Supporting Information

Ultrahigh-rate, ultralong-life asymmetric supercapacitors based on few-

crystalline, porous NiCo₂O₄ nanosheet composites

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Fig. S1 FESEM images of the $NiCo_2O_4$ at (a) low and (b) high magnifications. TEM images of the $NiCo_2O_4$ at (c) low and (d) high magnifications.

ASC type	C _T (F g ⁻¹)	C _T (F g ⁻¹)	E _{max}	P _{max}	Cycle stability	Reference
/Potential	(small current)	(large current)				
CQDs/ NiCo ₂ O ₄	88.9 (0.5 A g ⁻¹)	42.1 (40 A g ⁻¹ , 47.4%)	27.8 Wh kg-1	10.2 kW kg-1	101.9%	1
//AC (1.5 V)			(128 W kg-1)	(13.1 W h kg ⁻¹)	(5,000 cycles, 3 A g ⁻¹)	
NiCo ₂ O ₄ /CNTs	105 (1 A g ⁻¹)	55 (20 A g ⁻¹ , 52.4%)	28.58 Wh kg-1	14.18 kW kg-1	Almost 100%	2
//AC (1.4 V)			(700 W kg ⁻¹)	(14.18 Wh kg ⁻¹)	(3,000 cycles, 2 A g ⁻¹)	
NiCo ₂ O ₄ @ Au	~55 (1 A g ⁻¹)	~15 (5 A g ⁻¹ , 27.3%)	19.56 Wh kg-1	2.12 kW kg ⁻¹	79.1%	3
nanotubes(1.45 V)			(782.6 W kg ⁻¹)	(2.75 Wh kg ⁻¹)	(4,000 cycles, 2 A g ⁻¹)	
NiCo ₂ O ₄ /MnO ₂	31.3 (0.25 A g-	24 (4 A g ⁻¹ , 76.7%)	9.4 Wh kg-1	2.5 kW kg-1	89.7%	4
//AC (1.4 V)	1)		(175 W kg ⁻¹)	(5.8 Wh kg ⁻¹)	(3,000 cycles, 5 A g ⁻¹)	
NiCo ₂ O ₄ //AC	54 (0.5 A g ⁻¹)	25 (8 A g ⁻¹ , 46.3%)	14.7 Wh kg-1	2.8 kW kg ⁻¹	85%	5
(1.4 V)			(175 W kg ⁻¹)	(6.8 Wh kg ⁻¹)	(5,000 cycles, 1.5 A g ⁻¹)	
NiCo ₂ O ₄ @HMRA	49.3 (1 A g ⁻¹)	20.4 (10.4 A g ⁻¹ ,	15.4 Wh kg-1	7.8 kW kg-1	106% (2,500 cycles, 100	6
//AC (1.5 V)		41.5%)			mV s ⁻¹)	
NiCo ₂ O ₄ /NGN/	128 (1 A g ⁻¹)	80.4 (10 A g ⁻¹ , 62.8%)	42.7 Wh kg-1	15.5 kW kg-1	86% (10,000 cycles,	7
CNTs//NGN/CNT			(775 W kg ⁻¹)	(24.7 Wh kg ⁻¹)	4 A g ⁻¹)	
(1.55 V)						
CNT@NiO//PCPs	72 (0.5 A g ⁻¹)	28 (20 A g ⁻¹ , 38.9%)	25.4 Wh kg-1	16 kW kg-1	93%	8
(1.6 V)			(400 W kg ⁻¹)	(9.8 Wh kg ⁻¹)	(10,000 cycles, 6 A g ⁻¹)	
GF-CNT @ NiO	65.8 (1 A g ⁻¹)	20.1 (8 A g ⁻¹ , 30.5%)	23.4 Wh kg-1	7.14 kW kg-1	81.7%	9
//G-CNT			(1060 W kg ⁻¹)	(11.9 Wh kg ⁻¹)	(30,000 cycles, 4A g ⁻¹)	
CNT/Ni(OH) ₂	78.3 (1 A g ⁻¹)	58.3 (30 A g ⁻¹ , 74.5%)	35.2 Wh kg-1	27.1 kW kg-1		10
//RGO (1.8 V)			(1807 W kg ⁻¹)	(26.2 Wh kg ⁻¹)		
Ni(OH) ₂ /AC/CNT	82.1 (0.5 A g ⁻¹)	~40 (10 A g ⁻¹ , 48.7%)	32.3 Wh kg-1	~8 kW kg ⁻¹	83.5%	11
// AC (1.6 V)			(504.8 W kg ⁻¹)	(14.2 Wh kg ⁻¹)	(1,000 cycles, 10 A g ⁻¹)	
Ni-Co Hydroxide	108 (1 A g ⁻¹)	66.7 (10 A g ⁻¹ , 61.8%)	33.8 Wh kg-1	7.5 kW kg-1	84.4%	12
/Graphene/Ni			(750 W kg ⁻¹)	(20.8 Wh kg ⁻¹)	(2,000 cycles, 4 A g ⁻¹)	
Foam//AC (1.5 V)						
Ni(OH) ₂ //AC	153 (5 mV s ⁻¹)	~50 (50 mV s ⁻¹ , 32.7%)	35.7 Wh kg-1	1.67 kW kg-1	81%(10,000 cycles, 100	13
(1.3 V)			(490 W kg ⁻¹)	(12.6 Wh kg ⁻¹)	mV s ⁻¹)	
Co ₃ O ₄ //Carbon	101 (2 A g ⁻¹)	43 (10 A g ⁻¹ , 42.6%)	36 Wh kg ⁻¹	8 kW kg ⁻¹	89%	14
(1.6 V)			(1600 W kg ⁻¹)	(15 Wh kg ⁻¹)	(2,000 cycles, 5 A g ⁻¹)	
Co ₃ O ₄ @MnO ₂ //	49.8 (1 A g ⁻¹)	~10 (10 A g ⁻¹ , 20.1%)	17.7 Wh kg-1	~10 kW kg-1	81.1%	15
MEGO (1.6 V)			(~800 W kg-1)	(~3.5 Wh kg ⁻¹)	(10,000 cycles, 3 A g ⁻¹)	
FCP-	107.8 (1 A g ⁻¹)	77.5 (10 A g ⁻¹ , 72.0%)	38.1 Wh kg-1	58.1 kW kg-1	104.8% (20,000 cycles)	This work
NiCo ₂ O ₄ /RGO/		66.5 (20 A g ⁻¹ , 61.7%)	797.8 W kg ⁻¹	13.3 Wh kg-1	81.2% (50,000 cycles)	
CNTs//AC		50.1 (50 A g ⁻¹ , 46.5%)			20 A g ⁻¹	
(1.6 V)		45.5 (80 A g ⁻¹ , 42.2%)				

Table S2 Comparison of electrochemical performances between the FCP-NiCo $_2O_4$ /RGO/CNT composite and the previously reportedresults based on the Ni and/or Co-based materials in a three-electrode system.

Sample	C _s (F g ⁻¹)	C _s (F g ⁻¹)	Rate capability	Reference
	(low current)	(large current)		
NiCo ₂ O ₄ /3D graphene	2300 (1 A g ⁻¹)	711 (20 A g ⁻¹)	30.9%	16
NiCo ₂ O ₄ nanowires	1283 (1 A g ⁻¹)	1010 (20 A g ⁻¹)	78.7%	17
/Carbon Textiles				
NiCo ₂ O ₄ /3D graphene foam	1402 (1 A g ⁻¹)	1080 (20 A g ⁻¹)	77.0%	18
NiCo ₂ O ₄ /CNTs	680 (1 A g ⁻¹)	480 (50 A g ⁻¹)	70.6%	19
NiCo ₂ O ₄ nanosheets @	678 (6 A g ⁻¹)	367 (47 A g ⁻¹)	54.1%	6
hollow microrod arrays				
NiCo ₂ O ₄ nanowires	1197 (1 A g ⁻¹)	625 (8 A g ⁻¹)	52.2%	20
(microemulsion technique)				
NiCo2O4 nanocrystal-based	1113 (1 A g ⁻¹)	765 (20 A g ⁻¹)	68.7%	21
electrode				
NiCo ₂ O ₄ /CNTs films	828 (1 A g ⁻¹)	656 (20 A g ⁻¹)	79.2%	2
CNS/ NiCo ₂ O ₄ core-shell	1420 (1 A g ⁻¹)	1018 (10 A g ⁻¹)	71.7%	22
sub-microspheres				
Core-shell NiCo ₂ O ₄ @	1325.9 (2 mA cm ⁻²)	730.9 (40 mA cm ⁻²)	55.1%	23
NiMoO ₄ nanowires				
Ni foam/N-CNT/ NiCo ₂ O ₄	1472 (1 A g ⁻¹)	1074.5 (30 A g ⁻¹)	73%	24
nanosheets				
Ni/Co Oxide (microsphere)	696 (1 A g ⁻¹)	614 (10 A g ⁻¹)	88.2%	25
NiCo ₂ O ₄ @ MnMoO ₄ Core-	1118 (1 A g ⁻¹)	746 (10 A g ⁻¹)	66.7%	26
shell Flower				
NiCo2S4 ball-in-ball	1036 (1 A g ⁻¹)	705 (20 A g ⁻¹)	68.5%	27
hollow spheres				
Co ₃ O ₄ /RGO	458 (0.5 A g ⁻¹)	416 (2 A g ⁻¹)	90.8%	28
Co ₃ O ₄ /RGO/CNTs	378 (2 A g ⁻¹)	297 (8 A g ⁻¹)	78.6%	29
CNTs @ NiO	996 (1 A g ⁻¹)	500 (20 A g ⁻¹)	50.2%	8
NiO/RGO/CNTs	1180 (1 A g ⁻¹)	840 (10 A g ⁻¹)	71.2%	30
FCP-NiCo2O4/RGO/CNTs	1618 (1 A g ⁻¹)	1283.2 (10 A g ⁻¹)	79.3% (10A g ⁻¹),	This work
		1164.2 (20 A g ⁻¹),	72.0% (20A g ⁻¹),	
		1079.7 (30 A g ⁻¹),	66.7% (30A g ⁻¹),	
		917.8 (50 A g ⁻¹)	56.7% (50 A g ⁻¹)	



Fig. S2 XPS spectra of O 1s for the $NiCo_2O_4$ nanobelts.



Fig. S3 HRTEM images of $NiCo_2O_4$ nanobelts at different magnifications.



Fig. S4 Nitrogen adsorption/desorption isotherms of the FCP-NiCo₂O₄/RGO/CNT composite and the NiCo₂O₄ nanobelts.



Fig. S5 (a) CV curves of the $NiCo_2O_4$ electrode at scan rates from 5 to 100 mV s⁻¹. (b) and (c) CP curves of the $NiCo_2O_4$ electrode at current densities from 1 to 50 A g⁻¹.



Fig. S6 (a) CV curves of AC electrode at scan rates from 10 to 50 mV s⁻¹. (b) CP curves of AC electrode at current densities from 1 to 20 A g^{-1} .

Current density (A g ⁻¹)	Specific capacitance (F g ⁻¹)			
1	281.5			
2	268.2			
5	251.5			
10	235.7			
20	214.2			





Fig. S7 A Ragone plot for NiCo₂O₄//AC and FCP-NiCo₂O₄/RGO/CNTs//AC ASC.



Fig. S8 Electrochemical behavior of the NiCo₂O₄//AC ASC. (a) and (b) CV curves of NiCo₂O₄//AC ASC at scan rates from 10 mV s⁻¹ to 100 mV s⁻¹. (c) and (d) CP curves of the NiCo₂O₄//AC at current densities from 1 to 50 A g⁻¹.



Fig. S9 A TEM image of the positive electrode (FCP-NiCo₂O₄/RGO/CNT composite) of our ASC device after 50,000 cycles, indicates that the NiCo₂O₄ held well the morphology of porous nanosheets, without visible pulverization.



Fig. S10 Cycling stability of the NiCo₂O₄ //AC at 20 A g⁻¹.

References

- 1 Y. Zhu, Z. Wu, M. Jing, H. Hou, Y. Yang, Y. Zhang, X. Yang, W. Song, X. Jia and X. Ji, *J. Mater. Chem. A*, 2014, **3**, 866–877.
- 2 S. Xu, D. Yang, F. Zhang, J. Liu, A. Guo and F. Hou, *Rsc Adv.*, 2015, 5, 74032-74039.
- 3 J. Zhu, Z. Xu and B. Lu, *Nano Energy*, 2014, 7, 114–123.
- 4 M. Kuang, Z. Q. Wen, X. L. Guo, S. M. Zhang and Y. X. Zhang, *J. Power Sources* 2014, **270**, 426–433.
- 5 D. Rui, Q. Li, M. Jia and H. Wang, *Electrochim. Acta*, 2013, **107**, 494-502.
- K. F. Lu, D. J. Wu, R. Z. Li, Q. Li, S. H. Ye, Y. X. Tong and G. R. Li, *J. Mater. Chem. A*, 2014, 2, 4706–4713.
- 7 S. Yue, H. Tong, L. Lu, W. Tang, W. Bai, F. Jin, Q. Han, J. He, J. Liu and X. Zhang, *J. Mater. Chem. A*, 2017, **5**, 689–698.
- 8 H. Yi, H. Wang, Y. Jing, T. Peng and X. Wang, *J. Power Sources* 2015, 285, 281-290.
- 9 C. Guan, Y. Wang, Y. Hu, J. Liu, K. H. Ho, Z. Fan, W. Zhao, Z. Shen, H. Zhang and J. Wang, J. Mater. Chem. A, 2015, 3, 23283-23288.

- 10 R. R. Salunkhe, J. Lin, V. Malgras, S. X. Dou, J. H. Kim and Y. Yamauchi, *Nano Energy*, 2015, 11, 211-218.
- 11 L. Sui, S. Tang, Y. Chen, Z. Dai, H. Huangfu, Z. Zhu, X. Qin, Y. Deng and G. M. Haarberg, *Electrochim. Acta*, 2015, **182**, 1159-1165.
- 12 Y. Bai, W. Wang, R. Wang, J. Sun and L. Gao, J. Mater. Chem. A, 2015, 3, 12530-12538.
- 13 L. Huang, D. Chen, Y. Ding, S. Feng, Z. L. Wang and M. Liu, *Nano letters*, 2013, **13**, 3135-3139.
- 14 R. R. Salunkhe, J. Tang, Y. Kamachi, T. Nakato, J. H. Kim and Y. Yamauchi, *Acs Nano*, 2015, **9**, 6288-6296.
- 15 M. Huang, Y. Zhang, F. Li, L. Zhang, Z. Wen and Q. Liu, J. Power Sources 2014, 252, 98-106.
- 16 S. Sun, S. Wang, S. Li, Y. Li, Y. Zhang, J. Chen, Z. Zhang, S. Fang and P. Wang, J. Mater. Chem. A, 2016, 4, 18646–18653.
- 17 L. Shen, Q. Che, H. Li and X. Zhang, Adv. Funct. Mater., 2014, 24, 2630-2637.
- 18 C. Zhang, T. Kuila, N. H. Kim, S. H. Lee and J. H. Lee, *Carbon*, 2015, 89, 328-339.
- 19 S. Abouali, M. A. Garakani, Z. L. Xu and J. K. Kim, *Carbon*, 2016, **102**, 262-272.
- 20 C. An, Y. Wang, Y. Huang, Y. Xu, L. Jiao and H. Yuan, *Nano Energy*, 2014, **10**, 125-134.
- 21 M. Rui, X. Li, L. Gan, T. Zhai and H. Zeng, Adv. Funct. Mater., 2016, 26, 5051-5060.
- 22 D. Li, Y. Gong, Y. Zhang, C. Luo, W. Li, Q. Fu and C. Pan, *Scientific reports*, 2015, 5, 12903.
- 23 L. Huang, W. Zhang, J. Xiang, H. Xu, G. Li and Y. Huang, Sci. Rep., 2016, 6, 31465.
- 24 J. Wu, P. Guo, R. Mi, X. Liu, H. Zhang, H. Liu, J. Mei, L. W. M. Lau and L. M. Liu, *J. Mater. Chem. A*, 2015, **3**, 15331-15338.
- 25 Y. Zhao, X. Zhang, J. He, L. Zhang, M. Xia and F. Gao, *Electrochim. Acta*, 2015, 174, 51-56.
- 26 Z. Gu and X. Zhang, *J. Mater. Chem. A*, 2016, **4**, 8249-8254.
- 27 L. Shen, L. Yu, H. B. Wu, X. Y. Yu, X. Zhang and X. W. Lou, Nat. Commun. , 2015, 6, 6694.
- 28 C. Xiang, M. Li, M. Zhi, A. Manivannan and N. Wu, J. Power Sources 2013, 226, 65 70.
- 29 C. Yuan, Y. Long, L. Hou, J. Li, Y. Sun, X. Zhang, L. Shen, X. Lu, S. Xiong and W. L. Xiong, *Adv. Funct. Mater.*, 2012, 22, 2560 - 2566.
- 30 Y. Bai, M. Du, J. Chang, J. Sun and L. Gao, *J. Mater. Chem. A*, 2014, **2**, 3834-3840.