Supporting Information

A Three-Dimensional Graphene Frameworks-Enabled High-Performance Stretchable Asymmetric Supercapacitor

Ke Li, Yanshan Huang, Jingjing Liu, Mansoor Sarfraz, Phillips O. Agboola, Imran Shakir and Yuxi Xu**

Detailed calculation methods of electrochemical data

In three-electrode system, the specific capacitance of an electrode (C) derived from CV curves or specific capacitance of an electrode (C_s) derived from galvanostatic charge/discharge curves were calculated based on the following equations:

$$C = \frac{\int I \times dV}{2 \times m \times \Delta V \times v} \quad (1)$$

$$C_s = \frac{I \times \Delta t}{m \times \Delta V} \quad (2)$$

where I is the discharge current, m is the mass of one electrode, ΔV is the potential change after a full discharge, v is potential scanning rate and Δt is the discharge time.

The mass ratio of the cathode and anode was obtained based on the following equation:

$$\frac{m_{cathode}}{m_{anode}} = \frac{C_{anode} \times V_{anode}}{C_{cathode} \times V_{cathode}} \quad (3)$$

The gravimetric specific capacitance (C_{wt}) and areal capacitance (C_{area}) of the whole SASC in two-electrode system was obtained by the equations:

$$C_{wt} = \frac{I \times \Delta t}{M \times \Delta V} \quad (4)$$

$$C_{areal} = \frac{I \times \Delta t}{S \times \Delta V} \quad (5)$$

Where M is the total mass of cathode and anode, S is the area of the electrode in device.

Gravimetric energy density (E_{wt}) and areal energy density (E_{areal}), gravimetric power density (P_{wt}) and areal power density (P_{areal}) of the device were calculated from the following equations:

$$E_{wt} = \frac{1}{2} \times C_{wt} \times \Delta V^2 \quad (6)$$

$$P_{wt} = \frac{E_{wt}}{\Delta t} \quad (7)$$

$$E_{areal} = \frac{1}{2} \times C_{areal} \times \Delta V^2 \tag{8}$$

$$P_{areal} = \frac{E_{areal}}{\Delta t} \tag{9}$$

Figures and tables

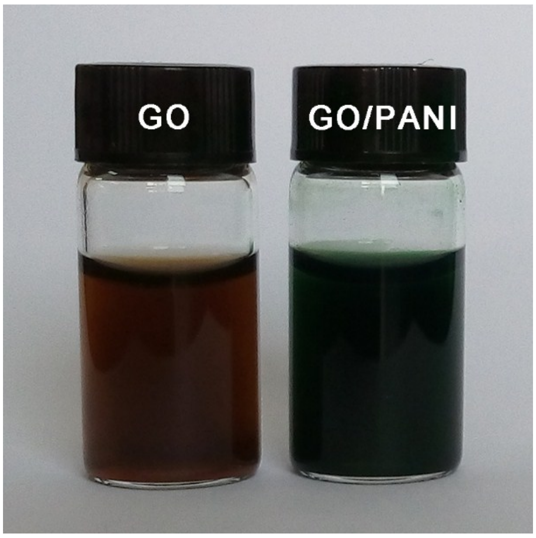

Fig. S1 Photograph of solution processable GO and GO/PANI composite nanosheets

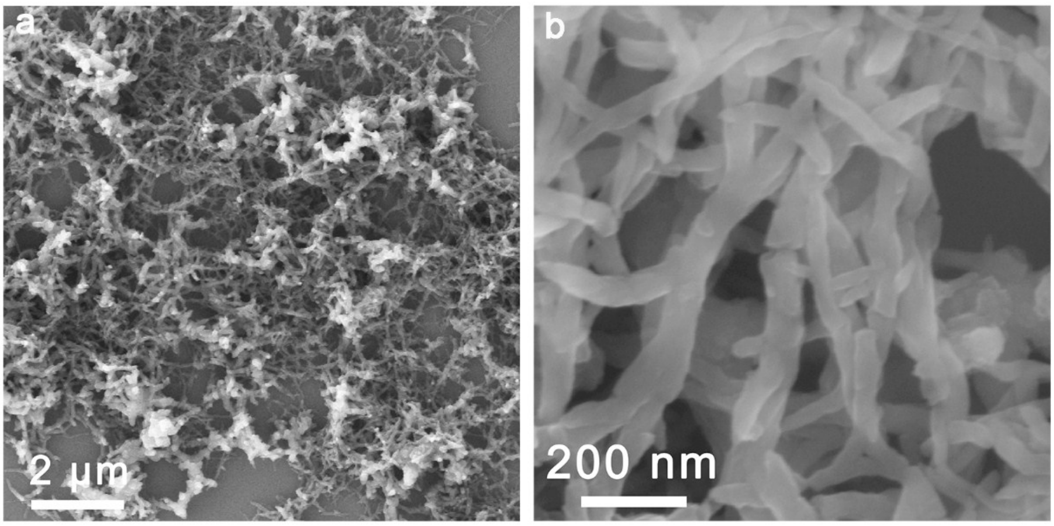

Fig. S2 (a) Low- and (b) high-magnification SEM images of PNFs

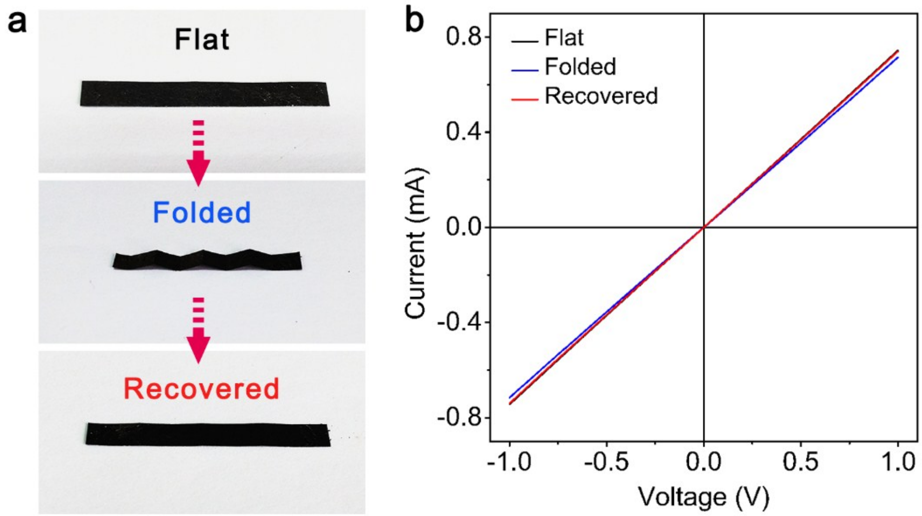

Fig. S3 (a) Photographs and (b) I-V curves of 3D-G/PANI film electrode at flat, folded and recovered states. The electrical conductivity was about 1973 S m^{-1}.

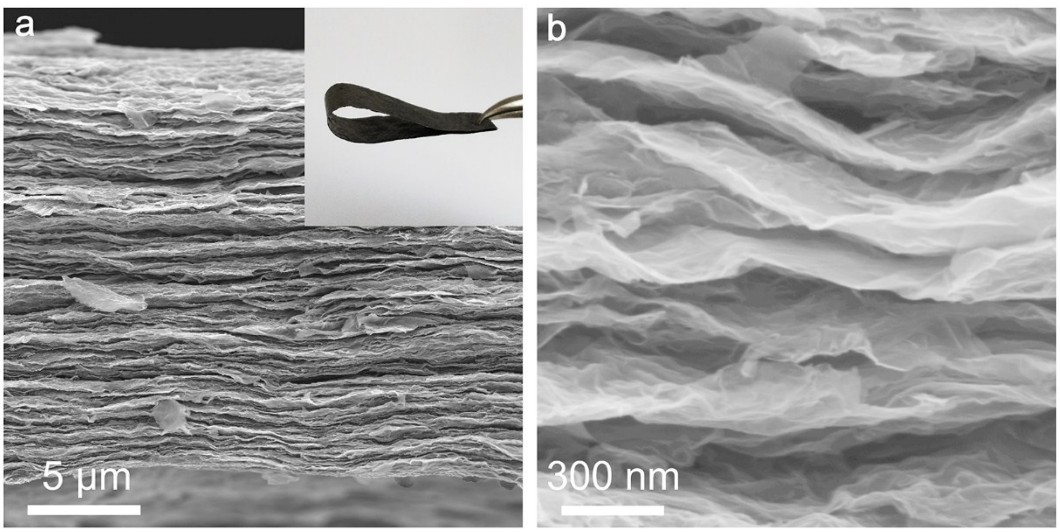

Fig. S4 (a) Low- and (b) high-magnification SEM side view images of 3D-G film

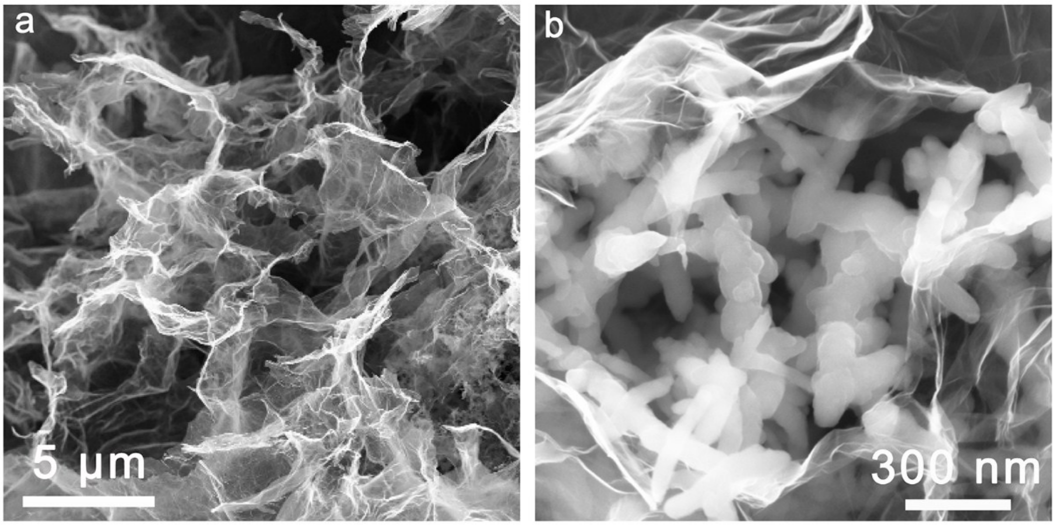

Fig. S5 (a) Low- and (b) high-magnification SEM images of 3D-G/PNF. Due to the aggregation and branched structure, only part of PNFs were in direct contact with graphene in 3D-G/PNF.

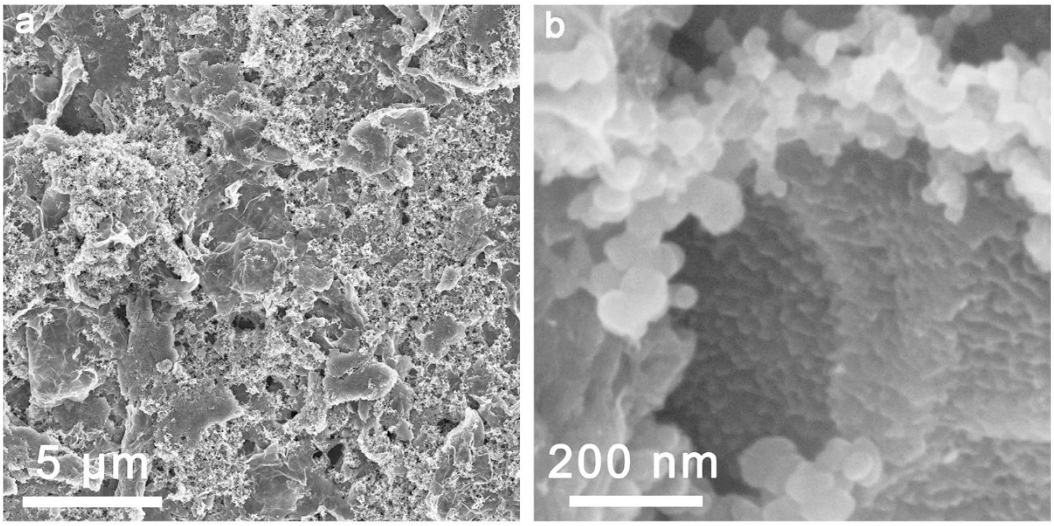

Fig. S6 (a) Low- and (b) high-magnification SEM images of CB/PANI. The CB/PANI showed a compact structure with CB nanoparticles simply attached on the surface of G/PANI composite nanosheets and most PANI exposed

.

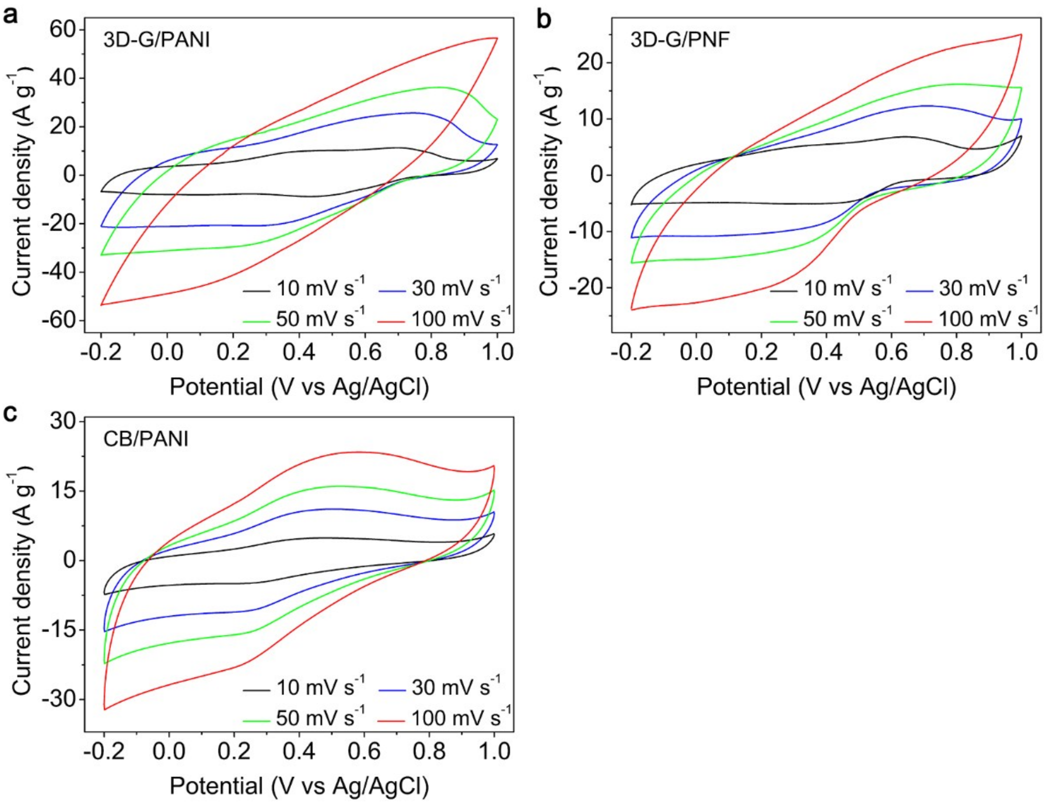

Fig. S7 CV curves of (a) 3D-G/PANI, (b) 3D-G/PNF and (c) CB/PANI electrodes at different scan rates in a three-electrode cell

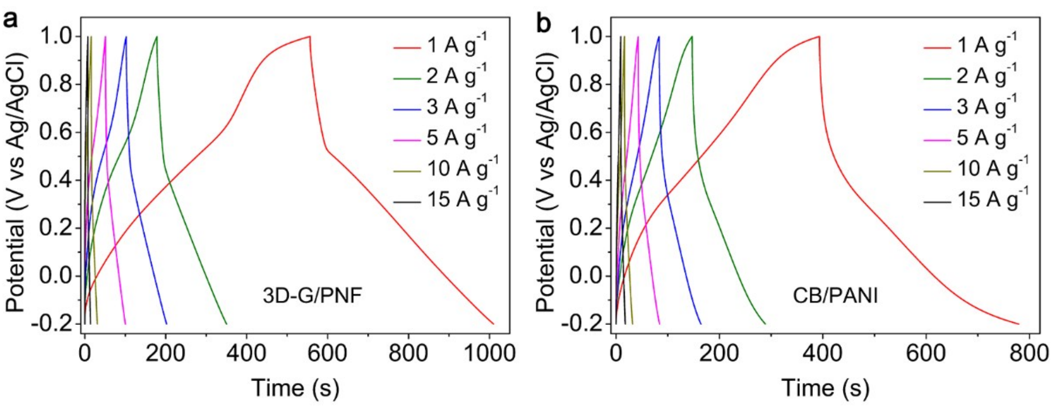

Fig. S8 Galvanostatic charge-discharge curves of (a) 3D-G/PNF and (b) CB/PANI electrodes at different current densities in a three-electrode cell

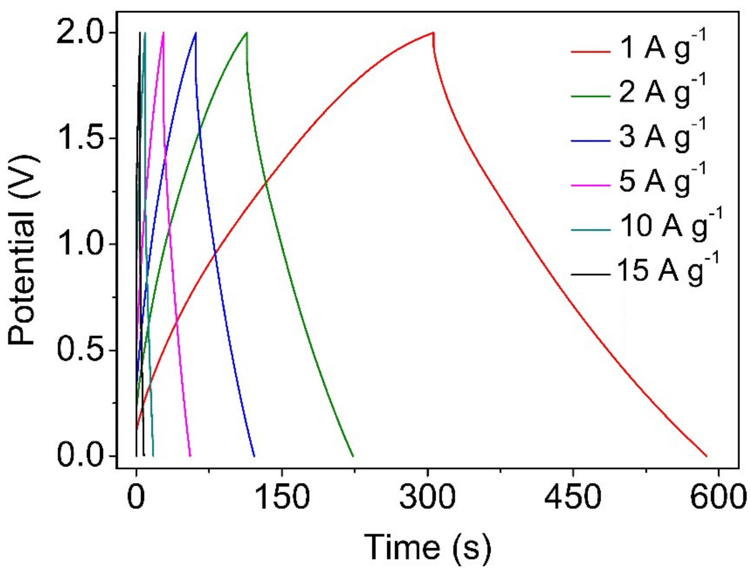

Fig. S9 Galvanostatic charge-discharge curve of 3D-G/PANI//3D-G SASC at different current densities

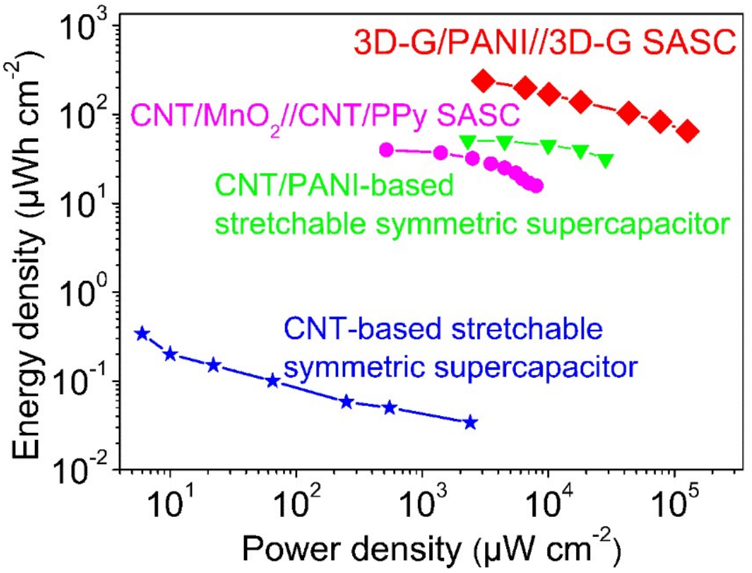

Fig. S10 Regone plots of 3D-G/PANI//3D-G SASC in comprision with CNT/MnO_2//CNT/Ppy SASC,[1] CNT/PANI-based stretchable symmetric supercapacitor,[2] and CNT-based stretchable symmetric supercapacitor[3]

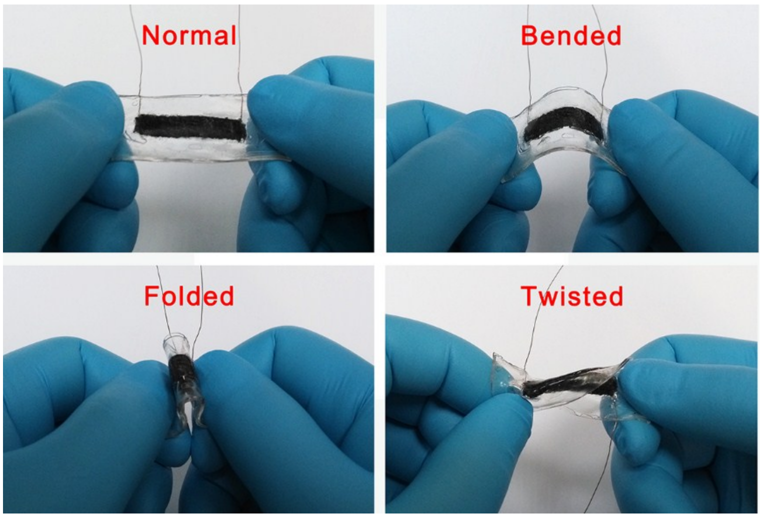

Fig. S11 Digital photographs of the flexible 3D-G/PANI//3D-G SASC at normal, bended, twisted and folded states

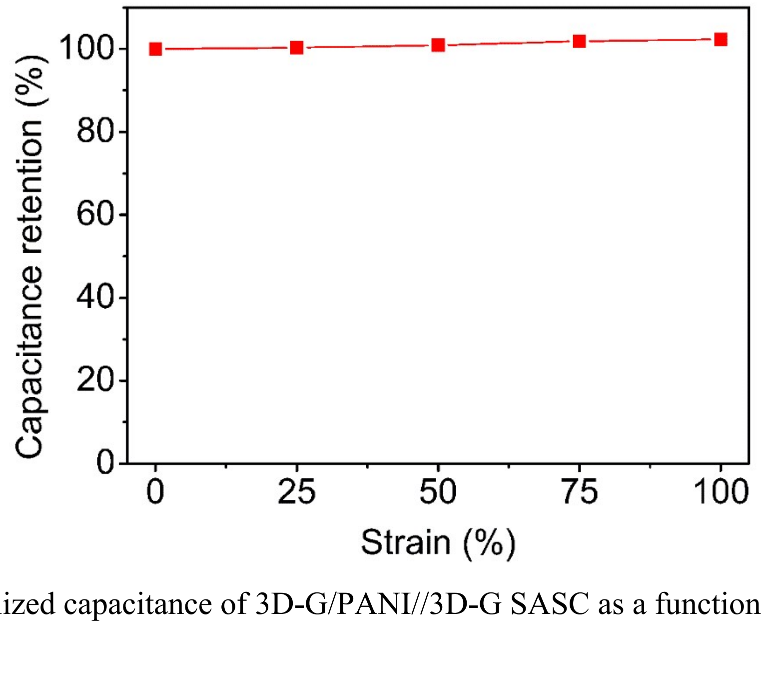

Fig. S12 Normalized capacitance of 3D-G/PANI//3D-G SASC as a function of tensile strains.

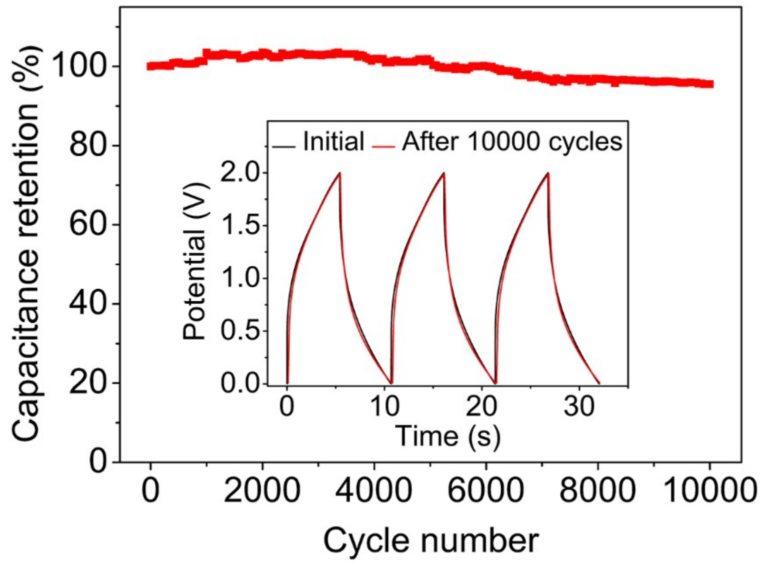

Fig. S13 Cycling stability of 3D-G/PANI//3D-G SASC at 5 A g^{-1}

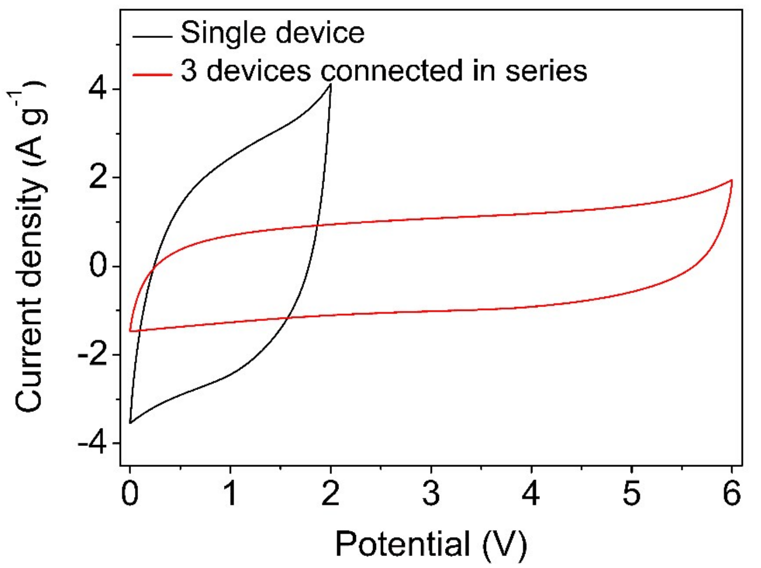

Fig. S14 CV curves of the three 3D-G/PANI//3D-G SASC group and a single supercapacitor at a scan rate of 30 mV s^{-1}

Table S1. Comparison of the 3D-G/PAN//3D-G SASC with previously reported SASCs

Cathode//Anode	Electrolyte	Potential window	Specific capacitance		Maximum energy density		Electrochemical stability	Mechanical durability	Reference
			C_{wt} (F g^{-1})	C_{areal} (mF cm^{-2})	E_{wt} (Wh Kg^{-1})	E_{areal} (μWh cm^{-2})			
3D-G/PANI//3D-G	**PVA/LiCl**	**2.0 V**	**140 (1 A g^{-1})**	**429 (1 A g^{-1})**	**77.8**	**238.5**	**95.6% (10000)**	**91.2% (100)**	**This work**
MnO_2/CNT//CNT	PVA/KOH	1.5 V	NA	33.75 (5 mV s^{-1})	NA	10.26	99% (10000)	100% (20)	ACS Nano, 2015, 9, 6088
Carbon fiber thread/PANI// Carbon fiber thread	PVA/H_3PO_4	1.6 V	NA	NA	NA	NA	81% (10000)	NA	Nano Energy, 2015, 11, 662
MnO_2/CNT// Fe_2O_3/CNT	PVA/ Na_2SO_4	2.0 V	82.4 (0.1 A g^{-1})	NA	45.8	NA	98.9% (10000)	99% (10000)	J. Mater. Chem. A ,2016, 4, 12289
CNT/PPy// CNT/PDAA	FKM/Et_4NBF_4-AN	2.7 V	57.9 (1 mA cm^{-2})	NA	58.2	NA	88% (10000)	NA	J. Ma ter. Chem. A, 2016, 4, 14839
CNT/MnO_2// CNT/PPy	KCl-CH_2=CH-SiO_2/PAAM	2.0 V	72 (2mA cm^{-2})	281.3 (2mA cm^{-2})	40	156	85% (5000)	96% (500)	ACS Appl. Mater. Interfaces. 2015, 7, 15303

Table S2. Comparison of the 3D-G/PAN//3D-G SASC with some previously reported stretchable symmetric supercapacitors

Electrode	Electrolyte	Potential window	Specific capacitance		Maximum energy density		Electrochemical stability	Stretchable durability	Reference
			C_{wt} ($F\ g^{-1}$)	C_{areal} ($mF\ cm^{-2}$)	E_{wt} ($Wh\ Kg^{-1}$)	E_{areal} ($\mu Wh\ cm^{-2}$)			
3D-G/PANI//3D-G	**PVA/LiCl**	**2.0 V**	**140 (1 A g^{-1})**	**429 (1 A g^{-1})**	**77.8**	**238.5**	**95.6% (10000)**	**91.2% (100)**	**This work**
CNT/PANI	PVA/H_2SO_4	0.8 V	NA	573 (10 mV s^{-1})	NA	50.98	NA	97% (20)	ACS Nano, 2016, 10, 5204
CNT/PANI	PVA/H_3PO_4	1.0 V	111.6 (0.5 A g^{-1})	50.1	15.5	7	NA	90% (5000)	Adv. Mater. 2015, 27, 356
CNT/PANI	PVA/H_2SO_4	0.8 V	123.8 (2 mA cm^{-2})	NA	11	NA	80% (2000)	95.6% (500)	Adv. Mater. 2014, 26, 4724
CNT/PANI	PVA/H_3PO_4	1.0 V	308.4	NA	42.8	NA	100% (2000)	100% (200)	Adv. Mater. 2014, 26, 4444
Graphene	PVA/H_3PO_4	0.8 V	7.6 (0.1 V s^{-1})	0.0058 (0.1 V s^{-1})	0.68	0.00052	NA	100% (100)	ACS Nano, 2014, 8, 1039
Graphene	PVA/H_3PO_4	0.8 V	NA	5.85	NA	0.52	92% (2000)	92% (5000)	Adv. Mater. 2015, 27, 5559
Graphene	PVA/H_3PO_4	1.0 V	49 (1 A g^{-1})	NA	6.8	NA	NA	NA	Sci. Rep, 4, 6492.
Graphene	PVA/H_2SO_4	0.8 V	3.475	0.00225	0.31	0.0002	98% (10000)	NA	ACS Nano, 2014, 8, 9437

Table S3. Comparison of the 3D-G/PAN//3D-G SASC with some previously reported non-stretachable asymmetric supercapacitors

Cathode//Anode	Extra electrode components (weight percent)	Electrolyte	Potential window	Specific capacitance		Maximum energy density		Electrochemical stability	Reference
				C_{wt} (F g^{-1})	C_{areal} (mF cm^{-2})	E_{wt} (Wh Kg^{-1})	E_{areal} (μWh cm^{-2})		
3D-G/PAN//3D-G	**None**	**PVA/LiCl**	**2.0 V**	**140 (1 A g^{-1})**	**429 (1 A g^{-1})**	**77.8**	**238.5**	**95.6% (10000)**	**This work**
ACT/Co-Al LDH// ACT/graphene	None	PVA/KOH	1.6 V	145.8 (12.5 mA cm^{-2})	NA	55	NA	87.54% (2000)	Nat. Commun. 2016, 7, 11586
MnO_2//Fe_2O_3/PPy	None	PVA/LiCl	1.6 V	NA	49.6 (1 mA cm^{-2})	NA	17.6	97 (5000)	Angew. Chem. Int. Ed. 2017, 56, 1105
MnO_2/TiN// N-MoO_{3-x}	None	PVA/LiCl	2.0 V	NA	NA	NA	NA	80.3% (5000)	Angew. Chem. Int. Ed. 2016, 55, 6762
MnO_2//Ni nanotube arrays /PPy	None	PVA/LiCl	1.6 V	141.9 (1 A g^{-1})	NA	50.5	NA	81% (10000)	Adv. Mater. 2016, 28, 7680
N-doped-CNTs/Au-doped-MnO_2//Fe_2O_3	None	PVA/LiCl	0.8 V	158 (0.42 mA cm^{-3})	NA	~14	NA	97% (5000)	Nano Lett. 2016, 16, 40
MoS_2-rGO/MWCNT// rGO/MWCNT	None	PVA/H_2SO_4	1.4 V	NA	NA	NA	NA	100% (7000)	Angew. Chem. Int. Ed. 2015, 54, 4651
Conductive paper/MnO_2// Carbon fiber	None	PVA/LiCl	1.8 V	56	NA	25.3	NA	98.9% (10000)	Angew. Chem. Int. Ed. 2015, 54, 6800
$SrCo_{0.9}Nb_{0.1}O_{3-\delta}$//AC	PTFE/Carbon black (15%)	6 M KOH	1.5 V	120 (0.5 A g^{-1})	NA	37.6	NA	98.3% (5000)	Angew. Chem. Int. Ed. 2016, 55, 9576
Ni nanotube arrays /MnO_2//Ni nanotube arrays/PPy	None	0.50 M Na_2SO_4	1.7 V	121.6	135.15 (3 A g^{-1})	48.8	54.2	100% (10000)	Adv. Mater. 2016, 28, 4105
PBOTT-BTD//PEDOT	None	PMMA/PC/ TBAPF6	1.4 V	23 (0.8 A g^{-1})	NA	6.3	NA	NA	Adv. Energy Mater. 2017, 7, 1601623

N-doped carbon/PANI// N-doped carbon	PTFE/ Acetylene black (20%)	1 M Na_2SO_4	1.8 V	134 (1 A g^{-1})	NA	60.3	NA	92% (5000)	Adv. Energy Mater. 2016, 6, 1601111
G/CNT/PANI//G	PTFE/ Acetylene black (15%)	1 M H_2SO_4	1.6 V	116.7 (0.2 A g^{-1})	NA	41.5	NA	91% (5000)	ACS Appl. Mater. Inter. 2013, 5, 8467

1 Q .Tang, M. Chen, C. Yang, W. Wang, H. Bao and G. Wang, *ACS Appl. Mater. Inter.*, 2015, **7**, 15303.

2 J. Yu, W. Lu, S. Pei, K. Gong, L. Wang, L. Meng, Y. Huang, J. P. Smith, K. S. Booksh, Q. Li, J.-H. Byun, Y. Oh, Y. Yan and T.-W. Chou, *Acs Nano*, 2016, **10**, 5204.

3 H. Kim, J. Yoon, G. Lee, S. H. Paik, G. Choi, D. Kim, B. M. Kim, G. Zi, J. S. Ha, *ACS Appl. Mater. Inter.*, 2016, **8**, 16016.