## **Supporting Information for**

## 2D MOF Nanoflake-Assembled Spherical-Micro-Structures for Enhanced Supercapacitor and Electrocatalysis Performances

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**Fig. S1 a** SEM images of ZIF-67. **b** TEM images of Ni/Co-MOF nanoflakes. **c** TEM images of Ni-MOF nanoflakes. **d** TEM-EDS mapping of Ni-MOF nanoflakes, and **e** TEM-EDS mapping of ZIF-67



Fig. S2 XPS spectra of Ni/Co-MOF nanoflakes



**Fig. S3**. **a** CV of as-prepared ZIF-67 eletrodes at different scan rates. **b** CV of as-prepared Ni-MOF nanoflakes eletrodes at different scan rates. **c** Galvanostatic charge/discharge curves of ZIF-67 at various current densities. **d** Galvanostatic charge/discharge curves of Ni-MOF nanoflakes at various current densities



Fig. S4 Galvanostatic charge/discharge curves of Ni/Co-MOF nanoflakes supercapacitor before and after 2000 and 3000 cycles measured at  $2 \text{ A g}^{-1}$ 



**Fig. S5** Electrochemical impedance spectra measured in the frequency range of 10 mHz to 100 kHz at the open circuit voltage -0.1V with an alternate current amplitude of 5 mV. Inset: Equivalent circuit for the electrode-solution interface.  $C_{\rm I}$ , double-layer capacitance. The Faradic impedance includes  $Z_{\rm w}$  (the Warburg impedance) and  $R_{\rm ct}$  (the charge-transfer resistance).  $R_{\rm s}$ , spreading resistance



**Fig. S6 a, b** RDE polarization curves at different rotation speeds. Scan rate: 10 mV s<sup>-1</sup>. **c, d** Koutech-Levich plots at various potentials

| Table S1 Comparison of the capacities | of Ni/Co-MOF with the recently reported MOF based materials |
|---------------------------------------|---|
|                                       |   |

| Sample         | Electrolyte solution                    | Test<br>Condition       | Specific<br>Capacitance<br>(F g <sup>-1</sup> ) | Resistance (ohm) | Ref.         |
|----------------|---|-------------------------|---|------------------|--------------|
| Ni/Co-MOF      | 1.0 M LiOH                              | $0.5 \text{ Ag}^{-1}$   | 530.4   | ~4.0             | This<br>work |
| Co-MOF         | 1.0 M LiOH                              | $0.5 \text{ A g}^{-1}$  | 230.5   | -                | 23           |
| N-doped Zn-MOF | 6.0 M KOH                               | $0.1 \mathrm{A g^{-1}}$ | 285.8   | ~6               | 48           |
| ZIF-67         | 0.5 M<br>H <sub>2</sub> SO <sub>4</sub> | 20 mV s <sup>-1</sup>   | 238   | -                | 49           |
| Co-MOF         | 6 M<br>KOH                              | 1 A g <sup>-1</sup>     | 321   | ~5.23            | 50           |