Supplementary Information

Durable and efficient BTC-assisted 2D/0D of Al-Ni-MOF nanostructures for modern electrochemical energy systems

Xiaolong Leng[a,#], S. V. Prabhakar Vattikuti[b,#], Mohan Rao Tamtam [c,*], Jaesool Shim[a,*], Huynh Thanh Liem[d,e,*], Nam Nguyen Dang[d,e]

[a]School of Mechanical Engineering, Hubei Engineering University, Xiaogan 432000, China.

[b]School of Mechanical Engineering, College of Engineering, Yeungnam University, Gyeongsan 38541, Republic of Korea.

[c]School of Computer Science and Engineering, College of Digital Convergence, Yeungnam University, Gyeongsan 38541, Republic of Korea.

[d]Future Materials & Devices Lab., Institute of Fundamental and Applied Sciences, Duy Tan University, Ho Chi Minh City 70000, Viet Nam.

[e]The Faculty of Environmental and Chemical Engineering, Duy Tan University, Danang 50000, Viet Nam.

*E-mail: mohanrtam@gmail.com, jshim@ynu.ac.kr, huynhthanhliem@duytan.edu.vn

[#] Authors equally contributed

1.1 Characterization

X-ray diffraction (XRD; PANalytical) experiments were conducted using programmable PANalytical X-ray generator operated at 40 kV and 40 mA. Scanning electron microscopy (SEM; Hitachi-S-4800) was employed to observe the surface morphologies of the catalysts. High-resolution transmission electron microscopy (HRTEM; Tecnai G2 F20 S-Twin) was performed to examine the nanocrystal shape and size. Energy-dispersive X-ray (EDX)-HRTEM was employed to distinguish the individual elements. X-ray photoelectron spectroscopy (XPS; Thermo Scientific photoelectron spectrometer) was employed to define the surface states.

1.2 Electrochemical testing

All the electrochemical measurements were carried out on a Biologic Sp-200 electrochemical workstation with a standard three-electrode setup. As-synthesized samples were drop casted on Ni foam and used the working electrode. The Hg/HgO electrode and Platinum mesh were served as the reference electrode and the counter electrode, respectively. CV, galvanostatic charge-discharge (GCD), and EIS were used to assess the electrochemical activity of the electrodes. The CV tests were performed at several scan rates, ranging from 1 to 20 mV/s at a potential of 0 to 0.6 V. The GCD tests were executed within the range of 0 to 0. 5 V versus Hg/HgO at various current densities (0.8-4 A/g). All the electrochemical experiments were performed on a Biologic SP-200 electrochemical workstation in a 1.0 M KOH solution.

Specific capacitance (C_s) from Galvanostatic charge-discharge can also be calculated as

$$C_s = \frac{I\Delta t}{m\Delta V} \qquad (1)$$

where, 'I' implies current intensity and 'Δt' entails discharge time (t) of the experimental charge-discharge curve. To achieve the mass of active material on each electrode based on their respective capacitance and operating voltage window. The formula is:

$$m^+/m^- = (C^- \times \Delta V^-) / (C^+ \times \Delta V^+) \qquad (2)$$

where, m^+ and m^-: Represent the mass of active material in the positive and negative electrodes,

respectively. C^+ and C^-: Represent the specific capacitance of the positive and negative electrodes, respectively. ΔV^+ and ΔV^-: Represent the potential window (voltage range) of the positive and negative electrodes, respectively. The optimal mass ratio of positive electrode to negative one was ~0.32.

Further specific energy (E) in Wh/kg associated with specific power (P) in W/kg of electrode were evaluated from the charge-discharge dimensions using the reckoning,

$$E = \frac{1}{2}\left[\frac{C_s \Delta V^2}{3.6}\right] \qquad (3)$$

$$P = \frac{3600 \times E}{\Delta t} \qquad (4)$$

1.3 PVA-KOH gel electrolyte

In order to prepare the alkaline PVA/KOH gel electrolyte, 3 g of polyvinylalcohol was dissolved in 24 mL of pure deionized water at 90 °C with continuous and vigorous stirring to obtain a clear solution. After 1 h, we obtained a clear viscous solution. KOH (3 g) was liquefied in 6 mL deionized water and then dropped into the cleared PVA solution with continuous stirring until complete dissolution and the formation of a gel-like solution. Finally, the PVA/KOH gel electrolyte was cooled to room temperature for further use.

1.4 Asymmetric device preparation procedure (ADs)

For an aqueous asymmetric supercapacitor, the Al-Ni-MOF//AC was used as both positive and negative electrodes. The wide potential window of 0-1.6 V was applied for the two-electrode system and gel electrolyte was used as the electrolyte. The positive electrode (7 mg of active material, 2 mg of carbon black, and 1 mg of PVDF, paste was prepared and drop casted on 14 mm diameter of Ni foam. The paste was then coated on the respective electrodes, and heated at 80°C for 12 h.

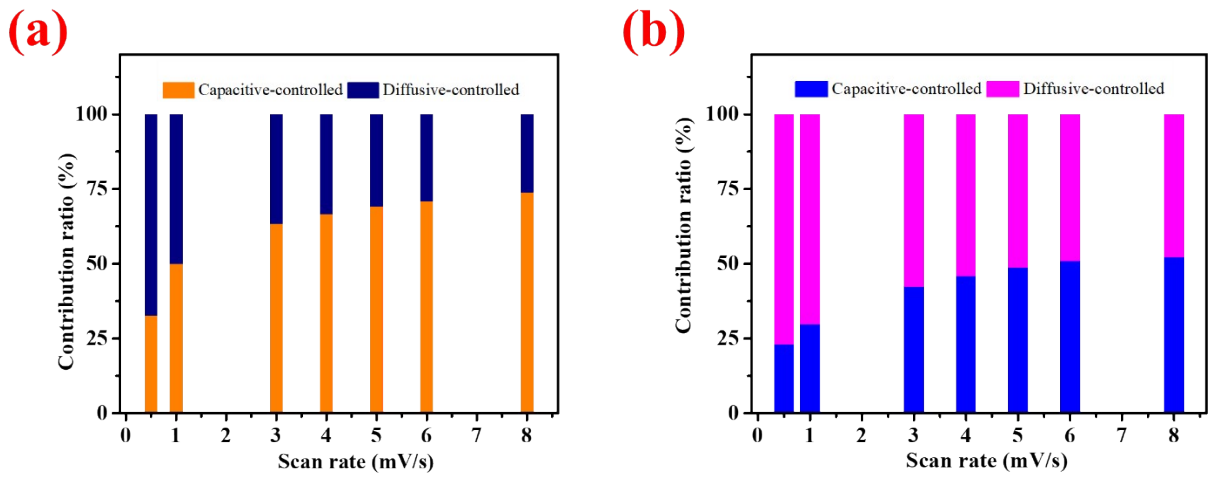

Fig. S1 Total contribution vs scan rate of (a) Al-MOF and (b) Ni-MOF electrodes.

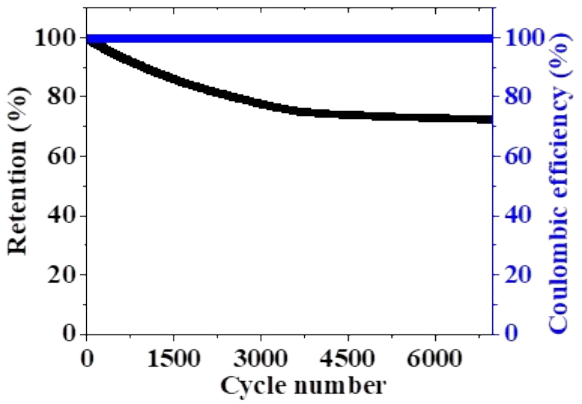

Fig. S2 Cycling stability and coulombic efficiency of the Al-Ni-MOF electrode.

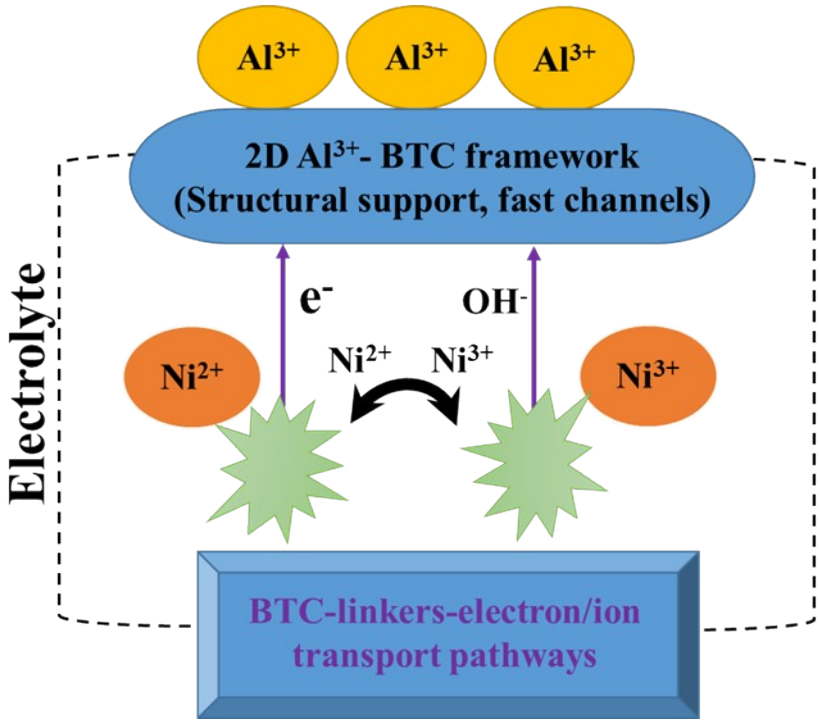

Fig. S3 Graphical representation of electrochemical storage mechanism of Al-Ni-MOF electrode.

Table: S1 Comparison of electrochemical activity of Al-Ni-MF electrode and other MOF-based electrodes in previous reports.

Materials	**Electrolyte**	**Scan rate or Current density**	**Sp. Capacitance (F/g)**	**References**
Mn-BDC MOF	1M Na_2SO_4	1 A/g	178	[1]
ZIF-8/PANI	1 M H_2SO_4	1 A/g	236	[2]
Co-Ni-MOF	2 M KOH	5 mA/cm^2	43.6 F/cm^2	[3]
Cu MOF/rGO	PVA-Na_2SO_4	1 A/g	385	[4]
UIO-66/rGO	6 M KOH	0.15 A/g	302	[5]
PANI-ZIF-67-CC	3 M KCl	10 mV/s	21.46 mF/cm^2	[6]
ZIF-67/polyprrole nanotubes	1M Na_2SO_4	0.5 A/g	597.6	[7]
Ni-CoMOF	3 M KOH	1 A/g	236.1 mAh/g	[8]
Al-MOF	1 M KOH	0.8 A/g	112 F/g	[This work]
Al-Ni-MOF	1 M KOH	0.8 A/g	1631.2 F/g	[This work]

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