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COMMUNICATION

Supporting Information

A new promising Ni-MOF superstructure for high-performance supercapacitors

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Experimental Section

Materials

2,5-thiophenedicarboxylate acid, Nickel(II) acetylacetonate, polyvinyl alcohol, NaOH, KOH and EtOH were purchased from Shanghai Sinopharm Chemical Reagent Co, and activated carbon (AC) was purchased from Nanjing Xfnano Company. All the aqueous solutions were freshly prepared with high purity water.

Synthesis of Ni-TDA

At first, 0.17 g of 2,5-thiophenedicarboxylate acid and 0.257 g of Nickel(II) acetylacetonate were dissolved in 10 mL H_2O and 10 mL EtOH with stirring at room temperature. Then, 2 mL of NaOH aqueous solution (0.5 M) was slowly added to the above solution drop by drop. After that, the solution was transferred into a 50 mL round-bottomed flask and heated to 80 ℃ for 5 h. The resulting precipitate was washed with H_2O and EtOH for several times. Finially, the product was dried at 110 ℃ for 12 h under ambient conditions.

Electrochemical measurements

The electrochemical performances were evaluated by using a conventional three electrode test pool with a platinum foil electrode and a Hg–HgO electrode as the counter and reference electrodes, respectively. The working electrode was made by mixing the active material (70 wt%), acetylene Black (20 wt%) and polytetrafluorethylene binder (10 wt%). The mass loading of the electrode material was in the range of 4–5 mg. The electrolyte was 2 M KOH solution. Cyclic voltammetry (CV) measurements, and galvanostatic charge–discharge (GCD) measurements measurements of gear-like Ni-MOF electrodeswere conducted on a Land automatic batteries tester (Land, CT 2001A, Wuhan, China), respectively.

Characterizations

The crystal structure of the sample was determined using X-ray diffraction (XRD) patterns recorded on Rigaku Ultima IV with Cu radiation of 1.5418 Å. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were taken on a Hitachi S4800 instrument and a TECNAI G2 F20 instrument, respectively. N_2 adsorption/desorption analysis was performed on a Micromeritics ASAP 2020 instrument. Brunauer–Emmett–Teller (BET) and Barrett–Joyner–Halenda (BJH) analysis was used to calculate the specific surface and the corresponding pore size distribution. Fourier transform infrared (FT-IR) transmission spectra were obtained on a Nicolet iS50 IR spectrophotometer. X-ray photoelectron spectroscopy (XPS) measurements (ESCALAB 250) were performed to analyze the surface species and their chemical states.

Fabrication of the PVA/KOH all-solid-state supercapacitors

The PVA/KOH gel electrolyte was prepared as follows: in a typical process, 2 g of PVA was dissolved in 20 mL water with continuous stirring for about 3 h at 80℃. After a clear viscous solution was obtained, 6 mL of NaOH aqueous solution (6M) was slowly added to the above solution drop by drop in which the resulting solution was continuously stirred for 2h at 80℃. The sandwich structured device was assembled in the following way: The sandwich structured device was assembled in the following way: The two as-prepared electrodes were soaked in the PVA/KOH gel electrolyte for 20 min and then pressed together on a sheet out roller after the excess water was vaporized.

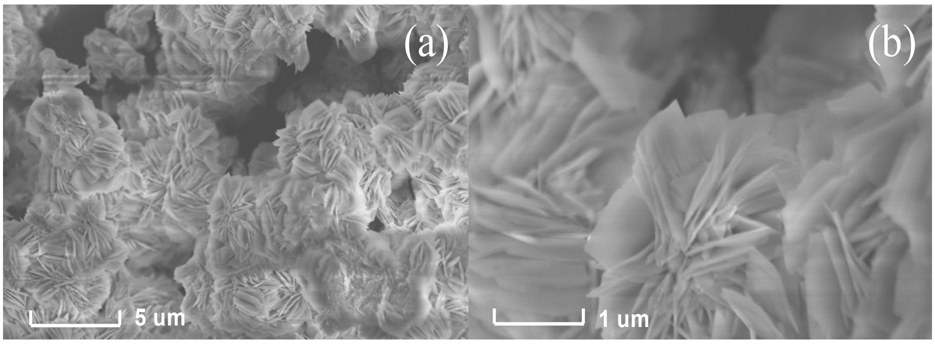

Fig. S1 (a, b) SEM images of the Ni-MOF.

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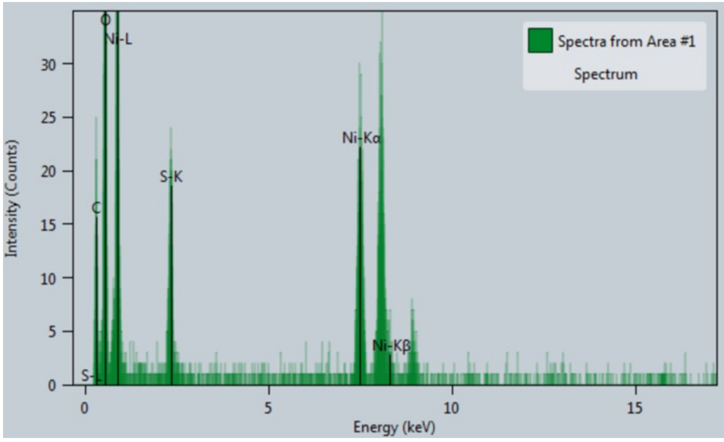

Fig. S2 The analysis of spectrum from EDS.

Table S1 The fitting results of Ni-MOF obtained from the analysis of spectrum.

Z	Element	Atomic Fraction (%)	Atomic Error (%)	Mass Fraction (%)	Mass Error (%)
6	C	26.29	4.32	13.90	1.32
8	O	47.37	12.18	33.95	7.46
16	S	10.39	2.57	13.89	2.89
28	Ni	15.95	3.34	38.25	6.16

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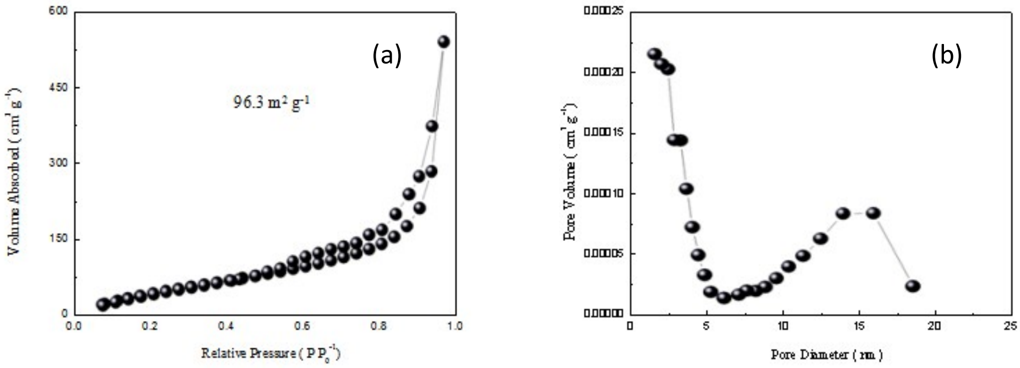

Fig. S3 (a) Nitrogen adsorption and desorption isotherms and (b) the corre sponding mesoporous analysis of Ni-MOF.

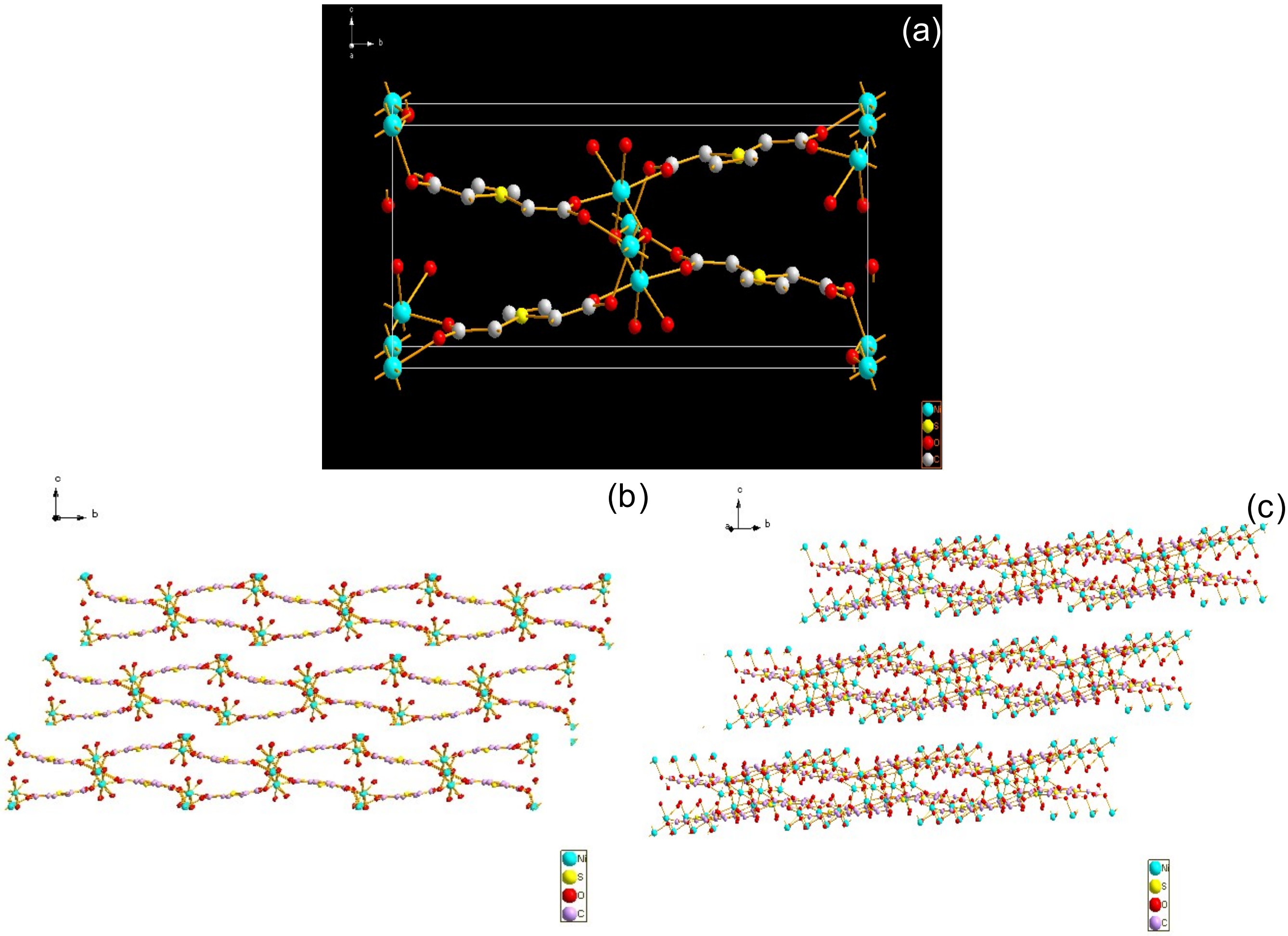

Fig. S4 (a) The crystal structure of the $Ni_3(OH)_2(C_2H_6O_4S)_2$ unit cell, (b, c) The structural model of $Ni_3(OH)_2(C_2H_6O_4S)_2$.

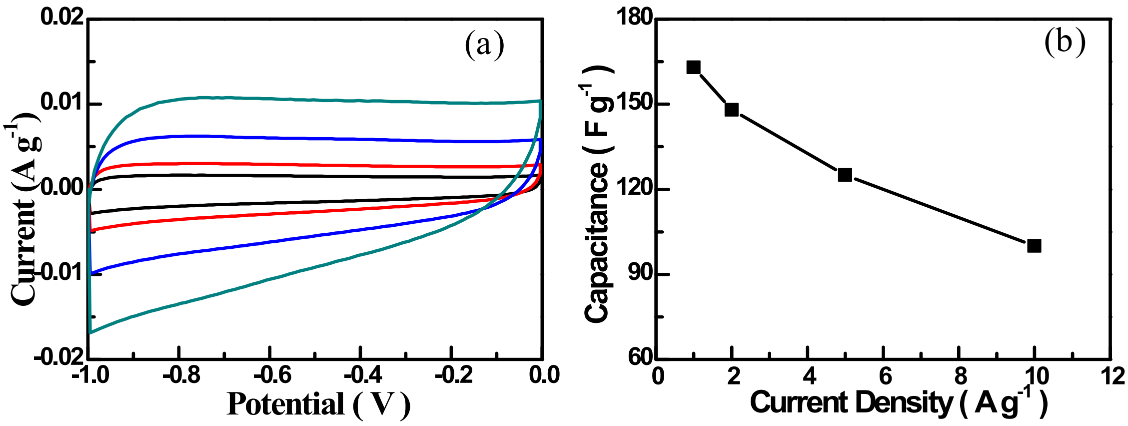

Fig. S5 (a) CV curves of the AC electrode at different scan rates in 2 M NaOH electrolyte , and (b) specific capacitances of the Ni-MOF electrode derived from the discharge curves at different current densities.

Table S2 Electrochemical performance comparison between recently reported studies and the present work.

Ni-MOFs	Capacitance (liquid electrolyte)	**FASCs**	Capacitance of **FASCs**	**Maximum energy density**	**Maximum power density**	**Ref.**
Ni-BDC	1127 F g^{-1} at 1 A g^{-1}	-	-	-	-	*Chem. Eur. J.*, 2017, **23**, 631-636.
Ni-BTC	726 F g^{-1} at 1 A g^{-1}	-	-	-	-	*Chinese.Chemical. Letters*, 2014, **25**, 957-961.
Ni-ADC-BADCO	522 F g^{-1} at 1 A g^{-1}	-	-	-	-	*Nano Energy*, 2016, **26**, 66-73.
Ni-PEDOT	-	Ni-PEDOT//PVA-H_3PO_4//AC	102 mF cm^{-2} at 2 A cm^{-2}	0.051 mW h cm^{-3}	2.1 mW cm^{-3}	*ChemistrySelect*, 2016, **1**, 285-289.
nMOF-867	726 F g^{-1} at 1 A g^{-1}	nMOF-867 //PVA-KOH//AC	197.6 mF cm^{-2} at 1 A cm^{-2}	0.0604 mW h cm^{-3} (0.00385 mW h cm^{-2})	1.097 mW cm^{-3} (0.0567 mW cm^{-2})	*RSC Adv*., 2014, **4**, 36366-36371.
CFs@UiO-66/PPY	90 F g^{-1} at 1 A g^{-1}	CFs@UiO-66/PPY//PVA-LiCl//CFs@UiO-66/PPY	93 mF cm^{-2} at 10 A cm^{-2}	0.0128 mW h cm^{-2}	2.102 mW cm^{-2}	*J. Am. Chem. Soc.*, 2015, **137**, 4920-4923.
PANI-ZIF-67-CC	436 F g^{-1} at 1 A g^{-1}	PANI-ZIF-67-CC//PVA-H_2SO4//PANI-ZIF-67-CC	56.2 mF cm^{-2} at 2 A cm^{-2}	0.0044 mW h cm^{-2}	0.0182 mW h cm^{-2}	*J. Colloid. Interf. Sci.*, 2019, **539**, 370-378.
S@Ni-BDC	1453.5 F g^{-1} at 1 A g^{-1}	S@Ni-BDC//PVA-KOH//AC	136.5 mF cm^{-2} at 0.6 A cm^{-2}	56.85 W h kg^{-1}	480 W kg^{-1}	*J. Mater. Chem. A*, 2018, **6**, 20254-20266.
ZIF-67−PPy	597.6 F g^{-1} at 0.5 A g^{-1}	ZIF-67−PPy//PVA-Na_2SO_4//ZIF-67−PPy	122.8 mF cm^{-2} at 1 A cm^{-2}	0.0113 mW h cm^{-2}	0.472 mW cm^{-2}	*Mater. Chem. Phys.*, 2016, **179**, 166-173.
Ni-TDA	1518.8 F g^{-1} at 1 A g^{-1}	**Ni-TDA//PVA-KOH//AC**	224 mF cm^{-2} at 0.5 A cm^{-2}	**0.0508 mW h cm^{-2} (0.7246 mW h cm^{-2}, 78.2 W h kg^{-1})**	**4.172 mW cm^{-2} (57.34 mW cm^{-2} 634.8 W kg^{-1})**	**This work**

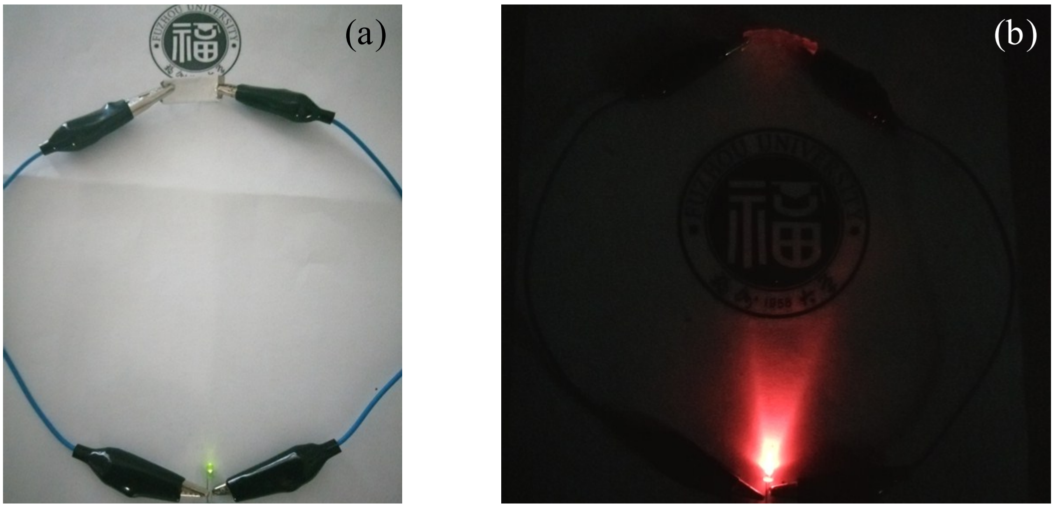

Fig. S6 the images of two LEDs can be powered for 4 min.

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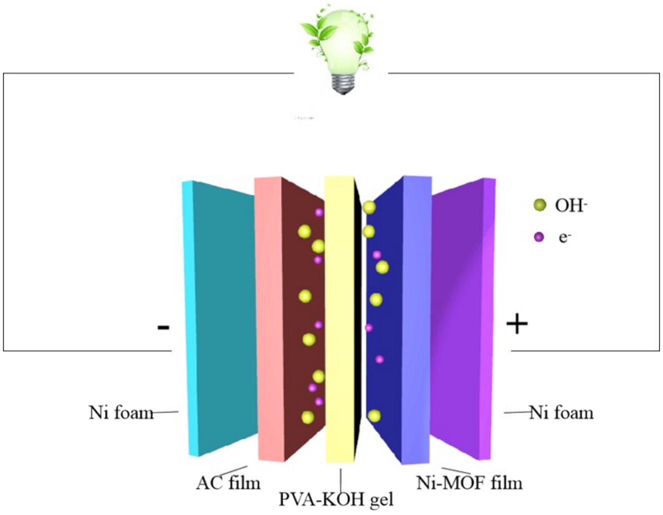

Fig. S7 Schematic diagram of the device structure for all-solid-state supercapacitors.

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