Supporting Information

Rational Integration of Inbuilt Aperture with Mesoporous Framework in Unusual Asymmetrical Yolk-Shell Structures for Energy Storage and Conversion

Ting Zhu,[†] Liangliang Zhu,[†] Jing Wang,[†] and Ghim Wei Ho[†‡§*]

[†]Department of Electrical and Computer Engineering, National University of Singapore, 4 Engineering Drive 3, Singapore 117583

[‡]Engineering Science Programme, National University of Singapore, 9 Engineering Drive 1, Singapore 117575

[§]Institute of Materials Research and Engineering, A*STAR (Agency for Science, Technology and Research), 3 Research Link, Singapore117602

Corresponding Author

*Email: elehgw@nus.edu.sg

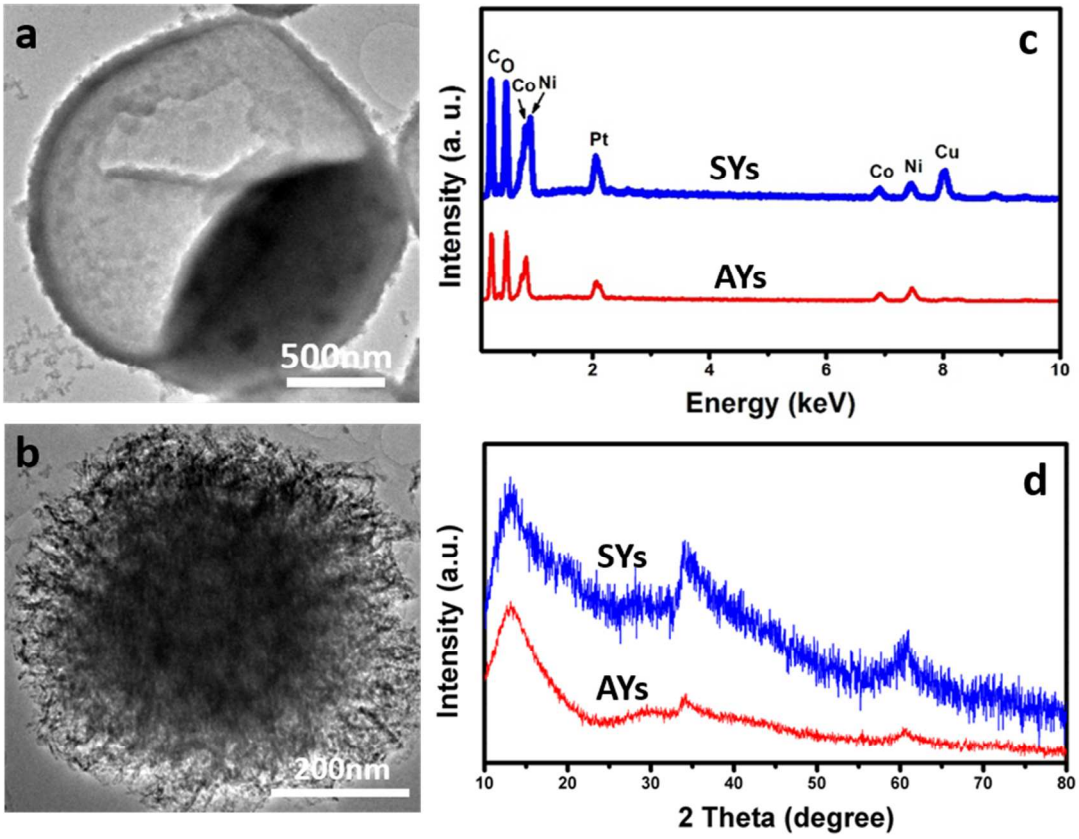

Figure S1. TEM images (a and b) of the SYs (a) and AYs (b), EDX (c) and XRD (d) results of the Ni-Co precursors.

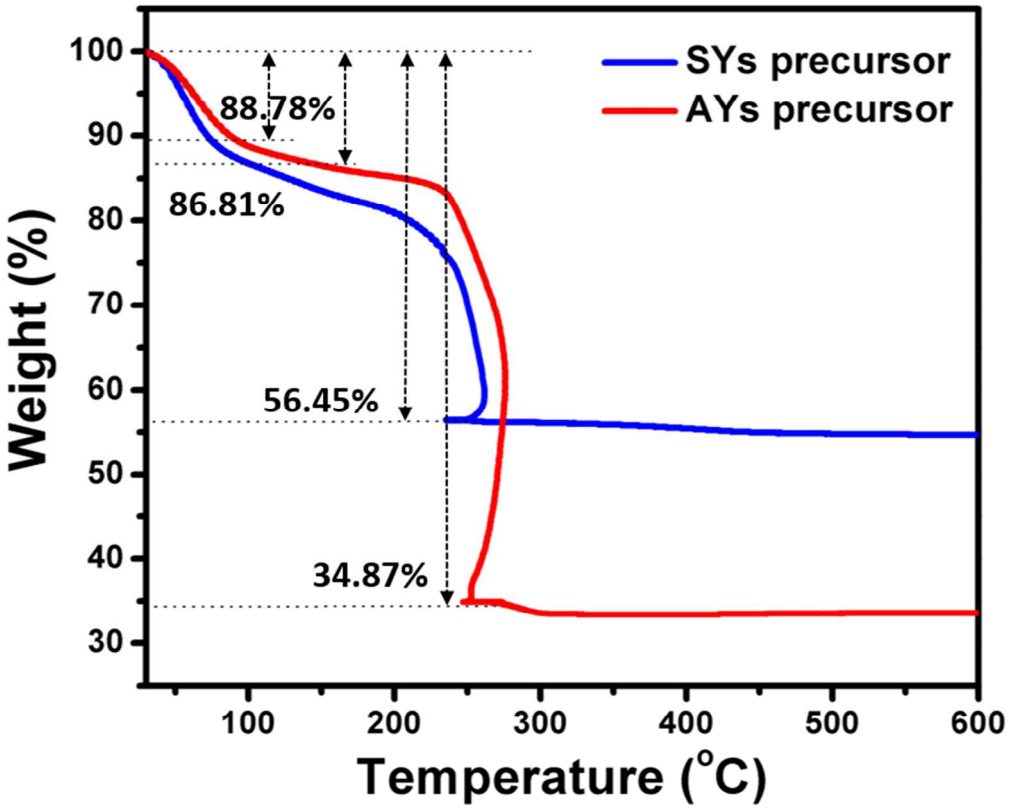

Figure S2. TGA curves of the SYs and AYs Ni-Co precursors.

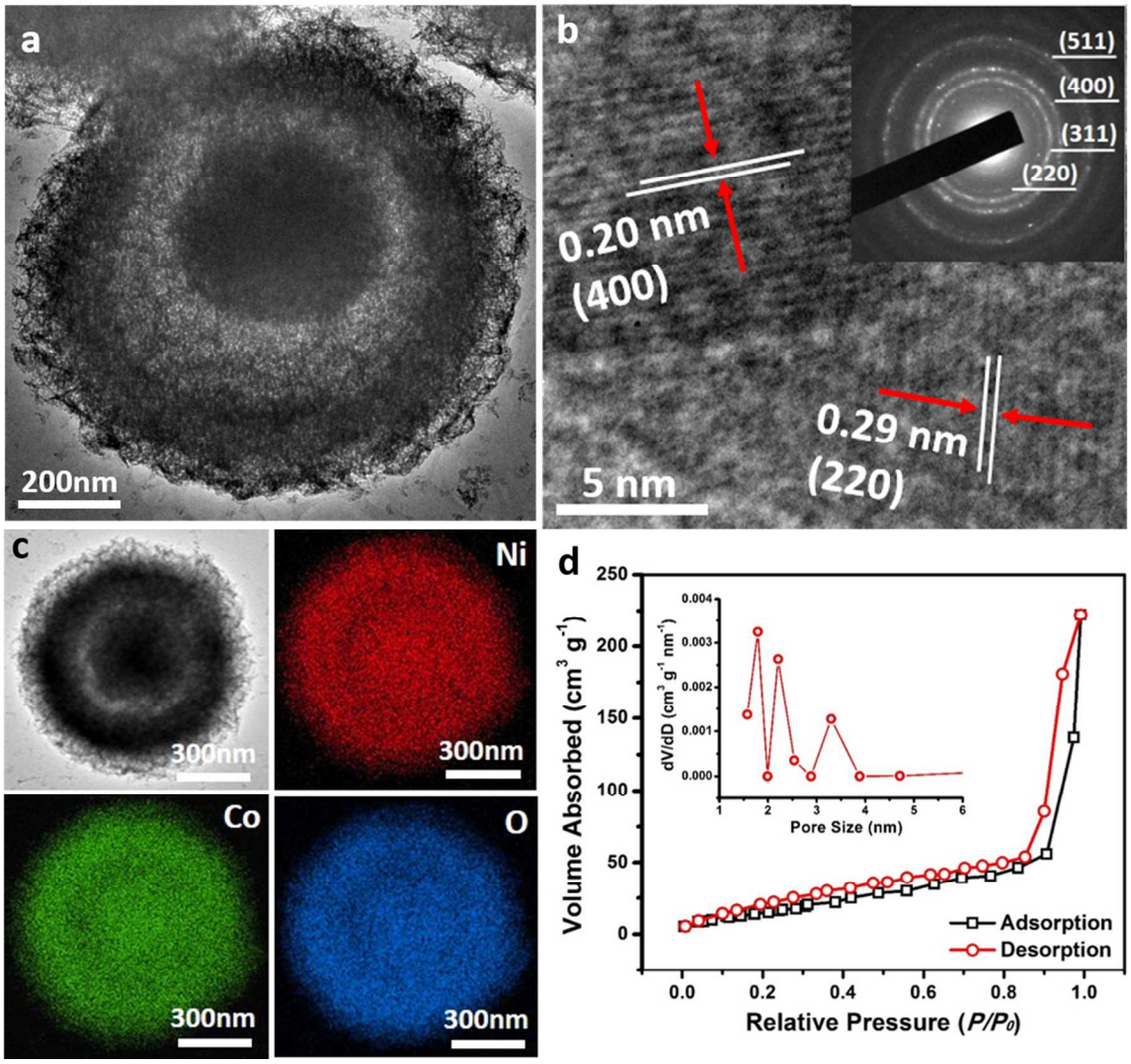

Figure S3. TEM (a), HRTEM (b) with SAED diffraction pattern (inset), TEM elemental mapping (c) and BET isotherm (d) with pore size distribution (inset) of the $NiCo_2O_4$ SYs structure.

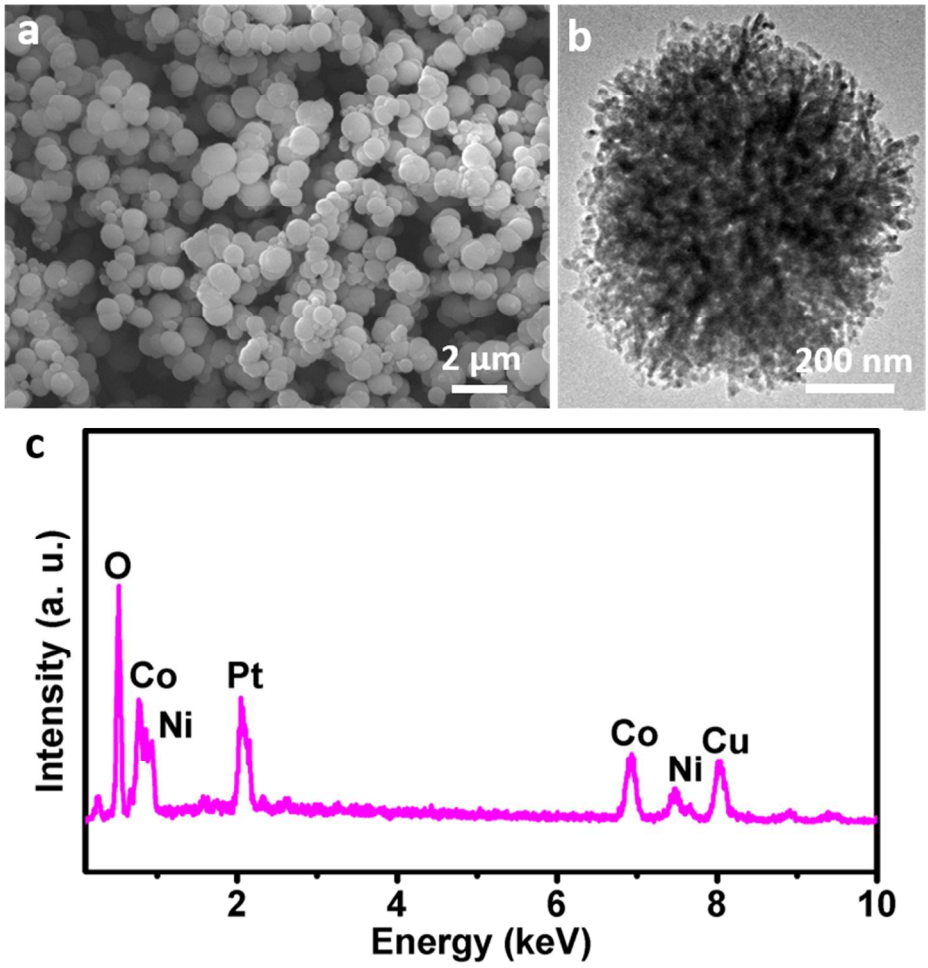

Figure S4. SEM (a), TEM (b) and EDX (c) of $NiCo_2O_4$ nanoparticles (control sample).

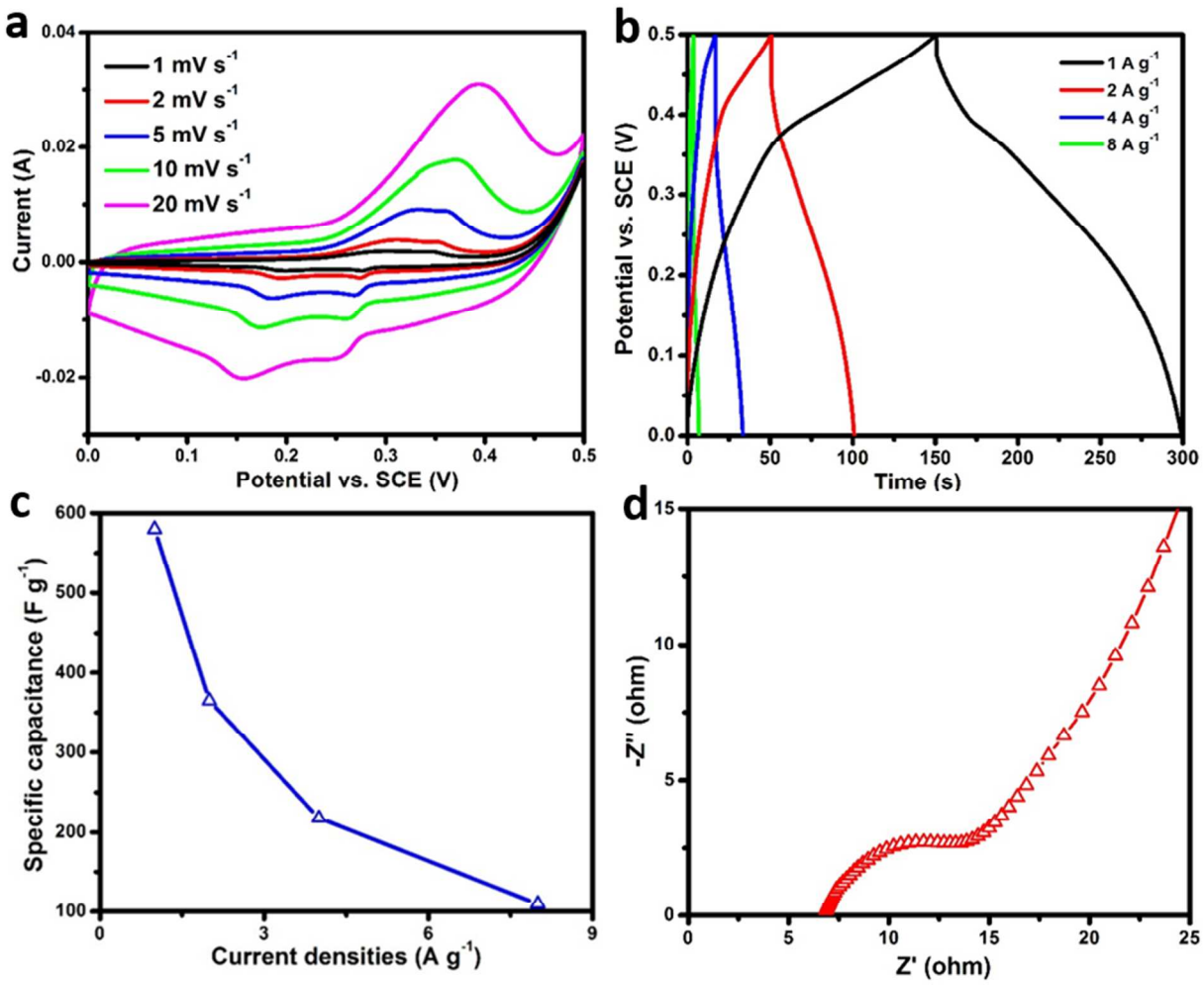

Figure S5. CV (a), GCCD (b), capacitances calculated from CV and EIS (d) curves of $NiCo_2O_4$ nanoparticles (control sample).

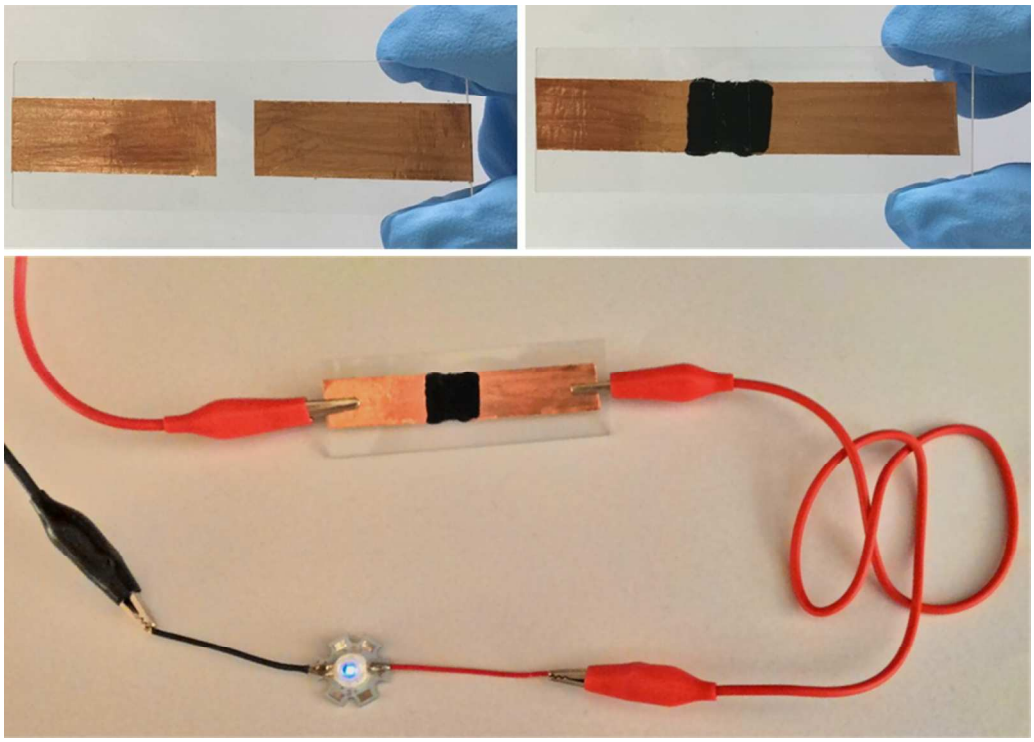

Figure S6. Electrical conductivity demonstration of $NiCo_2O_4$ nanoparticles (control sample).

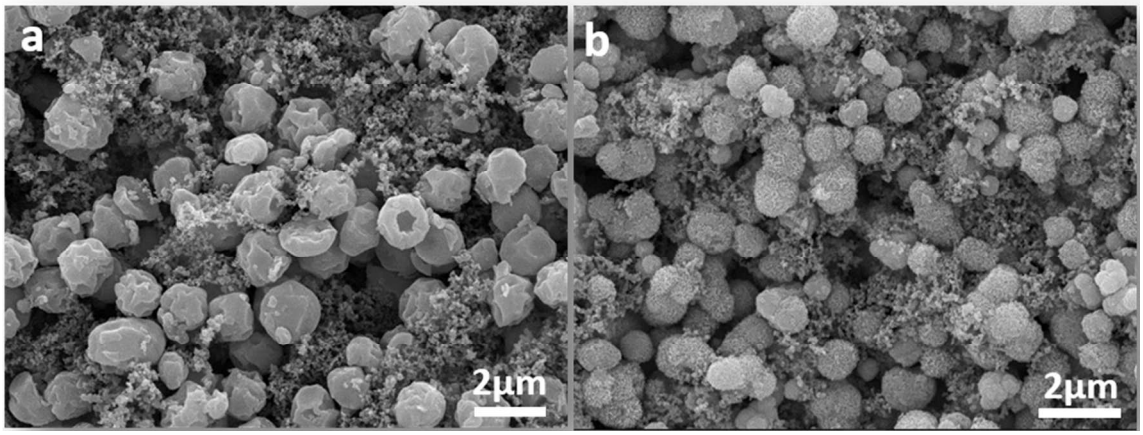

Figure S7. SEM images of the AYs (a) and SYs (b) structures after 5000 cycles as supercapacitor electrodes.

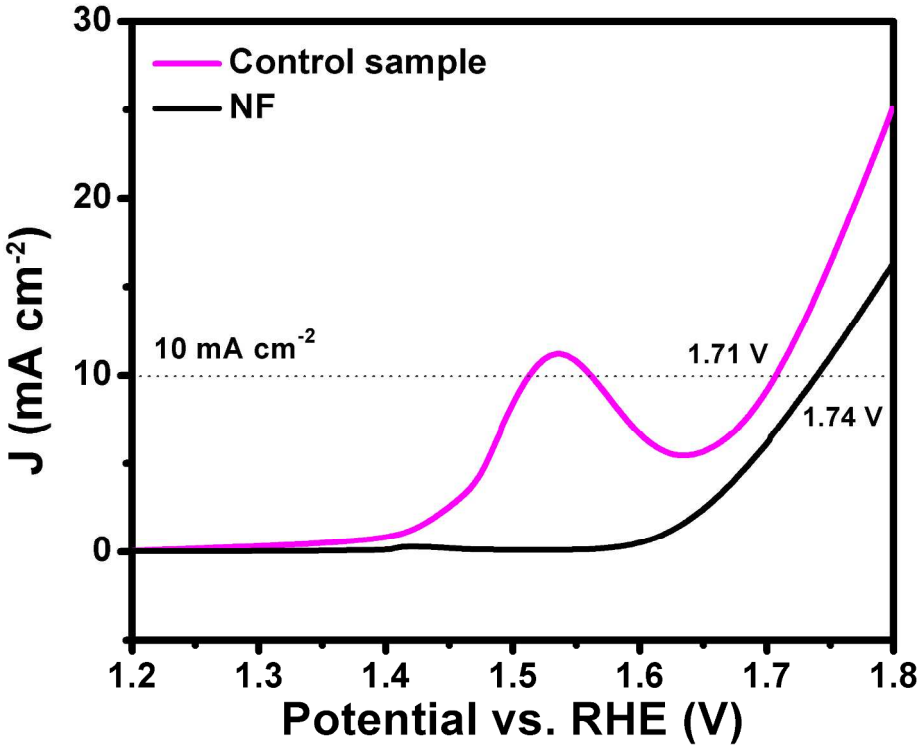

Figure S8. Polarization curves of $NiCo_2O_4$ nanoparticles (control sample) and NF.

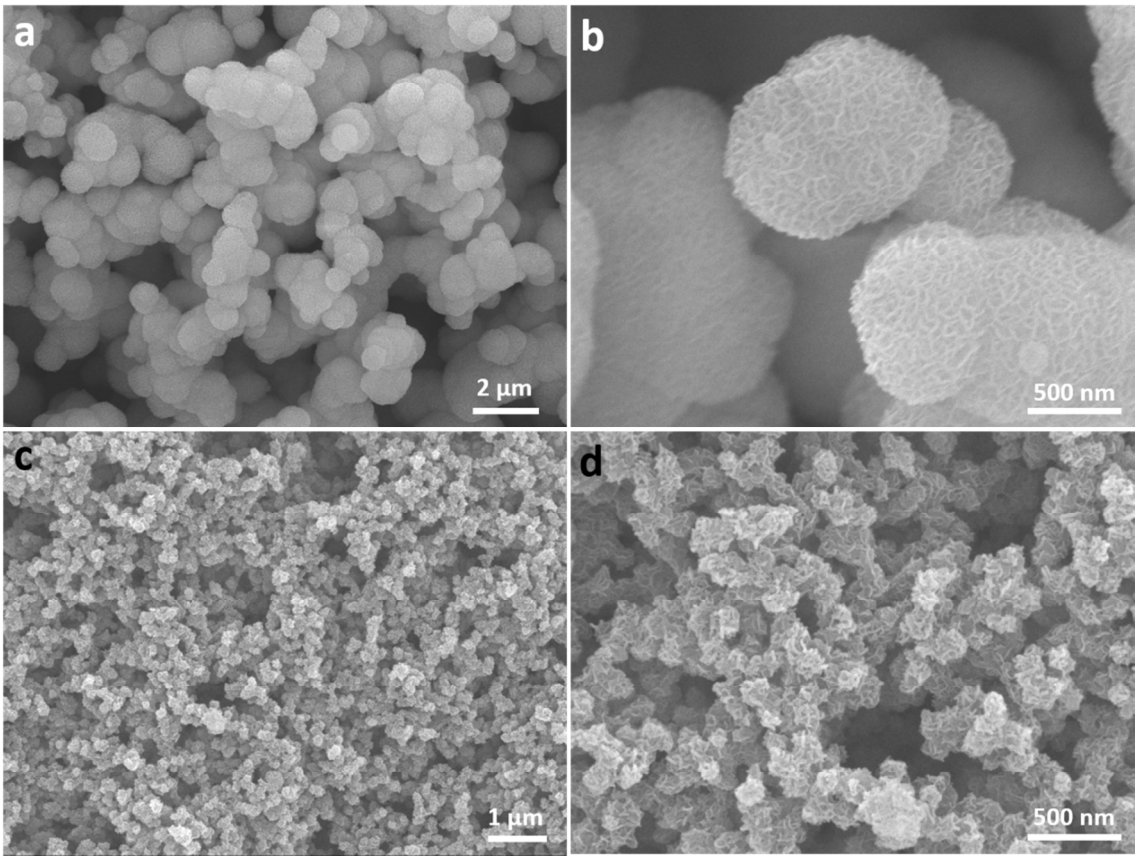

Figure S9. SEM images of the samples prepared by the recipes of SYs (a and b) and AYs (c and d) but using ethanol as solvents instead of IPA.

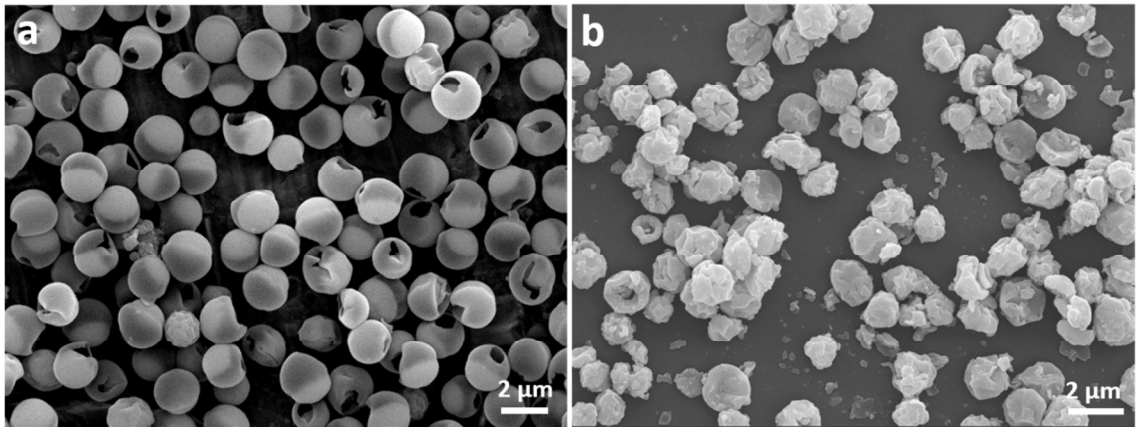

Figure S10. SEM images of the AYs sample annealed at 300 ^{o}C (a) and 500 ^{o}C (b), respectively.

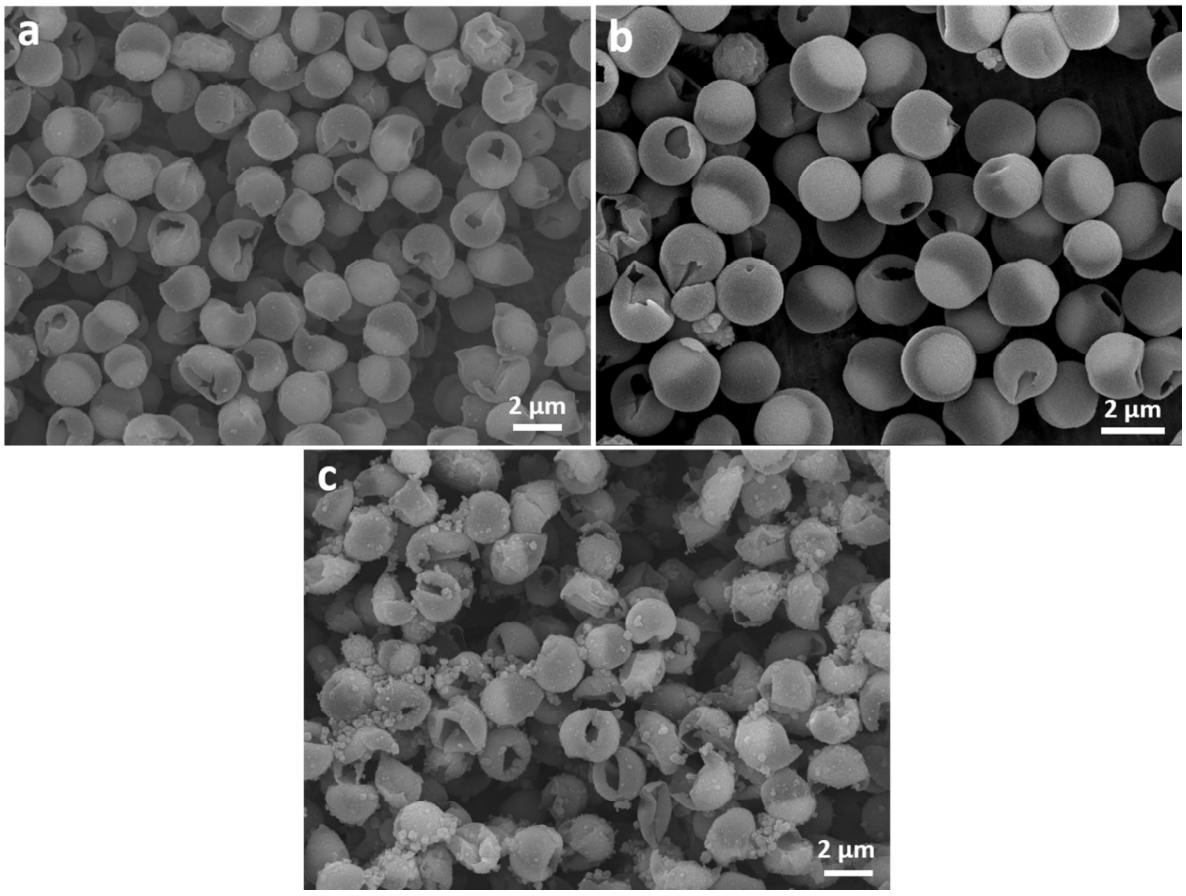

Figure S11. SEM images of the AYs sample precursor collected at 2 h (a), 4 h (b), and 8 h (c), respectively.

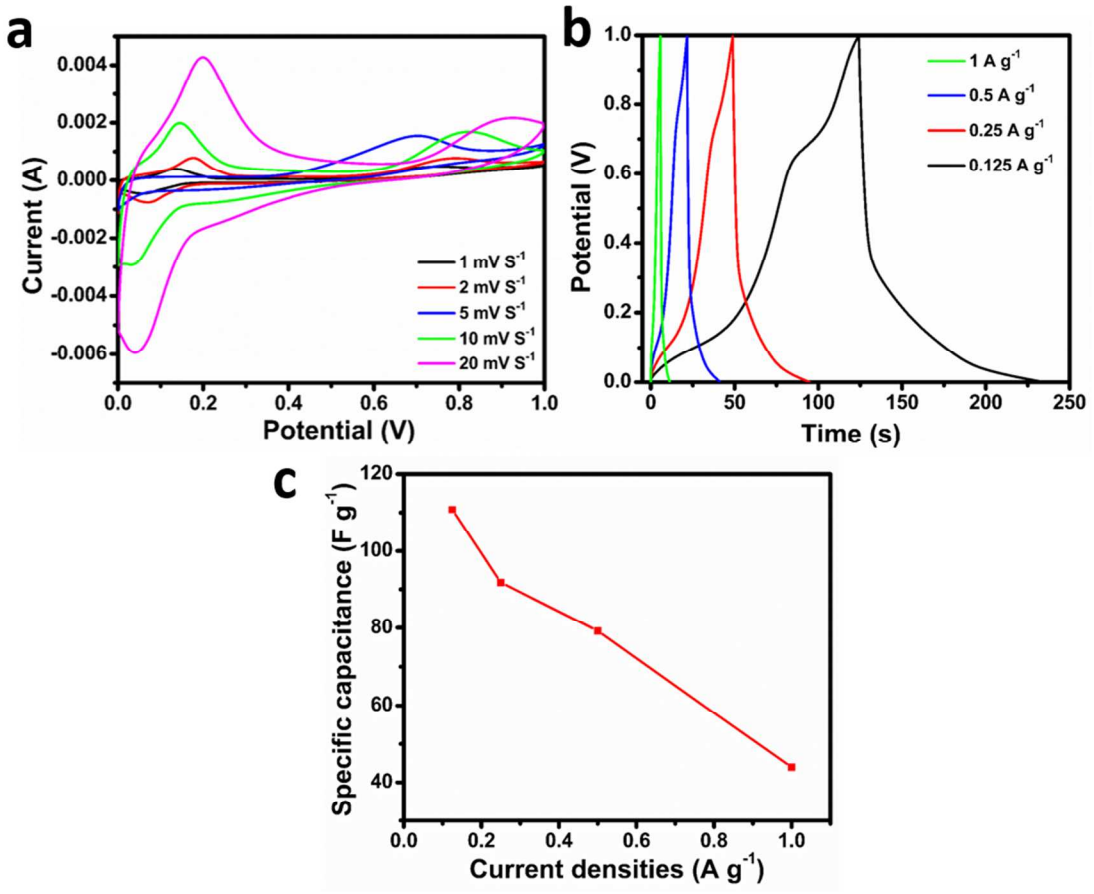

Figure S12. CV (a), GCCD (b) and specific capacitances calculated from GCCD discharge curve (c) of the AYs sample. The electrochemical data are derived from an asymmetrical two-electrode system with 1 M KOH as electrolyte.

Table S1. A comparison of supercapacitor performances of $NiCo_2O_4$ based electrode materials in previous work

$NiCo_2O_4$ Nanostructures	**Current Collector**	**Capacitance F g^{-1}**	**Capacitance Retention**	**Reference**
$NiCo_2O_4$ AYs	Ni foam	1212	97.9% after 5000 cycles	This work
$NiCo_2O_4$ micro flowers	Ni foam	750	102% after 3000 cycles	*ACS Appl. Mater. Interfaces*, 2015, **7**, 17630−17640
$NiCo_2O_4$ nanowires	Ni foam	743	93.8% after 3000 cycles	*Chem. Commun.,* 2012, **48**, 4465–4467
$NiCo_2O_4$ double-shell hollow spheres	Ni foam	568	85.8% after 2000 cycles	*NPG Asia Mater.*, 2015, **7**, e165
$NiCo_2O_4$ nanorods/nanosheets @carbon fibers	Ni foam	1023	91.5% after 2000 cycles	*Sci. Rep.*,2013, **3**, 1470
$NiCo_2O_4$ aerogels	graphite	1400	91% after 2000 cycles	*Adv. Mater.*, 2010, **22**, 347–351
$NiCo_2O_4$ nanosheets	Ti plates	678	100% after 2500 cycles	*J. Mater. Chem. A*, 2014,**2**, 4706-4713
$NiCo_2O_4$ hierarchical spheres	Ni foam	1191	78.3% after 1200 cycles	*CrystEngComm.*, 2014, **16**, 385–392
$NiCo_2O_4$ nanosheets@CNT	Ni foam	1038	100% after 1000 cycles	*J. Mater. Chem. A*, 2014, **2**, 11509–11515

Table S2. A comparison of OER activity of $NiCo_2O_4$ based electrocatalysts in previous work

$NiCo_2O_4$ Catalysts	**Substrate**	**Electrolyte**	**η (mV) at 10 mA cm^{-2}**	**Tafel slope (mV dec^{-1})**	**Reference**
$NiCo_2O_4$ AYs	-	1M KOH	380	159	This work
$NiCo_2O_4$ -Graphene	-	0.1 M KOH	~460	164	*J. Mater. Chem. A*, 2013, **1**, 4754–4762
N-doped $NiCo_2O_4$ - Graphene	-	0.1 M KOH	~450	156	*ACS Nano*, 2013 , **7** , 10190-10196
$NiCo_2O_4$ nanowire array	-	1M KOH	~415	-	*J. Mater. Chem. A*, 2013, **1**, 12170–12177
$NiCo_2O_4$ core-shell nanowire	Carbon cloth	1M NaOH	320	63.1	*Nano Energy*, 2015, **11**, 333−340
$NiCo_2O_4$ – Graphene-MnO_2	Ni foam	0.1 M KOH	-	371	*Chem. Commun.,* 2014, **50**, 207-209
Urchin-like $NiCo_2O_4$	-	1 M NaOH	419	51	*J. Power Sources*, 2014, **268**, 341-348
Au-$NiCo_2O_4$	3D HPG	0.1 M KOH	~560	-	*Sci. Rep.*, 2016, **6**, 23398
$NiCo_2O_4$ nanowire	-	0.1 M KOH	340	63	*Nanoscale*, 2016, **8**, 1390-1400