>2</sub>/C with partial enlarged view of the lattice fringes, showing the coexistence of two crystalline phases. The red lines denote the lattice fringes of the MoO<sub>2</sub> phase, while the blue ones indicate the lattice fringes of the MoS<sub>2</sub> phase.

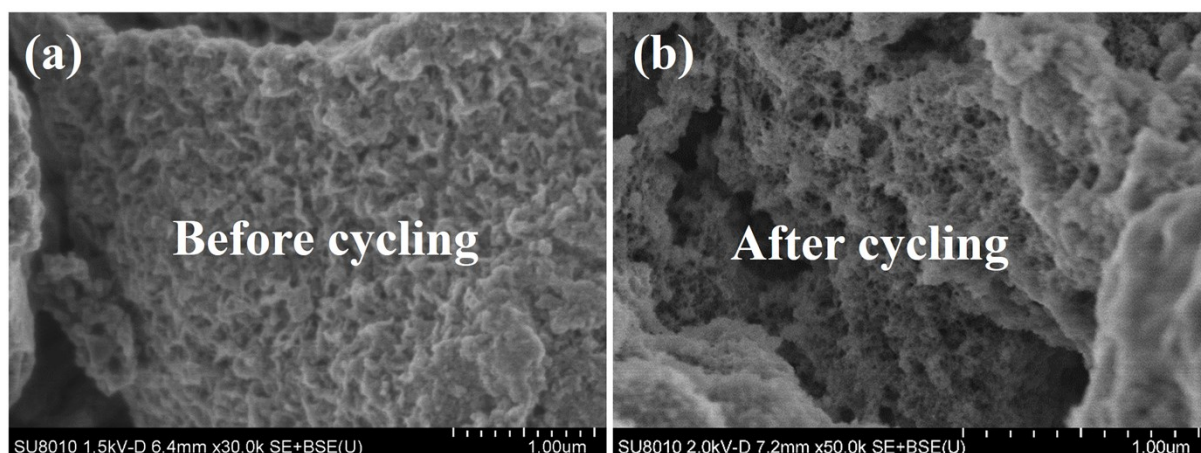


**Figure S2** (a)  $\text{N}_2$  adsorption–desorption isotherm, and (b, c) PSDs of  $\text{MoO}_2/\text{MoS}_2/\text{C}$  in the micropore and mesopore ranges, respectively.

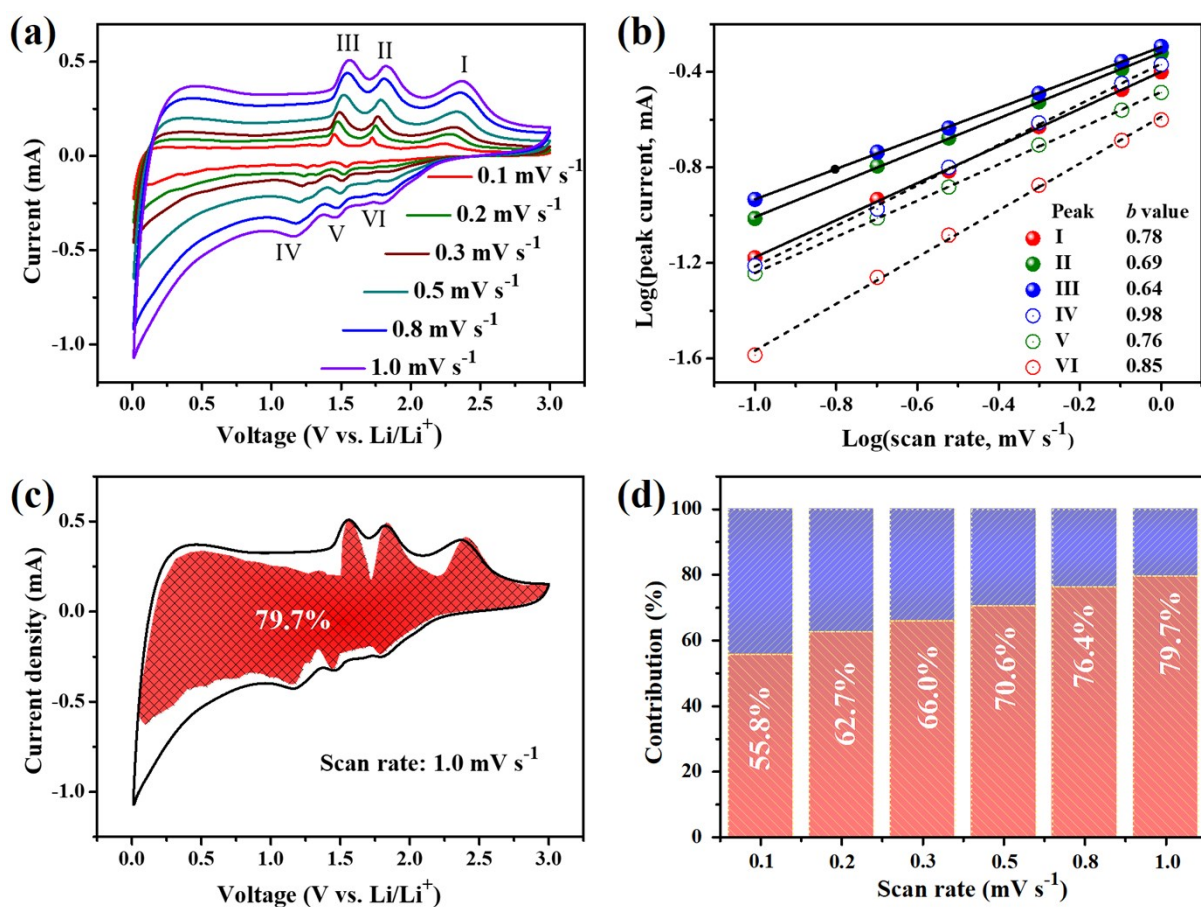


**Figure S3** (a) Raman spectrum and (b) XPS spectrum of  $\text{MoO}_2/\text{MoS}_2/\text{C}$ .

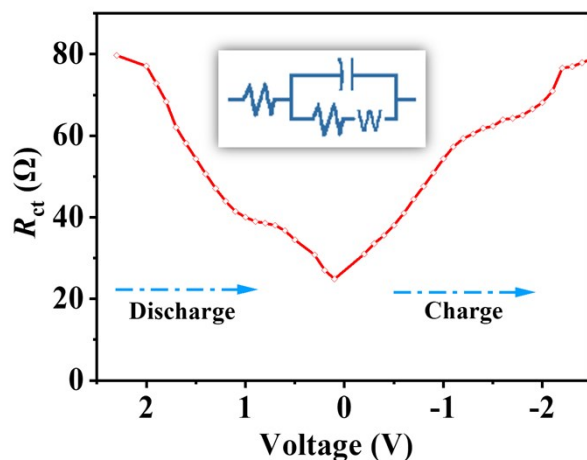




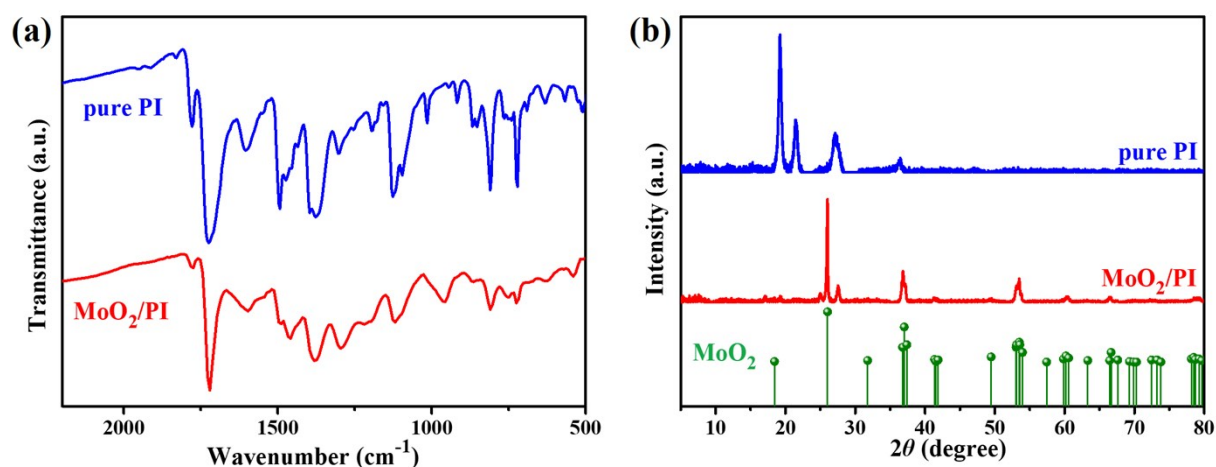
**Figure S4** SEM images of the MoO<sub>2</sub>/MoS<sub>2</sub>/C electrode before (a) and (b) after cycling at 5 A g<sup>-1</sup> for over 2000 cycles.



**Figure S5** (a) CV curves at different scan rate. (b) Log *i* vs. log *v* plots at different oxidation and reduction states. (c) Separation of the capacitive and diffusion currents at a scan rate of 1.0 mV s<sup>-1</sup>. (d) Normalized contribution ratio of capacitive and diffusion-controlled capacities at various scan rates.



**Figure S6** The fitting data of  $R_{ct}$  values at various charge/discharge voltages. The inset represents the equivalent circuit.



**Figure S7** (a) FTIR spectra and (b) powder XRD patterns of the pure PI and MoO<sub>2</sub>/PI precursors obtained after the hydrothermal polymerization.

**Note for Figure S7:** The appearance of the characteristic absorption peaks (the peaks of C=O at 1720 and 1778 cm<sup>-1</sup>, and the peak of imide C-N at 1380 cm<sup>-1</sup>)<sup>1</sup> in the FT-IR spectra shown in Figure S7a clearly demonstrates the successful synthesis of PI in both cases. The XRD patterns in Figure S7b clarify the existence of both PI and MoO<sub>2</sub> crystal phases, indicating the successful synthesis of MoO<sub>2</sub>/PI composite through the hydrothermal polymerization in the presence of PMo<sub>12</sub>.<sup>2</sup>

**Table S1** The elemental compositions of N-S-C obtained by XPS and CHNS analyses.

Sample	XPS analysis (atom%)				CHNS analysis (atom%)			
	C	O	N	S	C	H	N	S
N-S-C	88.35	4.91	5.25	1.48	73.97	2.27	5.91	4.36

## References

1. X. Liu, S. Qiu, P. Mei, Q. Zhang and Y. Yang, *J. Mater. Sci.*, 2020, **56**, 3900-3910.
2. J. Xie, K. Zhu, J. Min, L. Yang, J. Luo, J. Liu, M. Lei, R. Zhang, L. Ren and Z. Wang, *Ionics*, 2019, **25**, 1487-1494.