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## Hierarchically Structured Layered-Double- Hydroxide@Zeolitic-

## **Imidazolate-Framework Derivatives for High-Performance**

## **Electrochemical Energy Storage**

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Figure S1. EDX spectrum of CoAl-LDH film.



Figure S2. SEM images of the CoAl-LDH@ZIF-67 film as a function of reaction time (A) 10 h, (B) 18 h, and (C) 24 h.



Figure S3. SEM image of ZIF-67.



Figure S4. EDX spectrum of CoAl-LDH@ZIF-67 film.



Figure S5. Digital photographs of (A) CoAl-LDH and (B) CoAl-LDH@ZIF-67 films.



Figure S6. EDX spectrum of (A) MMO@Co<sub>3</sub>O<sub>4</sub>, (B) Spinelle@C, and (C) LDH@CoS, respectively.



**Figure S7.** SEM images of (A) MMO@Co<sub>3</sub>O<sub>4</sub>, (B) Spinelle@C, and (C) LDH@CoS obtained by oxidation at 300 °C under air for 5 h, carbonization at 800 °C under N<sub>2</sub> for 5 h, and sulfurization with thioacetamide for 3 h, respectively.



Figure S8. SEM image of Co<sub>3</sub>O<sub>4</sub> powder derived from ZIF-67 powder.



Figure S9. SEM image of nitrogen-doped carbon powder derived from ZIF-67 powder.



Figure S10. SEM image of CoS powder derived from ZIF-67 powder.



**Figure S11.** N<sub>2</sub> adsorption-desorption isotherms and pore size distributions (inset) of (A) MMO@Co<sub>3</sub>O<sub>4</sub>, (B) Spinelle@C, and (C) LDH@CoS, respectively.



Figure S12. CV curves of (A) LDH, (B) MMO@Co<sub>3</sub>O<sub>4</sub>, (C) Spinelle@C, and (D) LDH@CoS at varied scan rates from 5 to 60 mV/s.



**Figure S13.** Galvanostatic discharge curves of (A) LDH, (B) MMO@Co<sub>3</sub>O<sub>4</sub>, (C) Spinelle@C, and (D) LDH@CoS at varied current densities from 1 to 20 A/g.



**Figure S14.** Specific capacity of LDH and LDH@CoS with varied amounts of CoS that derived based on different sulfurization time of 20, 40, and 60 min, respectively.

MOFs	MOFs derivatives	Specific capacitance (F g <sup>-1</sup> )	Current density (A g <sup>-1</sup> )	Reference
MOF-5	NPC	258	0.25	1
Bi/MOF-5	HPCs	241	0.1	2
Al-PCP	Carbon	232.8	0.1	3
ZIF-11	Nitrogen-doped porous carbon	307	1	4
ZIF-8	PCPs	245	1	5
IRMOF-3	N doped Carbon	213	0.5	6
HKUST-1	Cu <sub>1.96</sub> S–C	200	0.5	7
ZIF-67	Co <sub>3</sub> O <sub>4</sub> //C	101	2	8
Co-MOF	Co <sub>3</sub> O <sub>4</sub>	150	1	9
LDH@ZIF-67	MMO@Co <sub>3</sub> O <sub>4</sub>	692	1	This work
LDH@ZIF-67	Spinelle@C	781	1	This
LDH@ZIF-67	LDH@CoS	1205	1	This work

 Table S1. Comparison study of LDH@ZIF-67 derivatives in this work and previously

 reported MOFs derivatives toward electrochemical energy storage.



Figure S15. SEM image of MMO@Co<sub>3</sub>O<sub>4</sub> after 2000 galvanostatic charge-discharge cycles.



Figure S16. SEM images of (A) Spinelle@C and (B) LDH@CoS after 2000 galvanostatic charge-discharge cycles.



Figure S17. Cycling stability of the LDH@CoS at consecutively varied current densities.

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