

## Supporting information

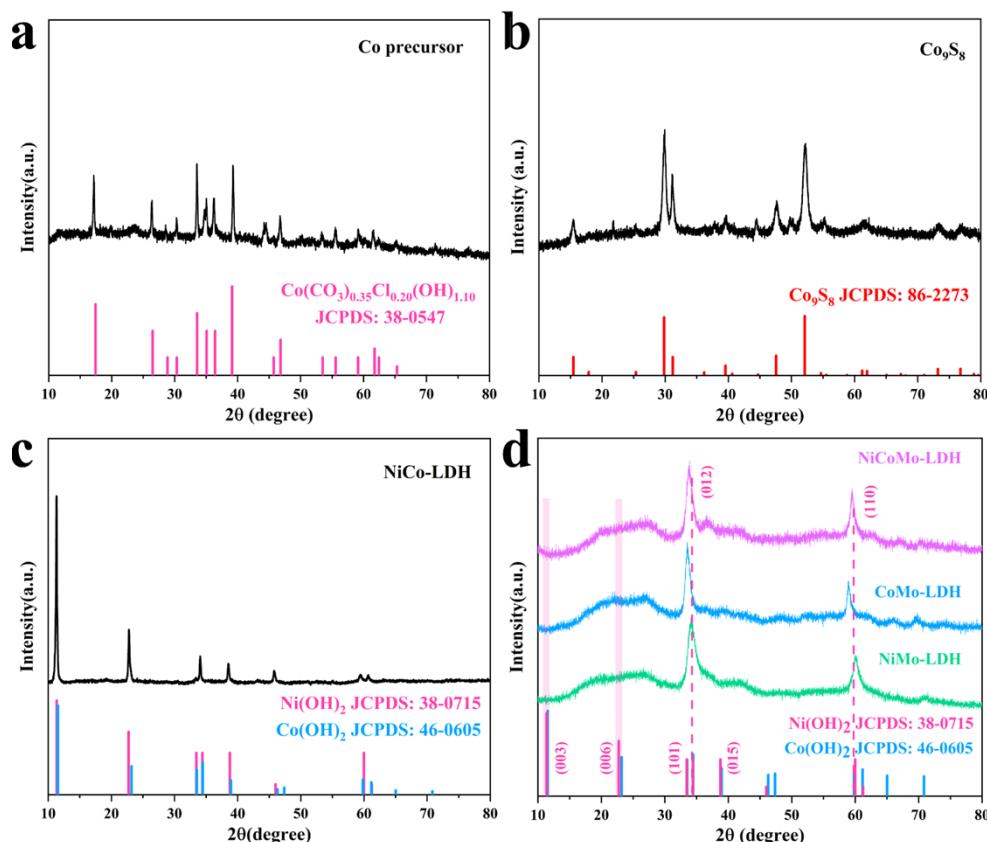
### Design and fabrication of $\text{MoO}_4^{2-}$ -intercalated LDHs nanosheets coated on $\text{Co}_9\text{S}_8$ nanotubes with enhanced cycle stability for high performance supercapacitors

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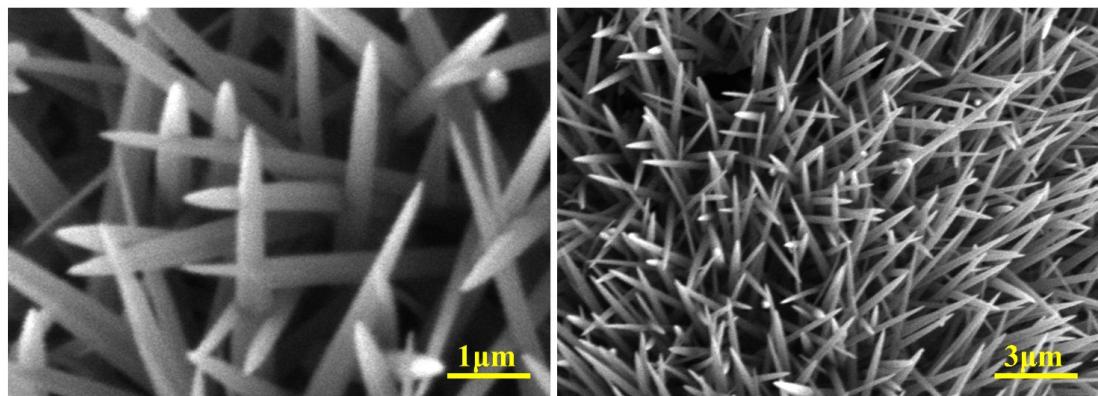
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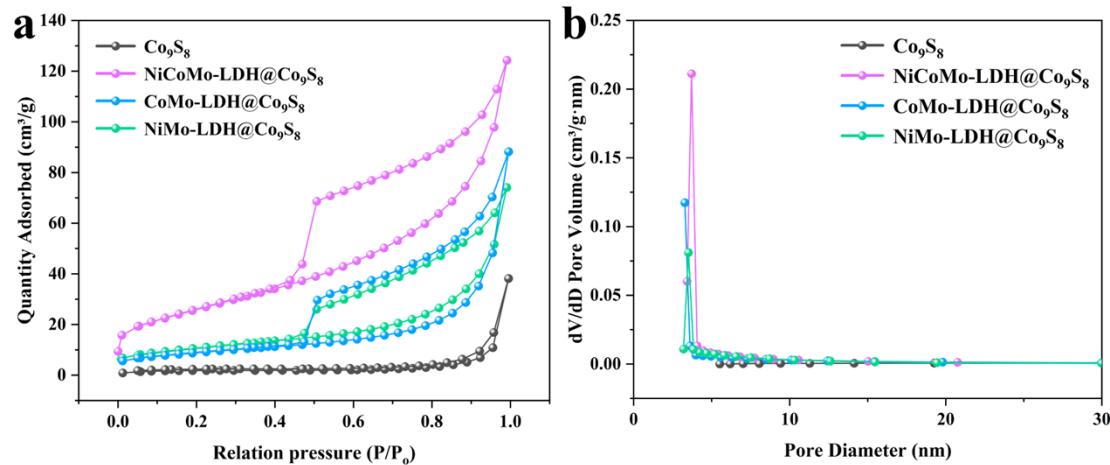
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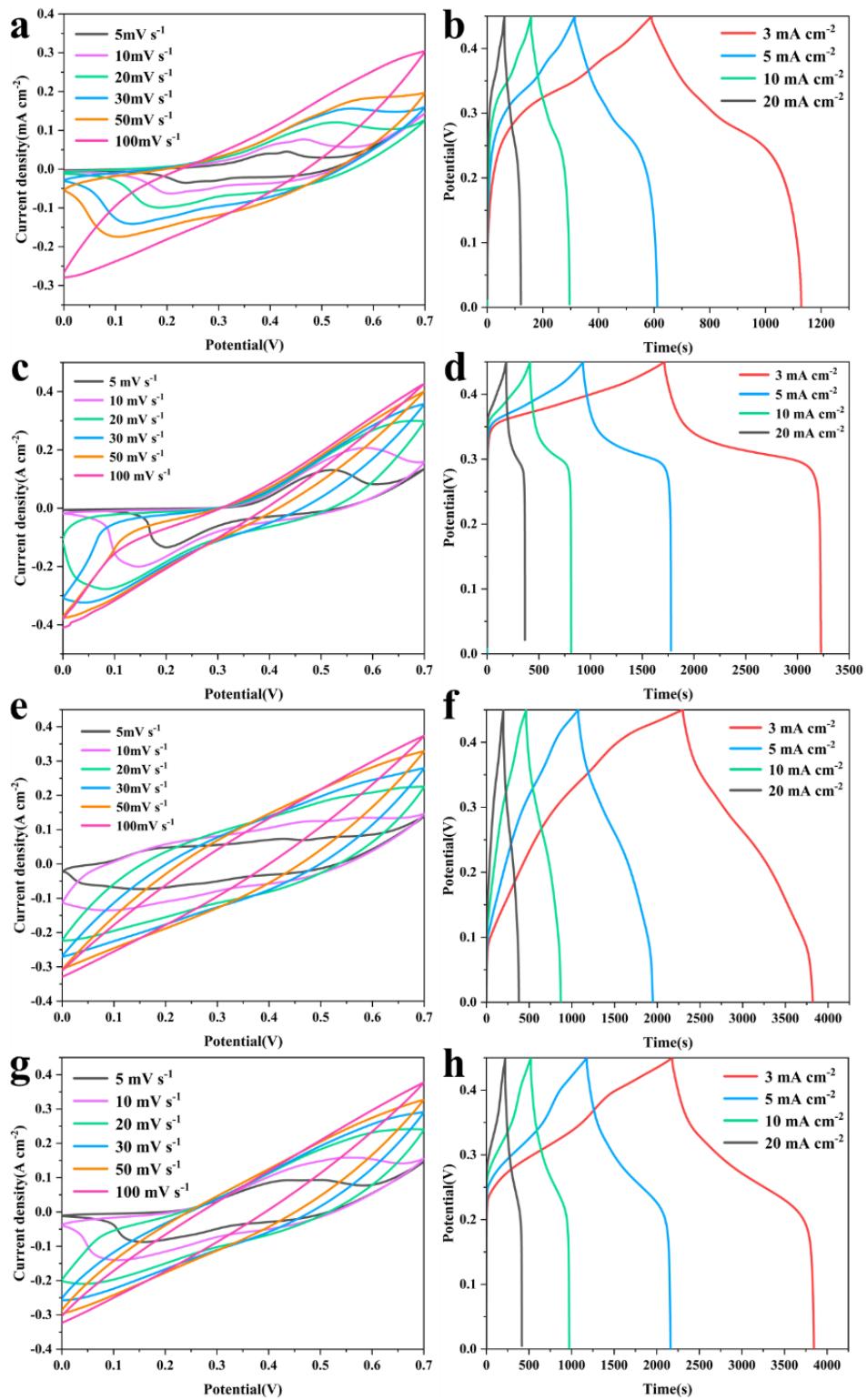
**Fig. S1** XRD images of Co precursor,  $\text{Co}_9\text{S}_8$ , NiCo-LDH and  $\text{MoO}_4^{2-}$ -intercalated LDHs.



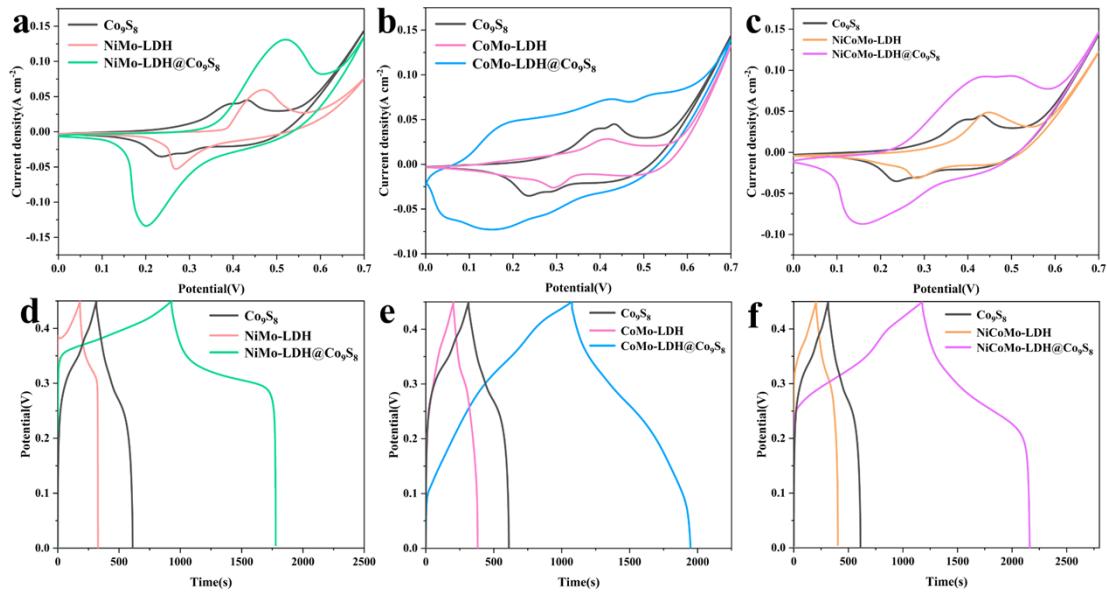
**Fig. S2** SEM images of the Co precursor.



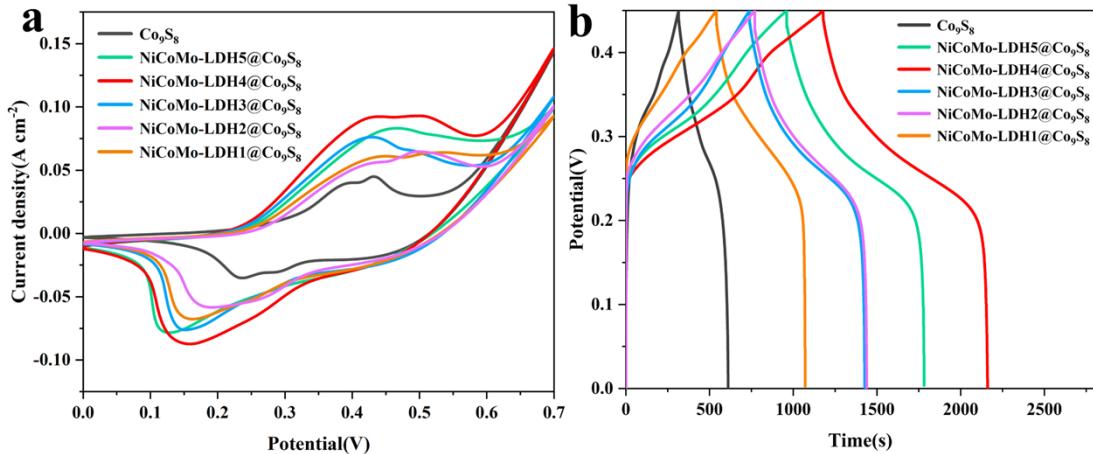
**Fig. S3** (a) Nitrogen adsorption-desorption isotherms of  $\text{Co}_9\text{S}_8$  and  $\text{MoO}_4^{2-}$ -intercalated LDHs@ $\text{Co}_9\text{S}_8$ ; (b) Pore diameter distribution of  $\text{Co}_9\text{S}_8$  and  $\text{MoO}_4^{2-}$ -intercalated LDHs@ $\text{Co}_9\text{S}_8$ .



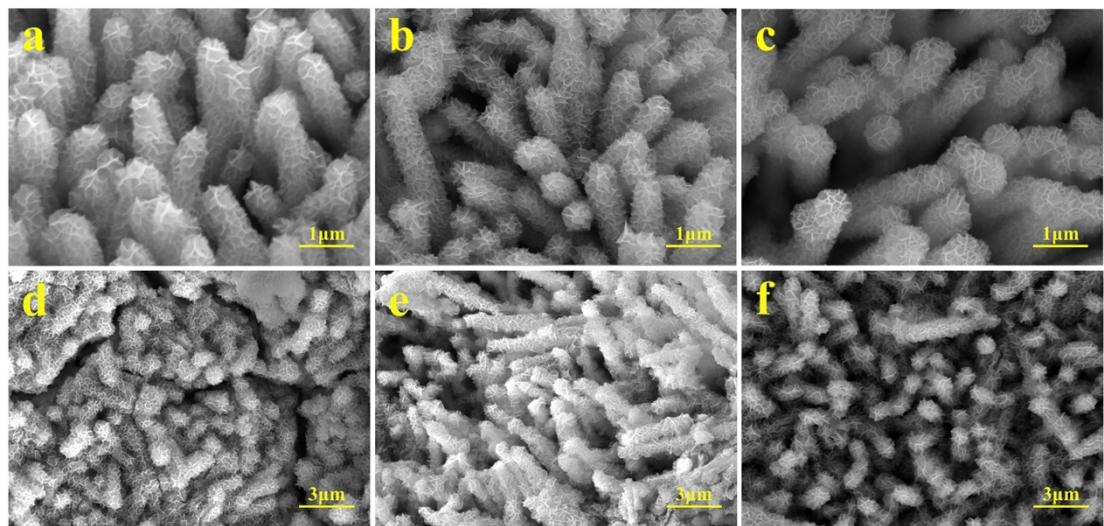
**Fig. S4** (a) CV and (b) GCD curves of  $\text{Co}_9\text{S}_8$ , (c) CV and (d) GCD curves of  $\text{NiMo-LDH@Co}_9\text{S}_8$ , (e) CV and (f) GCD curves of  $\text{CoMo-LDH@Co}_9\text{S}_8$ , (g) CV and (h) GCD curves of  $\text{NiCoMo-LDH@Co}_9\text{S}_8$ .



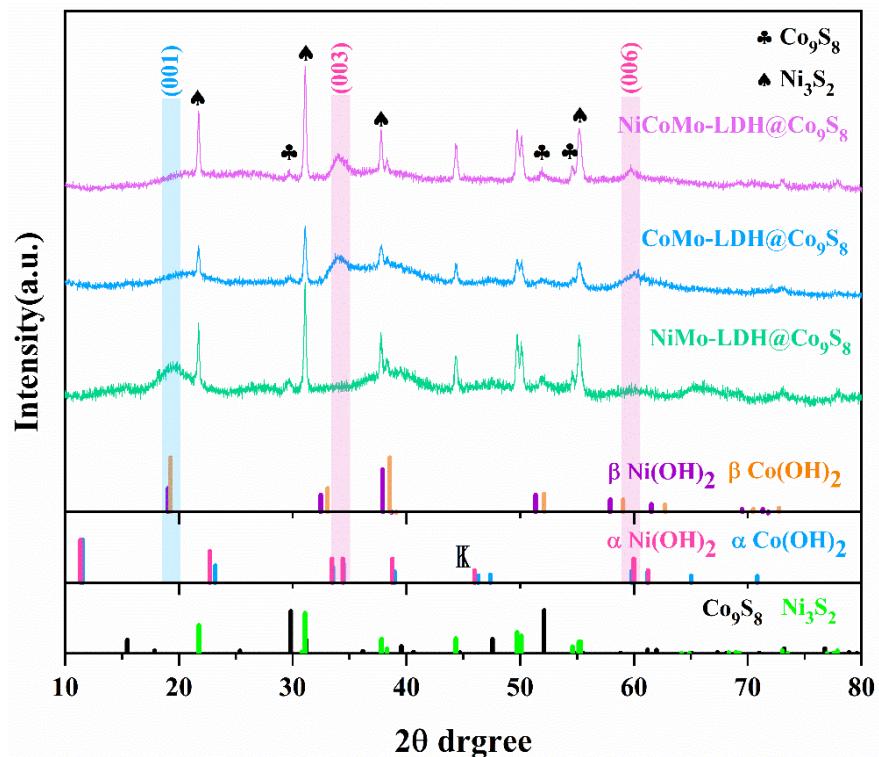
**Fig. S5** (a, b, c) CV curves comparing of  $\text{Co}_9\text{S}_8$ , LDH and LDH@ $\text{Co}_9\text{S}_8$  at  $5 \text{ mV s}^{-1}$ ; (b, d, f) GCD curves comparing of  $\text{Co}_9\text{S}_8$ , LDH and LDH@ $\text{Co}_9\text{S}_8$  at  $5 \text{ mA cm}^{-2}$ .



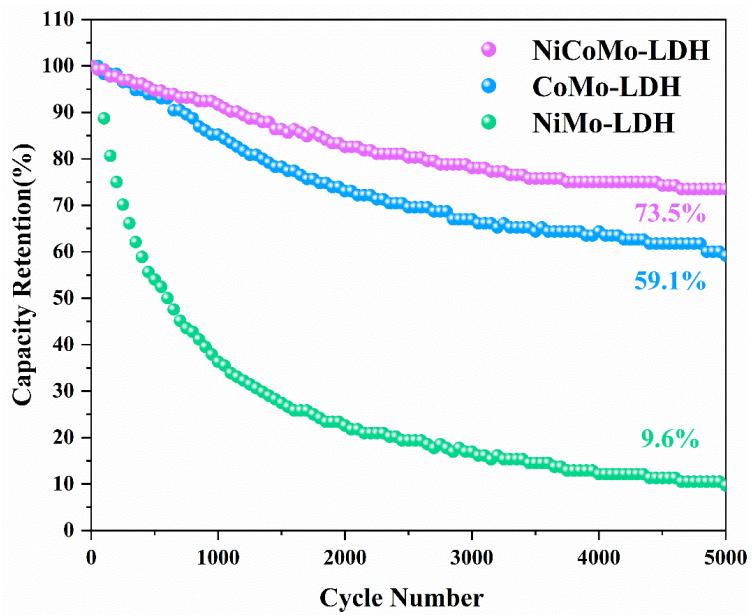
**Fig. S6** (a) CV curves of NiCoMo-LDH@ $\text{Co}_9\text{S}_8$  with different contents of  $\text{MoO}_4^{2-}$  at  $5 \text{ mV s}^{-1}$ ; (b) GCD curves of NiCoMo-LDH@ $\text{Co}_9\text{S}_8$  with different contents of  $\text{MoO}_4^{2-}$  at  $5 \text{ mA cm}^{-2}$ .



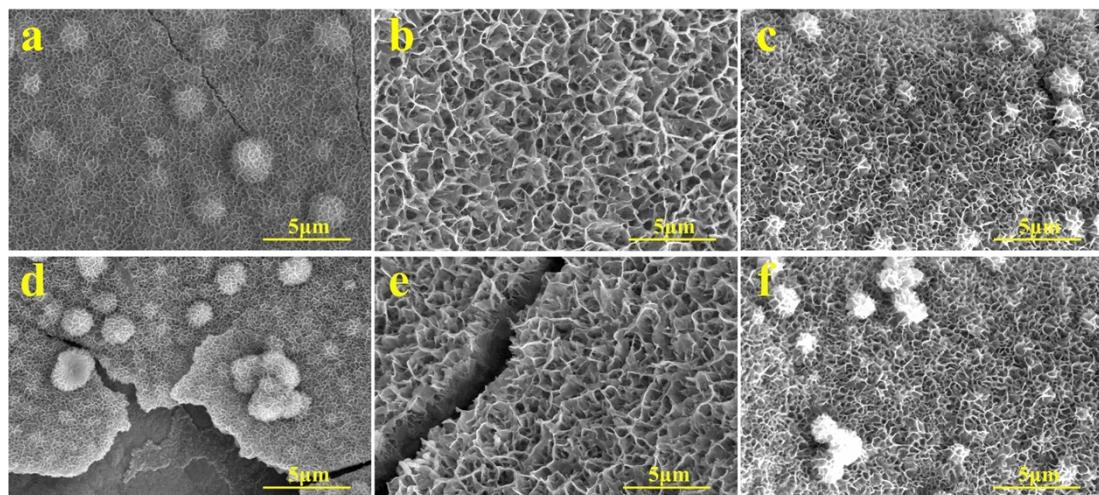
**Fig. S7** SEM images of (a) NiMo-LDH@Co<sub>9</sub>S<sub>8</sub>, (b) CoMo-LDH@Co<sub>9</sub>S<sub>8</sub> and (c) NiCoMo-LDH@Co<sub>9</sub>S<sub>8</sub> before 5000 cycles; SEM images of (d) NiMo-LDH@Co<sub>9</sub>S<sub>8</sub>, (e) CoMo-LDH@Co<sub>9</sub>S<sub>8</sub> and (f) NiCoMo-LDH@Co<sub>9</sub>S<sub>8</sub> after 5000 cycles, respectively.



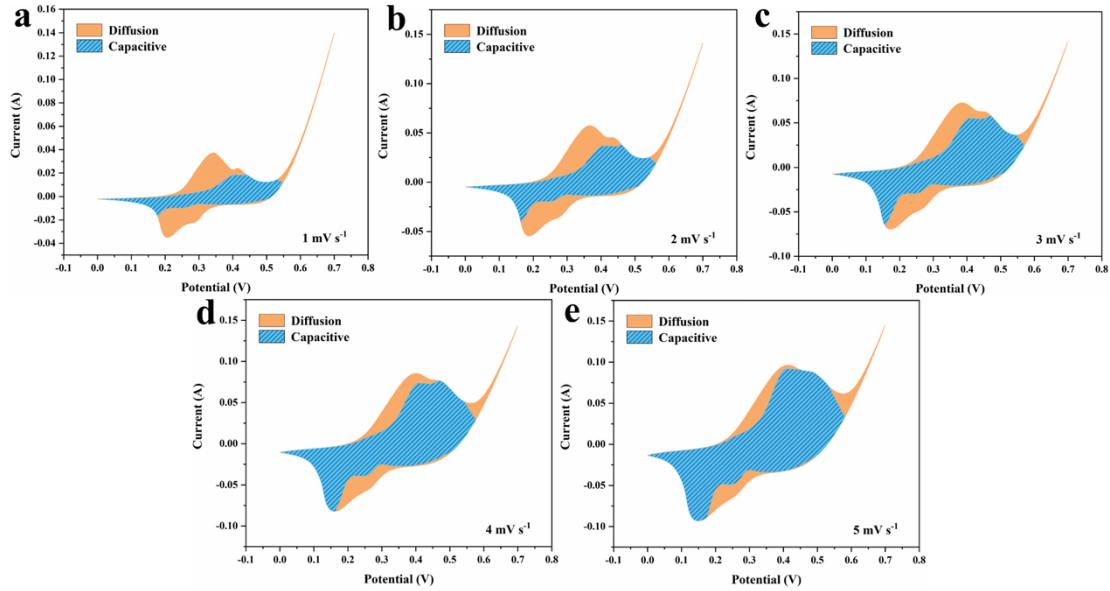
**Fig. S8** XRD of MoO<sub>4</sub><sup>2-</sup>-intercalated LDHs@Co<sub>9</sub>S<sub>8</sub> after 5000 cycles.



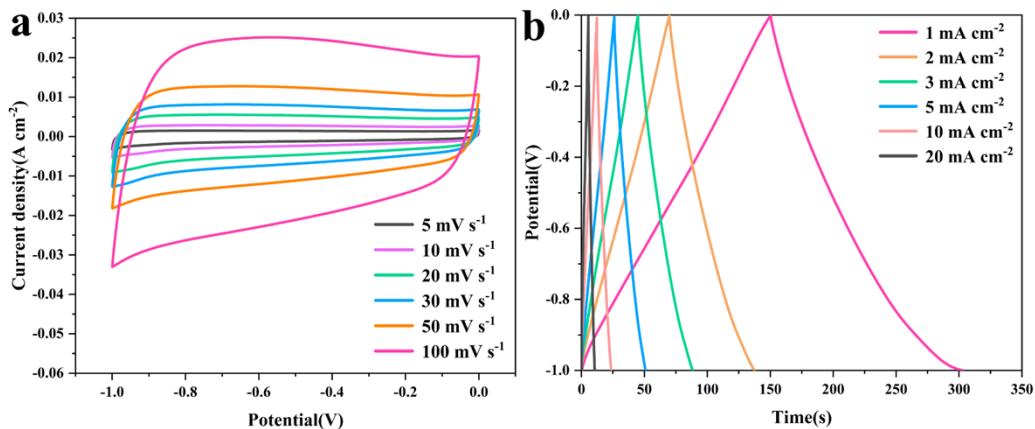
**Fig. S9** The cycle stability of NiMo-LDH, CoMo-LDH and NiCoMo-LDH as electrode materials.



**Fig. S10** SEM images of (a) NiMo-LDH, (b) CoMo-LDH and (c) NiCoMo-LDH before 5000 cycles; SEM images of (d) NiMo-LDH, (e) CoMo-LDH and (f) NiCoMo-LDH after 5000 cycles, respectively.



**Fig. S11** Capacitive contribution (blue) during charge storage at (a)  $1 \text{ mV s}^{-1}$ , (b)  $2 \text{ mV s}^{-1}$ , (c)  $3 \text{ mV s}^{-1}$ , (d)  $4 \text{ mV s}^{-1}$ , (e)  $5 \text{ mV s}^{-1}$ .



**Fig. S12** Electrochemical performance of AC: (a) CV curves; (b) GCD curves.

**Table S1** The atomic ratio of Co, Ni, Mo, O and S in NiCoMo-LDH@Co<sub>9</sub>S<sub>8</sub>, CoMo-LDH@Co<sub>9</sub>S<sub>8</sub>, NiMo-LDH@Co<sub>9</sub>S<sub>8</sub> obtained by XPS test.

Element	NiCoMo-LDH@Co <sub>9</sub> S <sub>8</sub>	CoMo-LDH@Co <sub>9</sub> S <sub>8</sub>	NiMo-LDH@Co <sub>9</sub> S <sub>8</sub>
Co	10.42%	24.99%	4.32%
Ni	13.89%	5.47%	19.34%
Mo	5.44%	6.21%	3.84%
O	64.95%	66.94%	68.91%
S	5.28%	4.92%	3.56%

**Table S2** Comparison of different electrode materials related to NiCoMo-LDH@Co<sub>9</sub>S<sub>8</sub>.

Electrode materials	Capacitance (F cm <sup>-2</sup> )	Current density (mA cm <sup>-2</sup> )	Cyclic stability	Ref
CoO@NiCo LDH	9.42 F cm <sup>-2</sup>	3 mA cm <sup>-2</sup>	88.76% (5000 cycles)	[1]
NC LDH NSs@Ag@CC	10.95 F cm <sup>-2</sup>	3 mA cm <sup>-2</sup>	80.47% (2000 cycles)	[2]
Co <sub>9</sub> S <sub>8</sub> @MnO <sub>2</sub>	0.80 F cm <sup>-2</sup>	2 mA cm <sup>-2</sup>	91.4% (5000 cycles)	[3]
NiMn-LDH@CuO/CF	6.07 F cm <sup>-2</sup>	2 mA cm <sup>-2</sup>	89.22% (8000 cycles)	[4]
Ni <sub>3</sub> S <sub>2</sub> /CoFe LDH/NF	5.08 F cm <sup>-2</sup>	2 mA cm <sup>-2</sup>	89.8% (8000 cycles)	[5]
Ag@NCM-LDH MFs@NSs/NF	6.40 F cm <sup>-2</sup>	5 mA cm <sup>-2</sup>	88.8% (15000 cycles)	[6]
NiV LDHs@P-NF	2.85 F cm <sup>-2</sup>	20 mA cm <sup>-2</sup>	Rarely decrease (5000 cycles)	[7]
Co <sub>9</sub> S <sub>8</sub> /NF	7.36 F cm <sup>-2</sup>	2 mA cm <sup>-2</sup>	/	[8]
CC/NiCoP@NiCo-LDH	4.68 F cm <sup>-2</sup>	1 mA cm <sup>-2</sup>	81.1% (5000 cycles)	[9]
NiCoMo-LDH@Co <sub>9</sub> S <sub>8</sub> //AC	11.0 F cm <sup>-2</sup>	3 mA cm <sup>-2</sup>	94.4% (5000 cycles)	This work

## References

- [S1] Jiao, Z.; Chen, Y.; Du, M.; Demir, M.; Yan, F.; Xia, W.; Zhang, Y.; Wang, C.; Gu, M.; Zhang, X.; Zou, J. 3D hollow NiCo LDH nanocages anchored on 3D CoO sea urchin-like microspheres: A novel 3D/3D structure for hybrid supercapacitor electrodes. *J. Colloid Interface Sci.* **2023**, *633*, 723-736.
- [S2] Sekhar, S. C.; Nagaraju, G.; Yu, J. S. Conductive silver nanowires-fenced carbon cloth fibers-supported layered double hydroxide nanosheets as a flexible and binder-free electrode for high-performance asymmetric supercapacitors. *Nano Energy* **2017**, *36*, 58-67.
- [S3] Li, Q.; Liu, M.; Huang, F.; Zuo, X.; Wei, X.; Li, S.; Zhang, H. Co<sub>9</sub>S<sub>8</sub>@MnO<sub>2</sub> core-shell defective heterostructure for High-Voltage flexible supercapacitor and Zn-ion hybrid supercapacitor. *Chem. Eng. J.* **2022**, *437*, 135494.
- [S4] Zhang, A.; Zheng, W.; Yuan, Z.; Tian, J.; Yue, L.; Zheng, R.; Wei, D.; Liu, J. Hierarchical NiMn-layered double hydroxides@CuO core-shell heterostructure in-situ generated on Cu(OH)<sub>2</sub> nanorod arrays for high performance supercapacitors. *Chem. Eng. J.* **2020**, *380*, 122486.
- [S5] Wang, Y.; Zhang, W.; Guo, X.; Liu, Y.; Zheng, Y.; Zhang, M.; Li, R.; Peng, Z.; Zhao, Y. Construction of high-performance asymmetric supercapacitor based on the hierarchical Ni<sub>3</sub>S<sub>2</sub>/CoFe LDH/Ni foam hybrid. *Appl. Surf. Sci.* **2021**, *561*, 150049.
- [S6] Ramulu, B.; Sekhar, S. C.; Arbaz, S. J.; Yu, J. S. Nano-Ag laminated ternary layered double hydroxides for hybrid supercapacitors. *Chem. Eng. J.* **2021**, *420*, 130376.
- [S7] Wang, G.; Jin, Z.; Guo, Q. Ordered Self-supporting NiV LDHs@P-Nickel foam

Nano-array as High-Performance supercapacitor electrode. *J. Colloid Interface Sci.* **2021**, *583*, 1-12.

- [S8] Aloqayli, S.; Ranaweera, C. K.; Wang, Z.; Siam, K.; Kahol, P. K.; Tripathi, P.; Srivastava, O. N.; Gupta, B. K.; Mishra, S. R.; Perez, F.; Shen, X.; Gupta, R. K. Nanostructured cobalt oxide and cobalt sulfide for flexible, high performance and durable supercapacitors. *Energy Storage Mater.* **2017**, *8*, 68-76.
- [S9] Gao, X.; Zhao, Y.; Dai, K.; Wang, J.; Zhang, B.; Shen, X. NiCoP nanowire@NiCo-layered double hydroxides nanosheet heterostructure for flexible asymmetric supercapacitors. *Chem. Eng. J.* **2020**, *384*, 123373.