Electronic Supplementary Information

In-plane β-$Co(OH)_2$/Co_3O_4 hybrid nanosheets for flexible all-solid-state thin-film supercapacitors with high electrochemical performance

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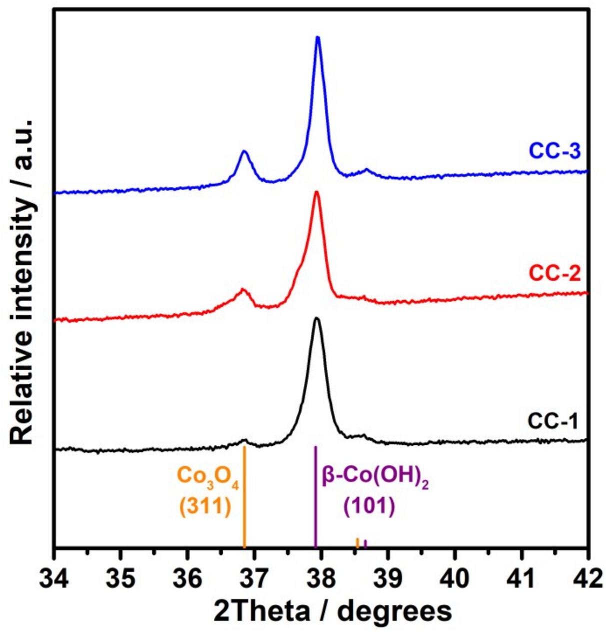

Fig. S1 Enlarged XRD patterns of the products to identify the intensity ratio of (311) diffraction peak of Co_3O_4 to (101) diffraction peak of β-$Co(OH)_2$.

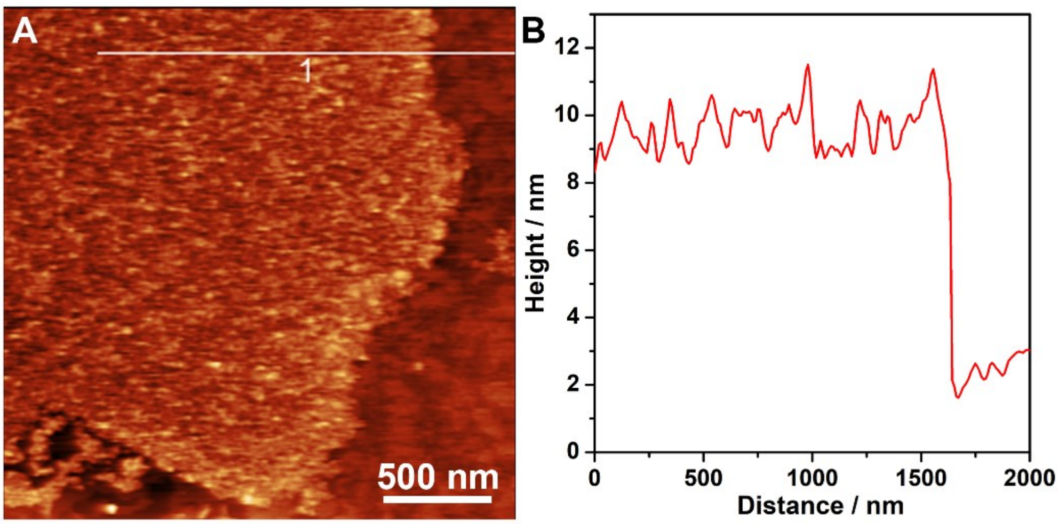

Fig. S2 (A) AFM image and (B) height profile plot of CC-2.

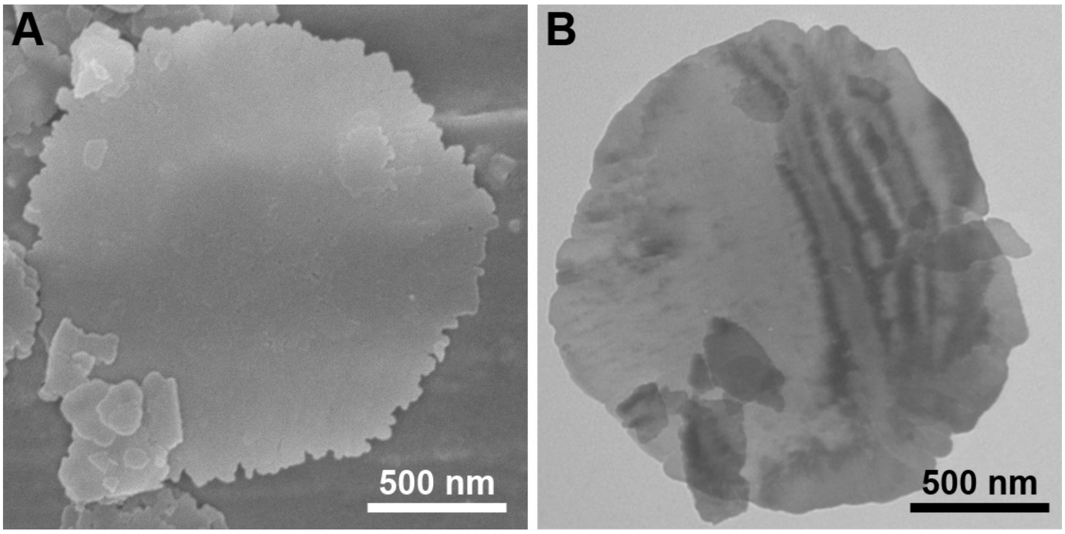

Fig. S3 (A) SEM and (B) TEM image of the β-$Co(OH)_2/Co_3O_4$ hybrid nanosheets synthesized at 80 °C for 6 hours (CC-1).

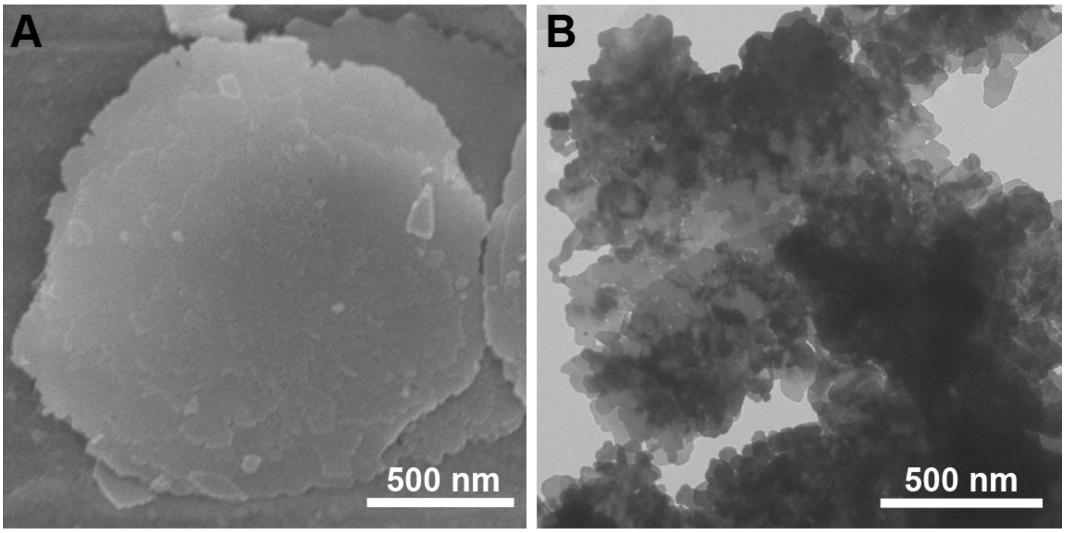

Fig. S4 (A) SEM and (B) TEM image of the β-$Co(OH)_2/Co_3O_4$ hybrid nanosheets synthesized at 120 °C for 12 hours (CC-3).

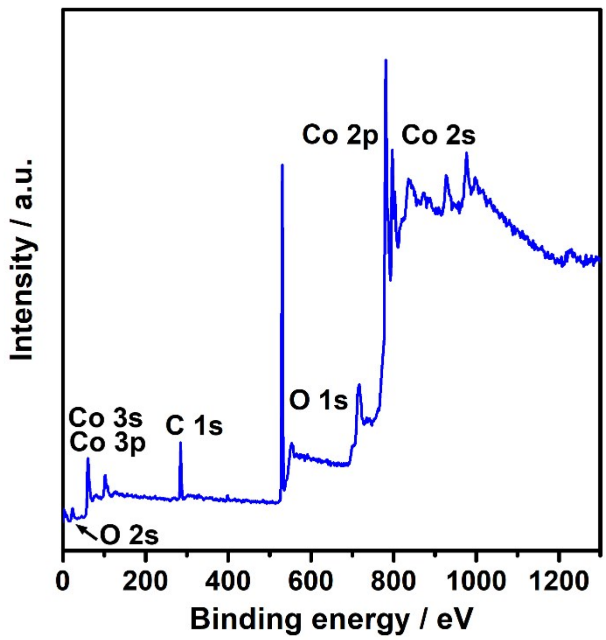

Fig. S5 XPS survey spectrum of CC-2 confirms the compositional purity.

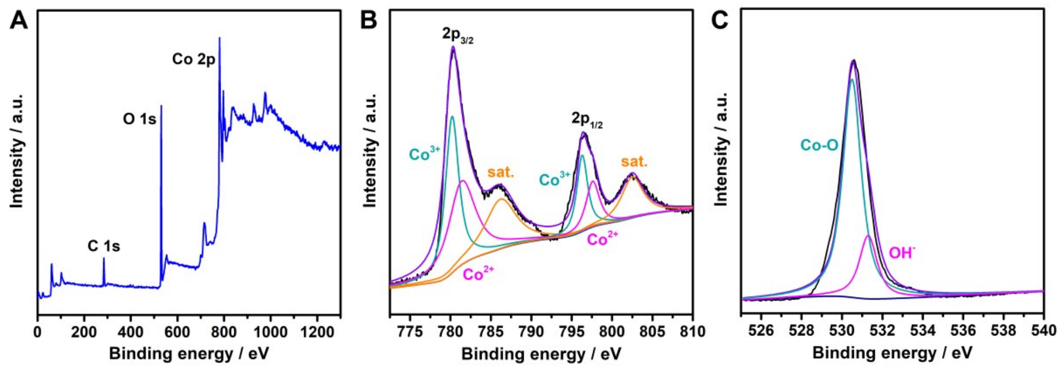

Fig. S6 XPS data of CC-1. (A) XPS survey spectrum. (B) Co 2p spectrum. (B) O 1s spectrum.

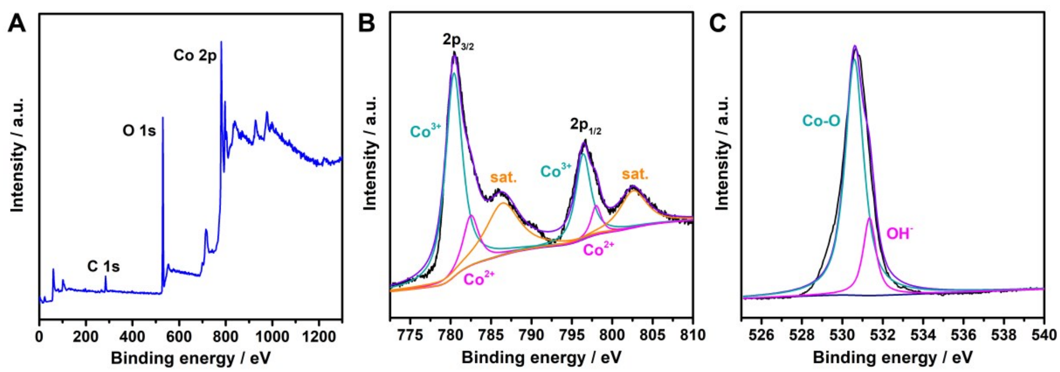

Fig. S7 XPS data of CC-3. (A) XPS survey spectrum. (B) Co 2p spectrum. (B) O 1s spectrum.

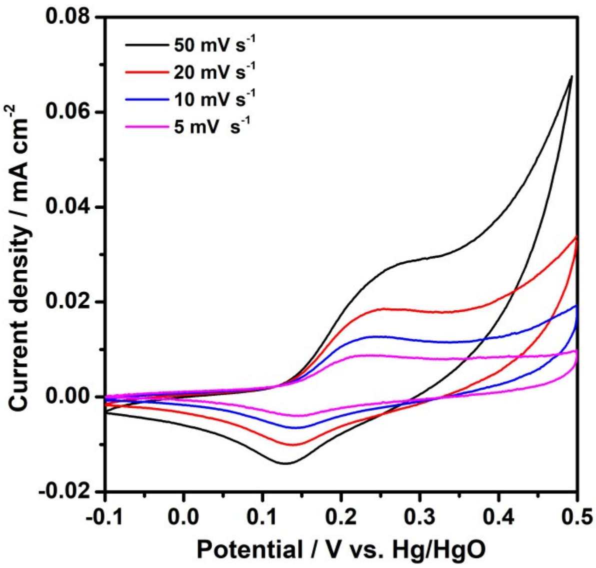

Fig. S8 CV curves of CC-1 at various scan rates.

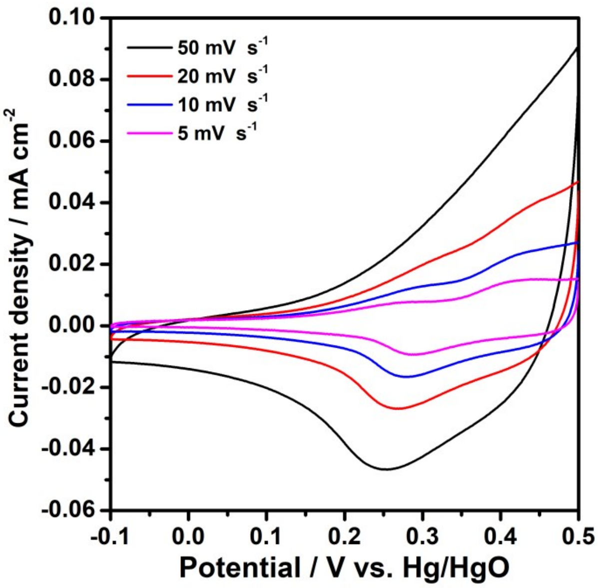

Fig. S9 CV curves of CC-2 at various scan rates.

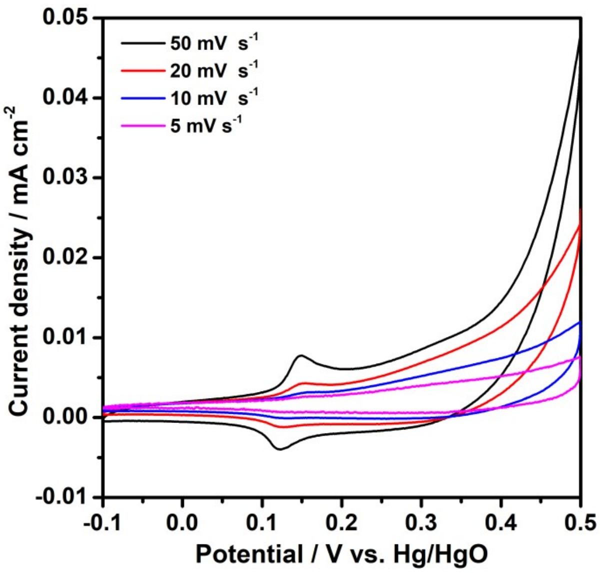

Fig. S10 CV curves of CC-3 at various scan rates.

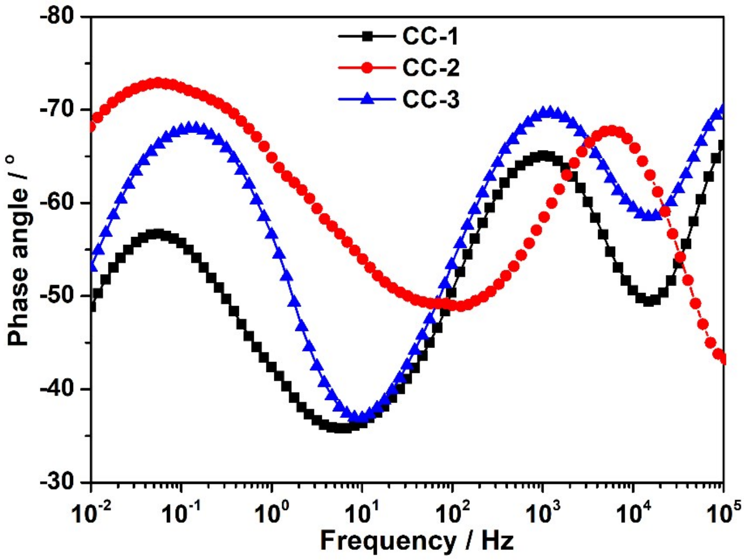

Fig. S11 Bode phase angle plot of the in-plane β-$Co(OH)_2/Co_3O_4$ hybrid nanosheets obtained open-circuit potentials. The phase angle of CC-2 is measured to be close to 73 at low frequency domain, further confirming the excellent capacitive behavior of the β-$Co(OH)_2/Co_3O_4$ hybrid nanosheets

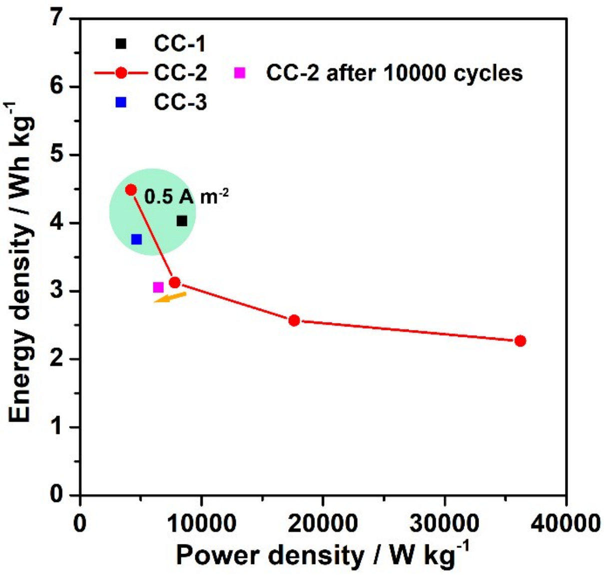

Fig. S12 Ragone plot of the as-fabricated ASSTFSs based on CC-1, CC-2 and CC-3. The energy density and power density of the CC-2-based ASSTFS display only slight degradation even after 10000 cycles, further confirming the superior stability of the CC-2-based ASSTFS device.

Table S1 Comparison of the electrochemical performance of the all-solid-state supercapacitors.

Material	Specific capacitance @ 100 mV s^{-1}	Specific capacitance @ 0.5 A m^{-2}	Parameters of the device	Reference
CC-1	166.3 F g^{-1}	80.6 F g^{-1}	PVA/KOH	this work
CC-2	232.9 F g^{-1}	89.7 F g^{-1}	PVA/KOH	this work
CC-2 after activation	289.1 F g^{-1}	-	PVA/KOH	this work
CC-3	211.7 F g^{-1}	75.2 F g^{-1}	PVA/KOH	this work
β-$Ni(OH)_2$/graphene nanohybrid	-	~0.5 F g^{-1}	thickness=50 nm, 1 cm^2, PVA/ KOH	1
N,B-doped graphene	26.2 F g^{-1}	-	thickness=1 mm, Φ=7-10 mm, PVA/H_2SO_4	2
N-doped graphene	21.7 F g^{-1}	-	thickness=1 mm, Φ=7-10 mm, PVA/H_2SO_4	2
B-doped graphene	21.3 F g^{-1}	-	thickness=1 mm, Φ=7-10 mm, PVA/H_2SO_4	2
graphene paper	6.5 F g^{-1}	-	thickness=1 mm,	2

			Φ=7-10 mm, PVA/H_2SO_4	
graphene	21.2 F g^{-1}	-	thickness=1 mm, Φ=7-10 mm, PVA/H_2SO_4	2
$Co(CO_3)_{0.5}(OH)$ /$Ni_2(CO_3)(OH)_2$ nanobelt	-	72.5 F g^{-1}	acetylene black/ /PVDF/NF, PVA/KOH	3
$NiMoO_4$-PANI	-	74.2 F g^{-1}	acetylene black/ /PVDF/NF, PVA/KOH	4
Ag nanoparticles/ activated carbon/ woven polyester	17.8 F g^{-1}	-	PVA/H_2SO_4	5
NiO nanosheets/ NF //α-Fe_2O_3 nanorods/CC	-	~50.0 F g^{-1}	thickness=0.12 mm, PVA/KOH	6

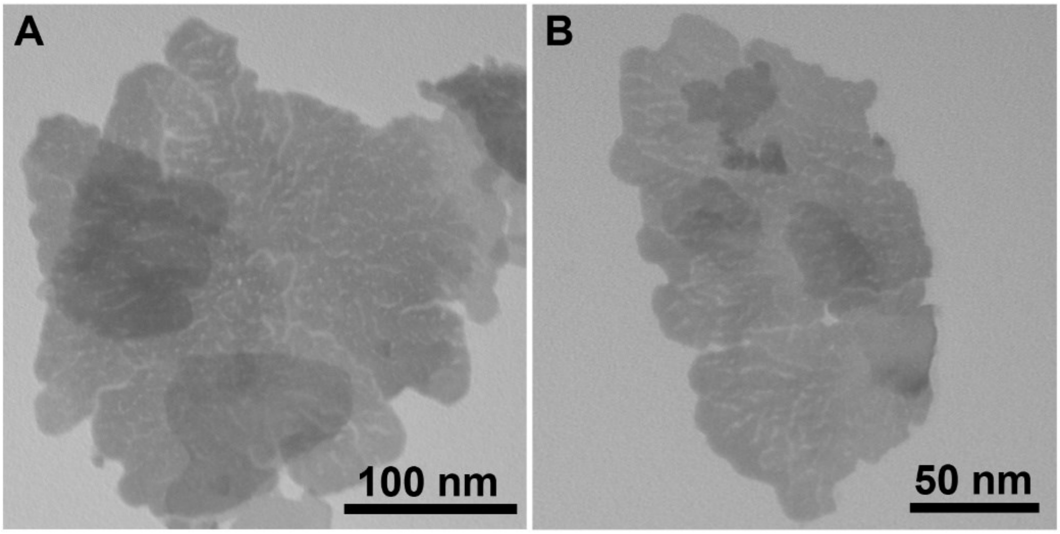

Fig. S13 TEM images of CC-2 hybrid nanosheets after conducting 10000 cycles. As can be seen, more cracks were generated during the activation process, and the surface of the β-$Co(OH)_2$/Co_3O_4 hybrid nanosheets is obviously roughened owing to the repeated surface redox reaction, therefore leading to the exposure of more surface sites for the pseudocapacitive charge storage and thus exhibiting significantly improved performance.

Reference

1. J. Xie, X. Sun, N. Zhang, K. Xu, M. Zhou, Y. Xie, *Nano Energy*. 2013, **2**, 65-74.

2. Z.-S. Wu, A. Winter, L. Chen, Y. Sun, A. Turchanin, X. Feng, K. Müllen, *Adv. Mater.* 2012, **24**, 5130-5135.

3. G. Zhang, P. Qin, R. Nasser, S. Li, P. Chen, J. Song, *Chem. Eng. J.* 2020, **387**, 124029.

4. H. Gao, F. Wu, X. Wang, C. Hao, C. Ge, *Int. J. Hydrogen Energy* 2018, **43**, 18349-18362.

5. Y. Jang, J. Jo, K. Woo, S.-H. Lee, S. Kwon, K.-Y. Kim, D. Kang, *Appl. Phys. Lett.* 2017, **110**, 203902.

6. S. Zhang, B. Yin, Z. Wang, F. Peter, *Chem. Eng. J.* 2016, **306**, 193-203.