## Supporting Information for

## Hybrid NiO-CuO mesoporous nanowires array with abundant oxygen vacancies and hollow structure for high-performance asymmetric supercapacitor

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Materials	Specific capacitance (F g <sup>-1</sup> )	Areal capacitance (F cm <sup>-2</sup> )	Current density or scan rate	Electrolyte	Reference
NiO	302	-	1 A g <sup>-1</sup>	6 M KOH	<b>S</b> 1
CuO	431	1.51	3.5 mA cm <sup>-2</sup>	3 М КОН	S2
Cu2O/CuO/Co <sub>3</sub> O <sub>4</sub>	318	-	$0.5  \text{A g}^{-1}$	3 M KOH	<b>S</b> 3
NiO/NiMn-LDH	937	-	$0.5  \text{A g}^{-1}$	3 М КОН	S4
CuCo <sub>2</sub> O <sub>4</sub> /CuO	781	-	2 mV s-1	1 M KOH	S5
Ni0.99Cu0.01O	559	-	0.3 A g-1	6 M KOH	<b>S</b> 6
Ni/NiO	526	-	1 A g <sup>-1</sup>	3 M KOH	S7
NiO/a-Ni(OH) <sub>2</sub>	707	-	$2 \text{ A g}^{-1}$	2 M KOH	<b>S</b> 8
Ni-Co binary hydroxide	1030	-	1 mg cm <sup>-2</sup>	6 M KOH	S9
NiO@MnO <sub>2</sub>	266.7	-	$0.5 \mathrm{A  g^{-1}}$	2 M KOH	S10
NiO-Co <sub>3</sub> O <sub>4</sub>	801	-	1 A g <sup>-1</sup>	3 M KOH.	S11
ZnO-NiO	649	-	5.8 A g <sup>-1</sup>	3 M KOH	S12
Cu/Ni-based manganese dioxide	374	-	$0.25 \text{ Ag}^{-1}$	1 M Na <sub>2</sub> SO4	S13
NiCo <sub>2</sub> O <sub>4</sub> nanowires	743	-	1 A g <sup>-1</sup>	1 M KOH	S14
NiCu(OH) <sub>2</sub> CO <sub>3</sub>	971	-	1 A g <sup>-1</sup>	6 M KOH	S15
NiO-CuO	1450.8	4.35	2 mA cm <sup>-2</sup>	3 M KOH	Our work

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## References

[S1] X. Ren, C. Guo, L. Xu, T. Li, L. Hou and Y.Wei, *ACS Appl. Mater. Inter.*, 2015, 7, 19930-19940.

[S2] S. E. Moosavifard, M. F. El-Kady, M. S. Rahmanifar, R. B. Kaner and M. F. Mousavi, *ACS Appl. Mater. Inter.*, 2015, 7, 4851-4860.

[S3] M. Kuang, T. T. Li, H. Chen, S. M. Zhang, L. L. Zhang and Y. X. Zhang, *Nanotechnology*, 2015, 26, 304002.

[S4] P. F. Liu, J. J. Zhou, G. C. Li, M. K. Wu, K. Tao, F. Y. Yi, W. N. Zhao and L. Han *Dalton. Trans.*, 2017, 46, 7388-7391.

[S5] A. Shanmugavani and R. K. Selvan, *Electrochim. Acta*, 2016, 188, 852-862.

[S6] G. Yuan, Y. Liu, M. Yue, H. Li, E. Liu, Y. Huang and D. Kong, *Ceram. Int.*, 2014, 40, 9101-9105.

[S7] Y. Zhang, M. Park, H. Y. Kim and S. J. Park, J. Colloid Interf. Sci., 2017, 500, 155-163.

[S8] B. K. Kim, V. Chabot and A. Yu, *Electrochim. Acta*, 2013, 109, 370-380.

[S9] X. Sun, G. Wang, H. Sun, F. Lu, M. Yu and J. Lian, J. Power Sources, 2013, 238, 150-156.

[S10] J. Chen, Y. Huang, C. Li, X. Chen and X. Zhang, *Appl. Surf. Sci.*, 2016, 360, 534-539.

[S11] X. W. Wang, D. L. Zheng, P. Z. Yang, X. E. Wang, Q. Q. Zhu, P. F. Ma and L. Y. Sun, *Chem. Phys. Lett.*, 2017, 667, 260-266.

[S12] H. Pang, Y. Ma, G. Li, J. Chen, J. Zhang, H. Zheng and W. Du, *Dalton. Trans.*, 2012, 41, 13284-13291.

[S13] H. Chen, X. Q. Qi, M. Kuang, F. Dong and Y. X. Zhang, *Electrochim. Acta*, 2016, 212, 671-677.

[S14] H. Jiang, J. Ma, and C. Li, Chem. Commun., 2012, 48, 4465-4467.

[S15] X. Zheng, Y. Ye, Q. Yang, B. Geng and X. Zhang, *Chem. Eng. J.*, 2016, 290: 353-360.