Flexible, Sandwich-Like CNTs/NiCo₂O₄ Hybrid Paper Electrodes for All-Solid State Supercapacitors

Yongjia Zheng¹, Zhiqiang Lin¹, Wenjun Chen¹, Binghao Liang¹, Huiwei Du¹, Rongliang

Yang¹, Xianfeng He¹, Zikang Tang^{1, 2}, Xuchun Gui^{1,*}

¹State Key Lab of Optoelectronic Materials and Technologies, School of Electronics and Information Technology, Sun Yat-sen University, Guangzhou, 510275, P. R. China

²Institute of Applied Physics and Materials Engineering, University of Macau, Avenida da

Universidade, Taipa, Macau, China

*Corresponding authors: <u>guixch@mail.sysu.edu.cn</u>;



Figure S1. SEM images of CNTs/NiCo₂O₄ hybrid paper synthesized with different solvothermal time.



Figure S2. Cross-sectional SEM images of $CNTs/NiCo_2O_4$ hybrid paper synthesized with different solvothermal time.



Figure S3. (a) Raman spectra of NiCo₂O₄ (NCO), CNT buckypaper (CNT) and CNTs/NiCo₂O₄ (CNT/NCO) hybrid paper. (b) N_2 adoption and desorption isotherms of the CNTs/NiCo₂O₄ sample synthesized with the solvothemal time of 2 hours; the inset is the pore size distributions of the sample.



Figure S4. (a) XPS spectrum of $CNTs/NiCo_2O_4$ showing the signals for Ni, Co, O and C elements. (b) Core level spectra of the Ni 2p region. (c) Core level spectra of the Co 2p region. (d) Core level spectra of the C 1s region. (e) Core level spectra of the O 1s region.



Figure S5. (a) and (d) CNTs buckypaper characterized by CV curves at scan rates varying from 5 to 50 mV/s and GCD curves at different current densities from 0.5 to 2 A/g. (b) and (e) The CNT/NCO-1h electrode characterized by CV curves at scan rates varying from 5 to 50 mV/s and GCD curves at different current densities varying from 1 to 20 A/g. (c) and (f) The CNT/NCO-6h electrode characterized by CV curves at scan rates varying from 5 to 50 mV/s and GCD curves at different current densities varying from 5 to 50 mV/s and GCD curves at scan rates varying from 1 to 20 A/g.



Figure S6. Ragone plot of CNT/NCO-1h hybrid paper electrode.



Figure S7. (a) and (b) SEM images, and (c) Raman spectrum of the CNT/NCO-2h electrode after 4000

cycles of CV scans.



Figure S8. Electrochemical performance of all-solid-state supercapacitors assembled with the CNT/NCO-1h electrodes. (a) CV curves of the supercapacitors at a scan rate of 50 mV/s under bending at various angles.(b) Galvanostatic charge/discharge curves of the supercapacitors at different current densities.

No.	Electrode materials	Preparation method	Specific	Rate	Capacity
			capacitance (F/g)	performance	retention
[1]	NiCo ₂ O ₄ @NiCo ₂ O	Solvothermal and	900 (1 A/g)	75% (20 A/g)	98.6% (4000
	₄ core/shell	chemical deposition			cycles)
[2]	NiCo ₂ O ₄ -RGO	Self-assembly and	835 (1 A/g)	74% (16 A/g)	85.7% (4000
		thermal treatment			clcles)
[3]	Ni@NiCo2O4	Solvothermal and	899 (1 A/g)	70.9% (10	84.9% (6000
	nanosheets	thermal treatment		A/g)	cycles)
[4]	Double-shelled	Template method and	972 (5 A/g)	89.5% (10	92.5% (12000
	Co ₃ O ₄ /NiCo ₂ O ₄	thermal treatment		A/g)	cycles)
[5]	NiCo ₂ O ₄	Solvothermal and	1023.6 (1 A/g)	61.5% (10	91.5% (2400
	nanorod/CF	thermal treatment		A/g)	cycles)
[6]	NiCo ₂ O ₄	Solvothermal and	1388 (2 A/g)	85% (10 A/g)	89.4% (12000
	nanosheets	thermal treatment			cycles)
[7]	NiCo ₂ O ₄	Solvothermal and	1743 (8.5	61.1% (25	93.2% (3000
	nanosheets/Ni	thermal treatment	mA/cm ²)	mA/cm ²)	cycles)
	foam				
[8]	NiCo ₂ O ₄	Co-electrodeposition	2010 (2 A/g)	79.4% (12	94% (2300
	nanosheets/Ni	and thermal		A/g)	cycles)
	foam	treatment			
Our	NiCo ₂ O ₄	Solvothermal and	1752.3 (1 A/g)	92% (10 A/g)	95.6% (7000
work	nanosheets/CNT	thermal treatment			cycles)

Table SI Comparison of the electrochemical performance of the NiCo ₂ O ₄ based electrode

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