## **Supporting Information**

Mechanically strong multifunctional three-dimensional crosslinked aramid nanofiber/holey graphene and aramid nanofiber/holey graphene/polyaniline hydrogels and derived films

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Figure S1. Illustration of the preparation of ANF dispersion from Kevlar nanofibers.

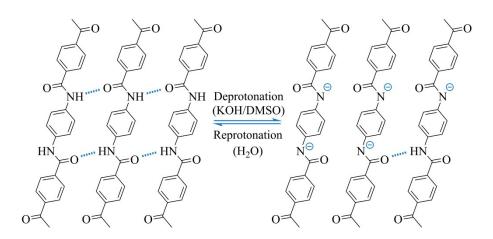
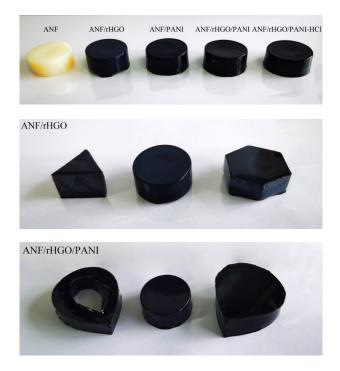


Figure S2. The deprotonation and reprotonation process of PPTA.



**Figure S3**. Illustrations of as-prepared hydrogel samples (upper), ANF/rHGO hydrogels (middle) and ANF/rHGO/PANI hydrogels (bottom) in different shapes casting from different models.

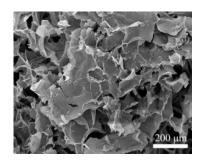


Figure S4. The SEM image of ANF/rHGO/PANI aerogel.

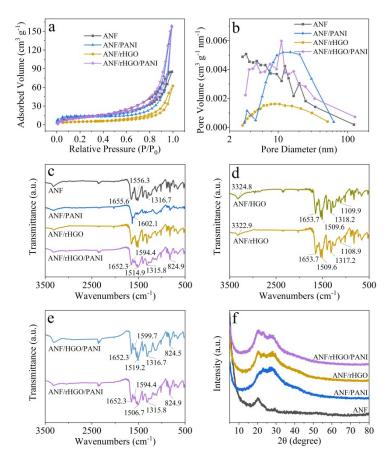


Figure S5. a) Adsorption-desorption isotherm curves, b) the pore size distribution by

the BJH method, c-e) FT-IR spectra, f) XRD patterns.

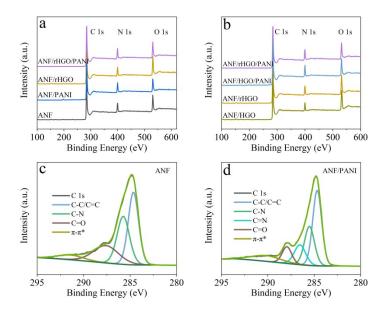
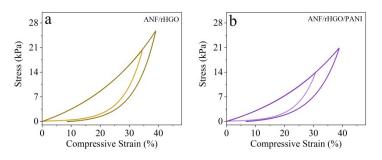


Figure S6. a-b) XPS spectra of as-prepared samples. c-d) C 1s spectra of c) ANF, d)

ANF/PANI.



Figure S7. The compressive process for ANF hydrogel.



**Figure S8**. Compressive stress-strain curves with different strains for ANF/rHGO and ANF/rHGO/PANI hydrogels.



Figure S9. Illustration of ANF aerogel on setaria.

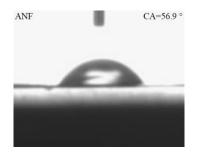
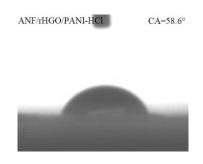


Figure S10. Illustration of static water contact angle on ANF film.



**Figure S11**. Illustration of static water contact angle on ANF/rHGO/PANI film after treating with 1 M HCl.

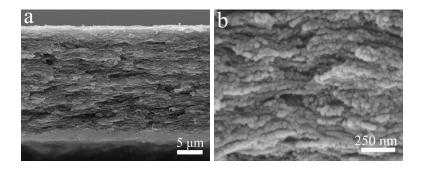


Figure S12. The SEM images of the rHGO/PANI film.

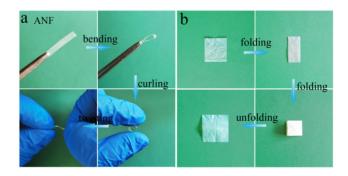
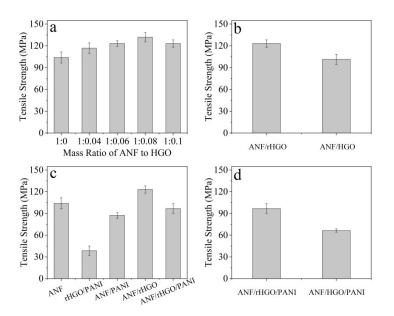
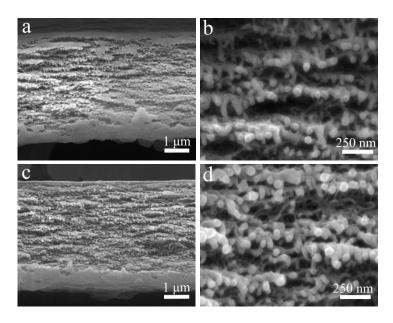


Figure S13. Illustrations of various mechanical states (initial, bending, folding, curling and twisting) of flexible ANF films.



**Figure S14**. Averaged breaking tensile strength of (a) ANF/rHGO films with different mass ratios of ANF to HGO, (b) comparison of ANF/rHGO and ANF/HGO films, (c) as-prepared flexible films and (d) comparison of ANF/rHGO/PANI and ANF/HGO/PANI film samples.



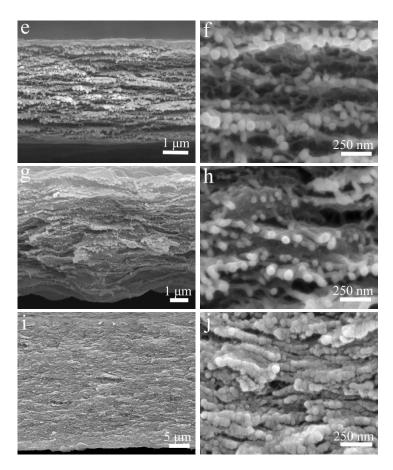
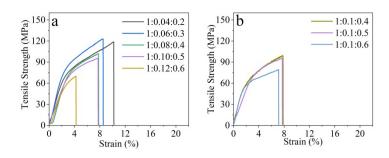
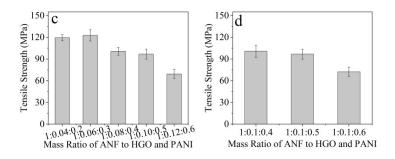
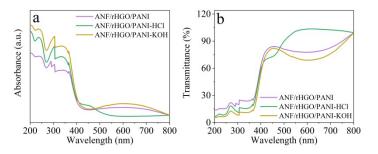


Figure S15. The SEM images of the fracture cross-section of a-b) ANF, c-d) ANF/PANI, e-f) ANF/rHGO, g-h) ANF/rHGO/PANI, i-j) rHGO/PANI films after tensile test.





**Figure S16**. (a-b) Tensile strength test results and (c-d) averaged breaking tensile strength of ANF/rHGO/PANI composite samples with (a, c) different mass ratios (1:0.04:0.2, 1:0.06:0.3, 1:0.08:0.4, 1:0.10:0.5 and 1:0.12:0.6) of ANF, HGO and PANI and (b, d) different PANI contents (1:0.1:0.4, 1:0.1:0.5 and 1:0.1:0.6).



**Figure S17**. The UV-vis spectra of ANF/rHGO/PANI film post-treated by acidic or alkaline solution.

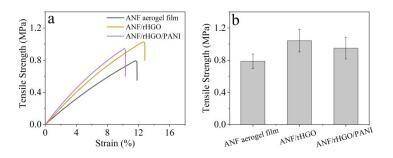


Figure S18. (a) The stress-strain curve and (b) averaged breaking tensile strength of

## ANF, ANF/rHGO and ANF/rHGO/PANI aerogel film.

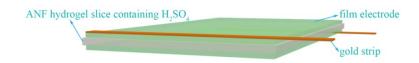


Figure S19. Schematic illustration for solid-state supercapacitor assembled using  $ANF/H_2SO_4$  hydrogel slice separating as-prepared film electrodes.

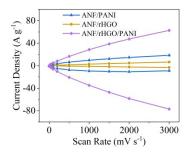


Figure S20. The plots of current density versus scan rate from CV curves.

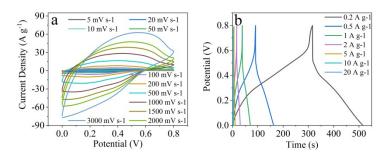
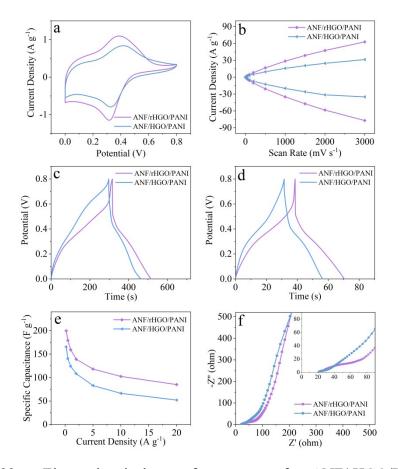
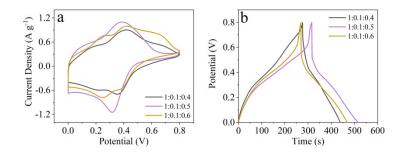


Figure S21. a) CV curves at various scan rates and b) GCD curves at various current

densities for ANF/rHGO/PANI supercapacitor.



**Figure S22**. Electrochemical performance of ANF/rHGO/PANI and ANF/HGO/PANI supercapacitors: a) CV curves at 10 mV s<sup>-1</sup>, b) The plot of current density versus scan rate from CV curves, c) GCD curves at 0.2 A g<sup>-1</sup>, d) GCD curves at 1 A g<sup>-1</sup>, e) Specific capacitance against current density, and f) EIS plot.



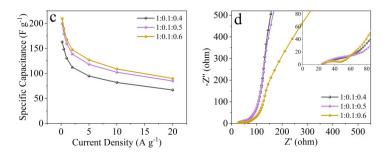
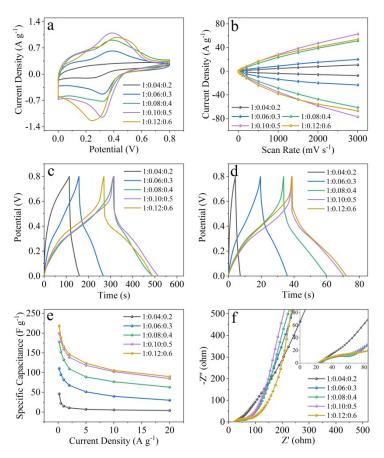
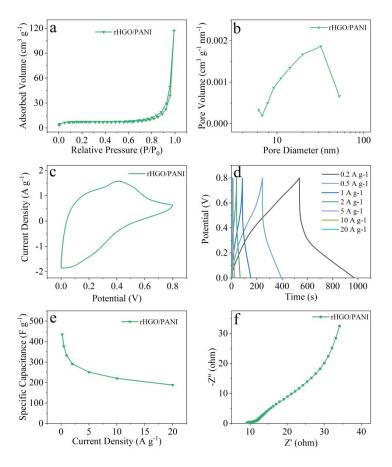


Figure S23. Electrochemical performance of ANF/rHGO/PANI composite supercapacitors with different PANI contents in composites: a) CV curves at 10 mV s<sup>-1</sup>, b) GCD curves at 0.2 A g<sup>-1</sup>, c) Specific capacitance against various current densities, and d) EIS plots.



**Figure S24**. Electrochemical performance of ANF/rHGO/PANI composite supercapacitors with different mass ratios of ANF, HGO and PANI: a) CV curves at 10 mV s<sup>-1</sup>, b) The plots of current density versus scan rate from CV curves, c) GCD curves at 0.2 A g<sup>-1</sup>, d) GCD curves at 1 A g<sup>-1</sup>, e) Specific capacitance against various

current densities, and f) EIS plots.



**Figure S25**. a) Adsorption-desorption isotherm curves, and b) the pore size distribution by the BJH method. c-f) The electrochemical performance of rHGO/PANI supercapacitor: c) CV curve at 10 mV s<sup>-1</sup>, d) GCD curves at different current densities, e) Specific capacitance against current density, and f) EIS plot.

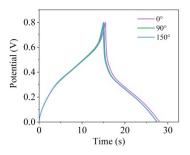
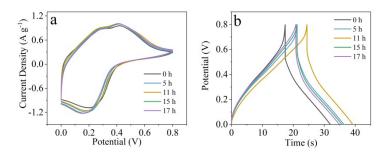


Figure S26. GCD curves at 2 A  $g^{-1}$  under different bending angles (0, 90 and 150 °)

for ANF/rHGO/PANI supercapacitors.



**Figure S27**. a) CV curves at 10 mV s<sup>-1</sup> and b) GCD curves at 2 A g<sup>-1</sup> for different times under keeping tensile force of 25 g for ANF/rHGO/PANI supercapacitors.

Assignment	Waven	umbers (cm <sup>-1</sup> )	
	ANF	ANF/rHGO	ANF/rHGO/PANI
C=O stretching vibrations	1655.6	1653.7	1652.3
C=C stretching vibrations of quinoid and benzenoid rings in PANI			1594.4, 1514.9
N-H deformation vibrations	1556.3	1542.8	1543.3
C-N stretching of aromatic amine	1316.7	1317.2	1315.8
C-C ring stretching vibrations	1268.9	1265.1	1268.9
C-O-C stretching vibrations		1108.9	1109.4
C-O stretching vibrations	1017.8	1017.3	1017.3
In-plane and out-of-plane bending vibrations of C-H	825.4	823.0	824.9

**Table S1.** The data of FT-IR spectra for the functional groups of as-prepared samples.

Samples	C (at.%)	N (at.%)	O (at.%)
ANF	79.7	9.3	11.0
ANF/PANI	80.7	10.0	9.3
ANF/HGO	79.6	6.6	13.8
ANF/rHGO	80.0	8.5	11.5
ANF/HGO/PANI	80.5	9.5	10.0
ANF/rHGO/PANI	81.0	9.6	9.4

 Table S2 Elemental Composition of XPS analysis for as-prepared samples.

Generalize	C 1s					
Samples	C-C/C=C	C-N	C-0	C=N	С=О	π-π*
ANF	39.4	31.3			22.9	6.4
ANF/PANI	41.9	24.1		13.9	9.7	10.4
ANF/HGO	43.1	20.5	18.6		11.5	6.3
ANF/rHGO	44.2	21.5	11.8		12.6	9.9
ANF/HGO/PANI	46.9	19.9	7.7	8.2	8.1	9.2
ANF/rHGO/PANI	47.8	20.7	5.8	6.9	8.9	9.9

Table S3 C 1s spectra of C species in as-prepared samples, relative ratio (at.%)

 Table S4 The tensile strength results of ANF based materials in literatures.

Sample	Tensile strength	Methods	Ref.
	(MPa)		
ANF,	125±8 at 10.2±1.6%	Py in-situ polymerization in ANF	S1
22% PPy/ANFs film	80±5 at 13.6±1.9%	and filtration	
ANF, 20%	93.5 at 21.3%	BNNS@PDA into ANF with a	S2
BNNS@PDA/ANF film	48 at 15%	homogenizer and filtration	
1% ANF, 20wt%-CNT FC-	1.8 at 10%	ANF/CNT dispersion coated on Al	S3
ANF/CNT aerogel films	3.4 at 8%	foil to form hydrogel	
ANF, RGO/1wt%ANF films	~95 at 2%,	GO/DMSO, ANF/DMSO mixed	S4
	41.7±2.0 at 1.2%	and filtration; 40 mg, 10-20 $\mu m$	
0.4 wt% ANF aerogel fiber	0.55 at 22%	~300 µm	S5
0.7 wt% ANFGS/PMMA	63.2 at 4.1%	ANFGS:GO/DMSO, ANF/DMSO	S6
film		mixed and centrifugation	
(ANFGS:ANFS/GO=2:1)		ANFGS/PMMA: solution casting	
		method	
8 wt% ANF	$855.7\pm18.9$	Wet Spinning: 8 wt% ANF	S7
ANF/CNT (1:20)	$818.4\pm16.2$ at 2%	dispersion and 0.6 wt% CNT	
		aqueous solutions	
0.1 wt% BANF	$138\pm5.3$ at 9%	aniline was polymerized in the	S8

73 wt% BANF/SWCNTs/15 $40 \pm 4$ at 1.8% wt% PAN1 filmpresence of BANFs and single- walled carbon nanotubesKNFs ~6.60 wt%72 at 19%KNFS hydrothermal treatment, spectra by stirring and membraneS9PDMS/PVDF@KNFs90 at 20% evenly dispersed by stirring and ineeduction stepssigersed by stirring and methraneS1SEBS/Kevlar/Ag/PVA fiber35 at 220% at 2.5 at 2.2%repeating adsorption and in situ still adsorption and in situS100.015% ANF147.3 ± 7.1 at 3.8%PEDOT:PSS into ANF/DMSO with a homogenizer and filtrationS1120%ANFs/80%PEDOT:PSS76.4 ± 2.5 at 2.2% a homogenizer and filtrationa homogenizer, adding curing agent for molding2% KNA1.27 at 12.9%KNA/PEG is KNA films were immersed in melting PCM and socied to room temperatureS13 cooled to room temperature2 wt% ANF196 at 16.1%Spin coating to deposit ANF film, S14 34.7 vol% Au-ANF film159 at 11%Spin coating to deposit ANF film, S144.1 vol% ANF155 at 13%ANF hydrogel: vacuum filtration to add PANIS150.5 mg/mI. ANF papers23.5 gat 24.8%ANF hydrogel: vacuum filtrateS160.5 mg/mI. ANF papers23.5 gat 24.8%ANF hydrogel: vacuum filtrate to deposit ANF film, S14S160.5 mg/mI. ANF papers2.0 MPa m² kg 1 at 2.7%ANF-MXenc/AgNW: vacuum filtrate to deposite MXenc/AgNWS173.8.1% rGO hydrogel-Kevlar2.7%ANF-MXenc/AgNW: vacuum filtrate to deposite MXenc/AgNWS183.9.1% rGO hydrogel-Kevlar2.0.92 at 1.96%ANF-MXenc/AgN				
KNFs $72$ at 19%KNFShydrothermaltreatment, $59$ PDMS/PVDF@KNFs90 at 20%evenly dispersed by stirring and mixed with PVDF and PDMSSEBS/Kevlar/Ag/PVA fiber $35$ at 220%repeating adsorption and in situ $S10$ reduction steps0.015% ANF $147.3 \pm 7.1$ at 3.8%PEDOT:PSS into ANF/DMSO with $S11$ a homogenizer and filtration $20%ANF*/80%PEDOT:PSS76.4 \pm 2.5 at 2.2%a homogenizer and filtrationfilm11\% ANF/IP plastic33.3 \pm 2.4 at 4%ANF into EP with a homogenizer,adding curing agent for molding2\% KNA1.27 at 12.9%KNA aerogel films: freez-dryingKNA/PEG aerogel film2.1 at 20%KNA/PEG: KNA films wereimmersed in melting PCM andsocied to room temperature2 wt% ANF196 at 16.1%Spin coating to deposit ANF film,96 at 9.1%spin coating to deposit ANF film,socied to room temperature2 wt% ANF196 at 16.1%Spin coating to deposit ANF film,91 at 7.5\%and vacuum filtration to add 4u NPs1.5 wt% ANF185 at 11%Spin coating to deposit ANF film,10 at 0.75\%ANF hydrogel: vacuum filtrate20 wt% PANI/ANF film179 at 7.5\%and vacuum filtrateS160.5 mg/NU20 wt% MXene/AgNW2.0 MPa m^3 kg^{-1} at2.7\%ANF-MXene/AgNW: vacuumfiltrate to deposite MXene/AgNWKevlar fibers2.0 MPa m^3 kg^{-1} at2.7\%ANFs-DMSO dispersion wasS180.1\% ANF with 30.292 at 1.96\%ANFW (water) Membranes20.392 at 1.96\%ANFs-DMSO dispersion wasS180.1\% ANF aerogel (axia$	73 wt% BANF/SWCNTs/15	$40\pm4$ at 1.8%	presence of BANFs and single-	
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$ \begin{array}{llllllllllllllllllllllllllllllllllll$	0.5 mg/mL ANF papers	235.9 at 24.8%	ANF hydrogel: vacuum filtrate	S16
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$\begin{array}{c} \mbox{followed by a desired amount of DI} \\ \mbox{water or ethanol to substitute} \\ \mbox{DMSO and precipitate the ANFs} \end{array}$	ANFW (water) Membranes	203.92 at 1.96%	ANFs-DMSO dispersion was	S18
water or ethanol to substitute DMSO and precipitate the ANFs 0.1% ANF aerogel (axial $0.095 \pm 0.003$ at 70% ANF dispersion: CaCl <sub>2</sub> , DME, PPD S19 direction) $0.165 \pm 0.005$ at 70% and TPC were added in NMP in ANF aerogel (radial (compression) turn, and diluted by NMP direction) ANF aerogel: filtration and	ANFE (ethanol) Membranes	141.34 at 3.28%	blended with a given ration	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			followed by a desired amount of DI	
$0.1\%$ ANF aerogel (axial $0.095 \pm 0.003$ at 70%ANF dispersion: CaCl2, DME, PPDS19direction) $0.165 \pm 0.005$ at 70%and TPC were added in NMP inANFaerogel (radial (compression))turn, and diluted by NMPdirection)ANF aerogel: filtration and			water or ethanol to substitute	
direction) $0.165 \pm 0.005$ at 70%and TPC were added in NMP inANF aerogel(radial (compression)turn, and diluted by NMPdirection)ANF aerogel: filtration and			DMSO and precipitate the ANFs	
ANF aerogel (radial (compression)turn, and diluted by NMPdirection)ANF aerogel: filtration and	0.1% ANF aerogel (axial	$0.095\pm0.003$ at 70%	ANF dispersion: CaCl <sub>2</sub> , DME, PPD	S19
direction) ANF aerogel: filtration and	direction)	$0.165 \pm 0.005$ at 70%	and TPC were added in NMP in	
	ANF aerogel (radial	(compression)	turn, and diluted by NMP	
filtration	direction)		ANF aerogel: filtration and	
			filtration	

0.2% ANF film	$255.1\pm7.1$ at 18%	ANF dispersion vacuum filtrate and	S20
4.6 wt% PANI@ANF film	$233.3\pm6.1$ at 17.5%	dry, and ANF film immerse in the	
		PANI dispersion	
PMF film (5.7 wt% Au/ANF)	14.42 at 5.25%	PA was added into the ANF	S21
		solution, and Au NP solution was	
		added, then filtration	
MXene/11% ANF membrane	101 at 4.6%	MXene (DMSO), ANF (DMSO)	S22
(ANF 1%)		with a homogenizer and filtration	
РРН	5.3 MPa at 250%	Polymerization of Ani in PVA and	S23
		ABA	
1.0% ANF	105.4 at 8.72%	ANF, HGO and PANI mixed and	This
ANF/rHGO	121.4 at 9.0%	adding water, hydrothermal	work
ANF/rHGO/PANI film	95.5 at 7.7%		

## Table S5 The comparison of electrochemical performance of ANF/rHGO/PANI

Samples	C <sub>s</sub>	Test system	Ref.
RGO/1wt% ANF	171 F g <sup>-1</sup> , 0.5 A g <sup>-1</sup>	Two-electrode	S4
	155 F g <sup>-1</sup> , 10 mV s <sup>-1</sup>		
25wt%TRGO/ANF	116 μF cm <sup>-2</sup> , 10 mV s <sup>-1</sup>	Three-electrode	S24
rGO(ANF <sub>2wt%</sub> ) hydrogel	$\sim 158 \text{ F g}^{-1}, 5 \text{ mV s}^{-1}$	Two-electrode	S25
ANF Sheath on Dry-Spun CNT Wires	0.75 mF cm <sup>-1</sup> , 3.25 μA cm <sup>-1</sup>	Solid state supercapacitor	S7
38.1% rGO hydrogel-Kevlar cloth	~50 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	Solid state supercapacitor	S17
20 wt%ANFs/PEDOT:PSS	111.5 F g <sup>-1</sup> , 0.5 mA cm <sup>-3</sup>	Three-electrode	S11
	12.8 F g <sup>-1</sup> , 0.1 A cm <sup>-3</sup>	Solid state supercapacitor	
PANI/BANF/12wt% CNT	~210 F g <sup>-1</sup> , 0.5 A g <sup>-1</sup>	Two-electrode	S8
	~180 F g <sup>-1</sup> , 10 mV s <sup>-1</sup>		

supercapacitor with reported results in literature.

4.6wt% PANI@ANF film	441.0 F g <sup>-1</sup> , 1 A g <sup>-1</sup>	Three-electrode	S20
	138 F g <sup>-1</sup> , 0.5 A g <sup>-1</sup>	Solid state supercapacitor	
62.5wt%ANF/rHGO/PANI	200 F g <sup>-1</sup> , 0.2 A g <sup>-1</sup>	Solid state supercapacitor	This work

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