Supporting Information

Bimetallic electronic effects of Mn-doped Ni-MOFs shuttle-like nanosheets remarkably enhance supercapacitive performance

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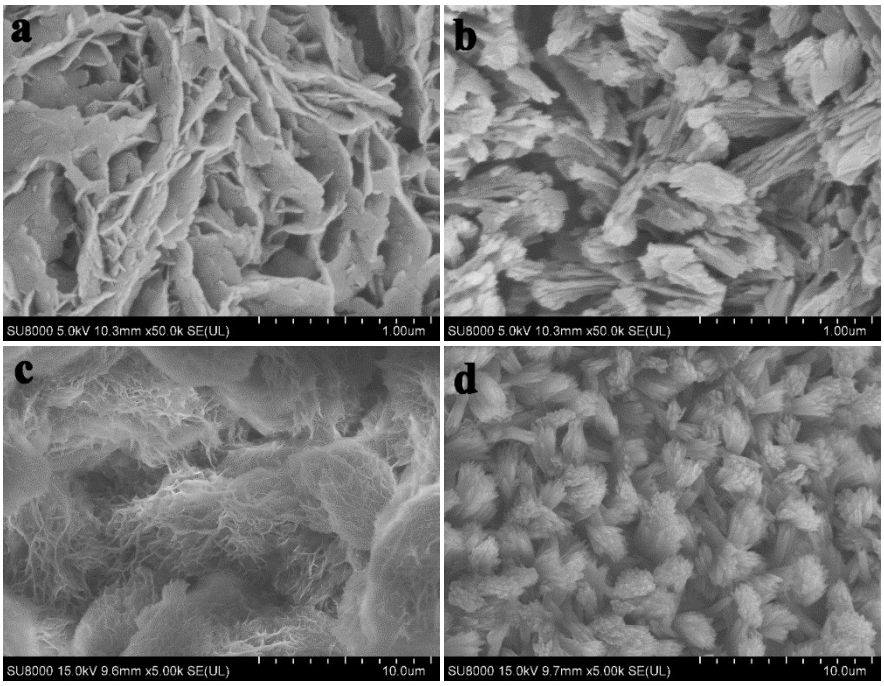

Figure S1. SEM images of (a) $Ni_{0.9}Mn_{0.1}$-MOF, (b) $Ni_{0.4}Mn_{0.6}$-MOF, (c) Ni-MOF and (d) Mn-MOF nanostructures

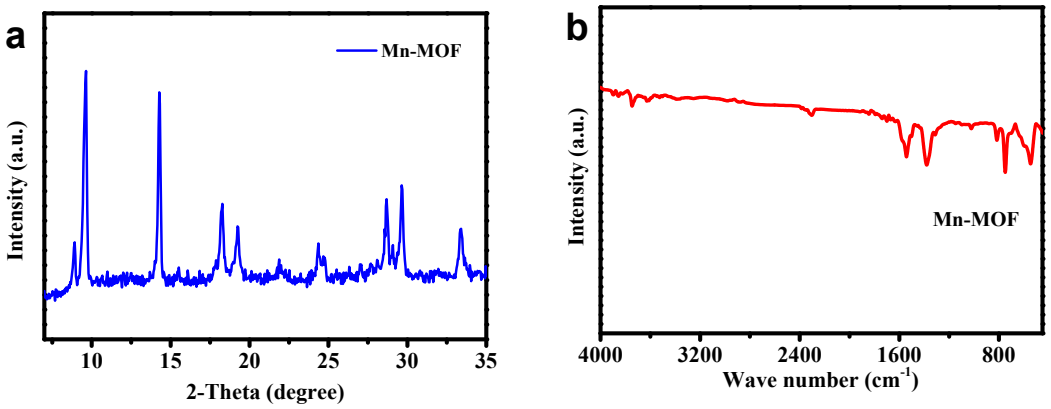

Figure S2. (a) XRD patterns, (b) FT-IR spectra of Mn-MOF.

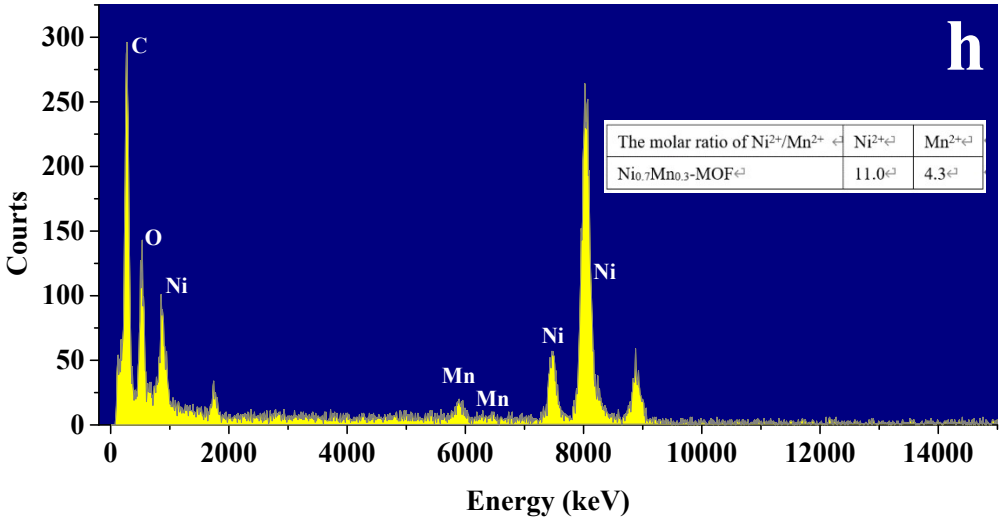

Figure S3. EDS mapping of the $Ni_{0.7}Mn_{0.3}$-MOF. The insert is the molar ratio of the Ni^{2+} and Mn^{2+}

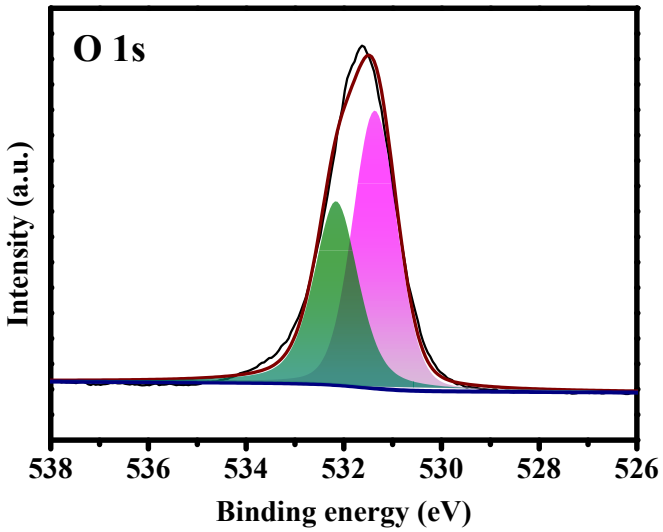

Figure S4. High-resolution XPS spectra for the $Ni_{0.7}Mn_{0.3}$-MOF in O 1s

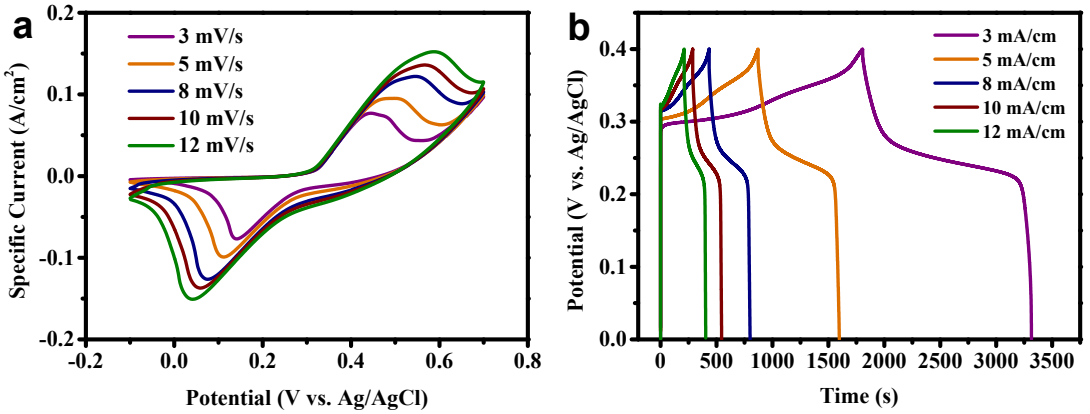

Figure S5. The electrochemical performance of the Ni-MOF electrode in 2 M KOH electrolyte: (a) CV curves, (b) GCD curves.

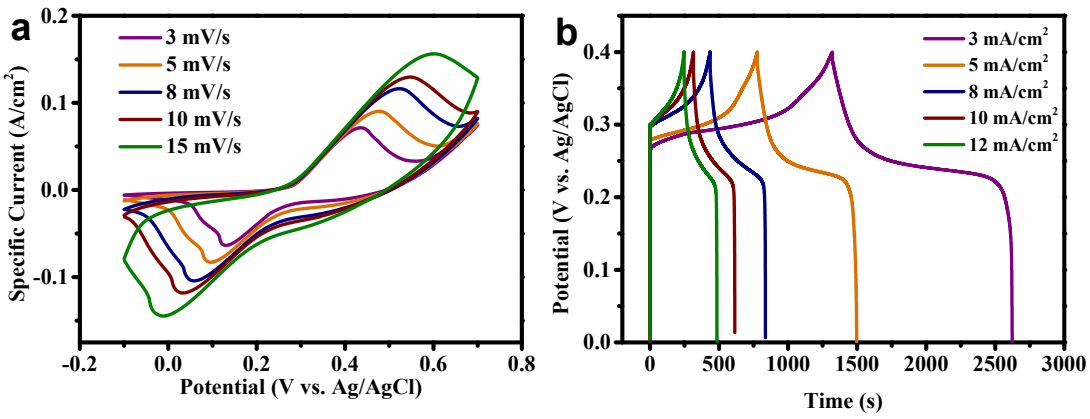

Figure S6. The electrochemical performance of the $Ni_{0.4}Mn_{0.6}$-MOF electrode in 2 M KOH electrolyte: (a) CV curves, (b) GCD curves.

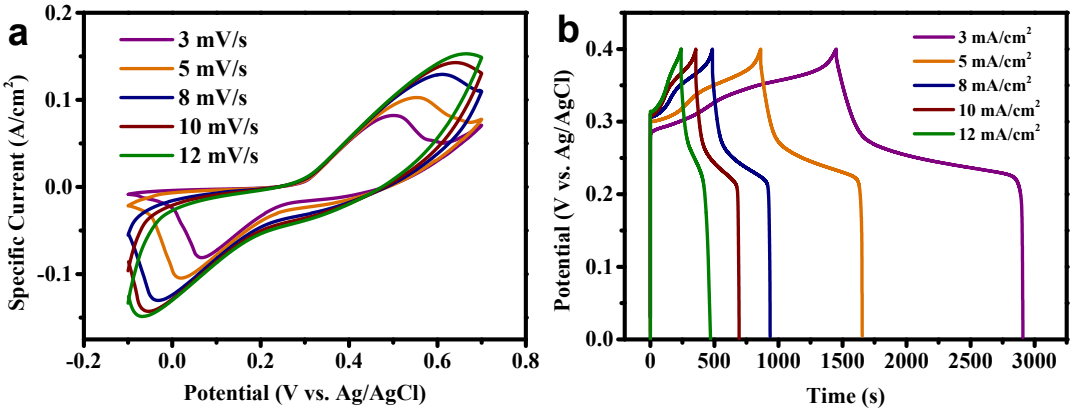

Figure S7. The electrochemical performance of the $Ni_{0.9}Mn_{0.1}$-MOF electrode in 2 M KOH electrolyte: (a) CV curves, (b) GCD curves.

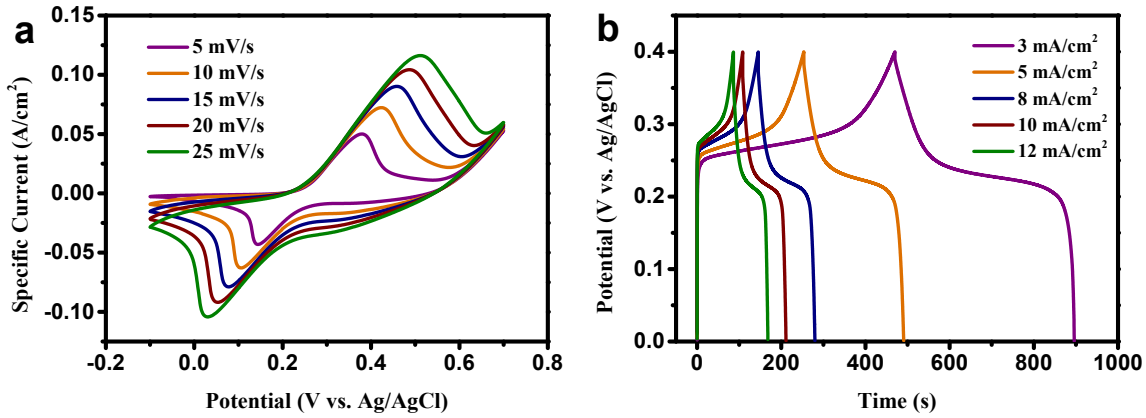

Figure S8. The electrochemical performance of the Mn-MOF electrode in 2 M KOH electrolyte: (a) CV curves, (b) GCD curves.

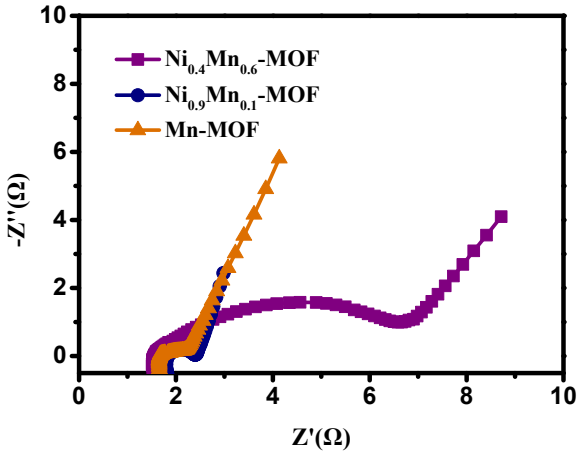

Figure S9. EIS curves of the $Ni_{0.4}Mn_{0.6}$-MOF, $Ni_{0.9}Mn_{0.1}$-MOF and Mn-MOF.

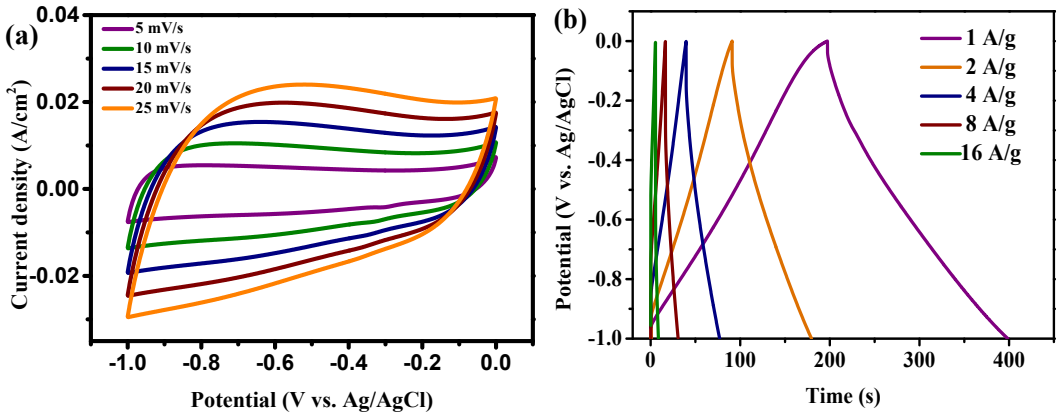

Figure S10. The electrochemical performance of the NF/AC electrode in 2 M KOH electrolyte: (a) CV curves, (b) GCD curves.

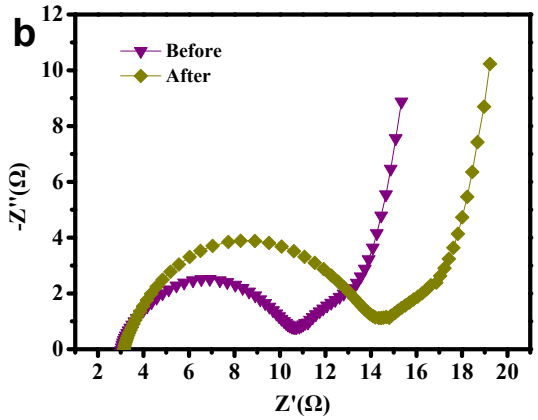

Figure S11. EIS curves of the NF/$Ni_{0.7}Mn_{0.3}$-MOF//AC HSC device before and after 10000 cycles.

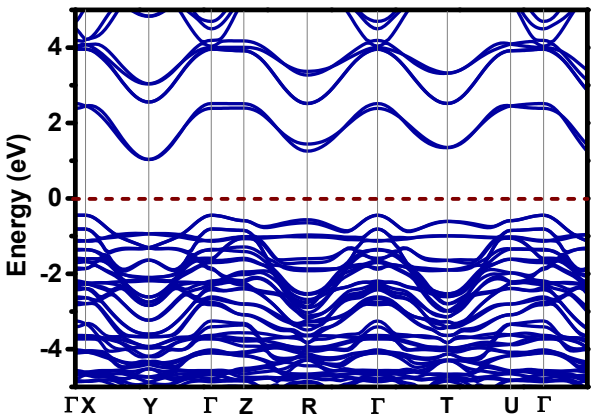

Figure S12. Calculated the energy band curve of Ni-MOFs.

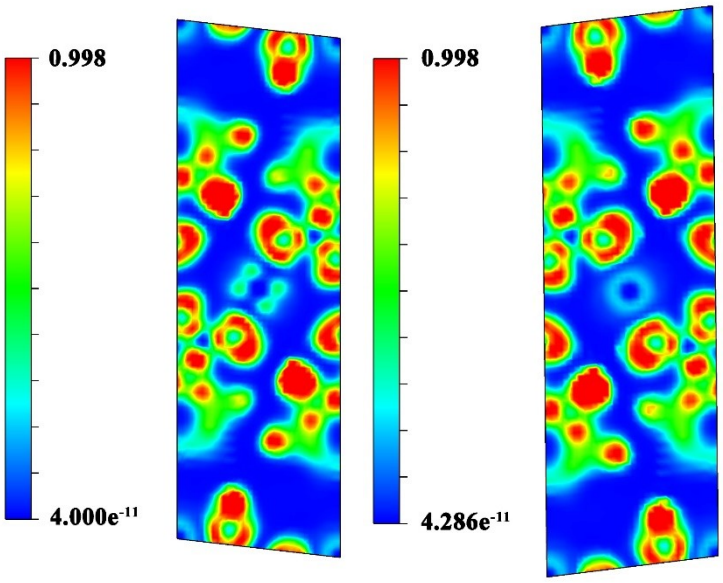

Figure S13. Calculated the electron localization density function plot.

References:

[S1] C. Qu, B. Zhao, Y. Jiao, D. C. Chen, S. G. Dai, B. M. Deglee, Y. Chen, K. S. Walton, R. Q. Zou and M. L. Liu, Functionalized bimetallic hydroxides derived from metal-organic frameworks for high-performance hybrid supercapacitor with exceptional cycling stability, *ACS Energy Lett.,* 2017, **2**, 1263-1269.

[S2] J. Yang, C. Zheng, P. X. Xiong, Y. F. Li and M. D. Wei, Zn-doped Ni-MOF material with a high supercapacitive performance, *J. Mater. Chem. A,* 2014, **2**, 19005-19010.

[S3] J. Wang, Q. Zhong, Y. Q. Zeng, D. Y. Cheng, Y. H. Xiong and Y. F. Bu, Rational construction of triangle-like nickel-cobalt bimetallic metal-organic framework nanosheets arrays as battery-type electrodes for hybrid supercapacitors, *J. Colloid Interface Sci.,* 2019, **555**, 42-52.

[S4] Y. Seo, P. A. Shinde, S. Park and S. C. Jun, Self-assembled bimetallic cobalte-manganese metale organic framework as a highly efficient, robust electrode for asymmetric supercapacitors, *Electrochim. Acta,* 2020, **335**, 135327.

[S5] P. A. Shinde, Y. Seo, S. Lee, H. S. Kim, Q. N. Pham, Y. Won and S. C. Jun, Layered manganese metal-organic framework with high specific and areal capacitance for hybrid supercapacitors, *Chem. Eng. J.,* 2020, **387**, 122982.

[S6] P. C. Du, Y. M. Dong, C. Liu, W. L. Wei, D. Liu and P. Liu, Fabrication of hierarchical porous nickel based metal-organic framework (Ni-MOF) constructed with nanosheets as novel pseudo-capacitive material for asymmetric supercapacitor, *J. Colloid Interface Sci.,* 2018, **518**, 57-68.

[S7] Q. Bi, Q. X. Ma, K. Tao and L. Han, Hierarchical core-shell 2D MOF nanosheet hybrid arrays for high-performance hybrid supercapacitors, *Dalton Trans.,* 2021, **50**, 8179.

[S8] L. L. Lei, X. C. Zhang, Y. T. Su, S. S. Wu and J. Shen, Metal-organic framework (MOF)-assisted construction of core-shell nanoflower-like CuO/CF@NiCoMn-OH for high-performance supercapacitor, *Energy Fuels,* 2021, **35**, 8387-8395.

[S9] J. Wang, Q. Zhong, Y. H. Xiong, D. Y. Cheng, Y. Q. Zeng and Y. F. Bu, Fabrication of 3D Co-doped Ni-based MOF hierarchical micro-flowers as a high-performance electrode material for supercapacitors, *Appl. Surf. Sci.,* 2019, **483**, 1158-1165.

[S10] Y. Jiao, J. Pei, D. H. Chen, C. S. Yan, Y. Y. Hu, Q. Zhang and G. Chen, Mixed-metallic MOF based electrode materials for high performance hybrid supercapacitors, *J. Mater. Chem. A,* 2017, **5**, 1094-1102.

[S11] Y. D. Zhang, B. P. Lin, Y. Sun, X. Q. Zhang, H. Yang and J. C. Wang, Carbon nanotubes@metal-organic frameworks as Mn-based symmetrical supercapacitor electrodes for enhanced charge storage, *RSC Adv.,* 2015, **5**, 58100-58106.