

## Supporting Information

### Free-Standing Composite Hydrogel Film for Superior Volumetric Capacitance

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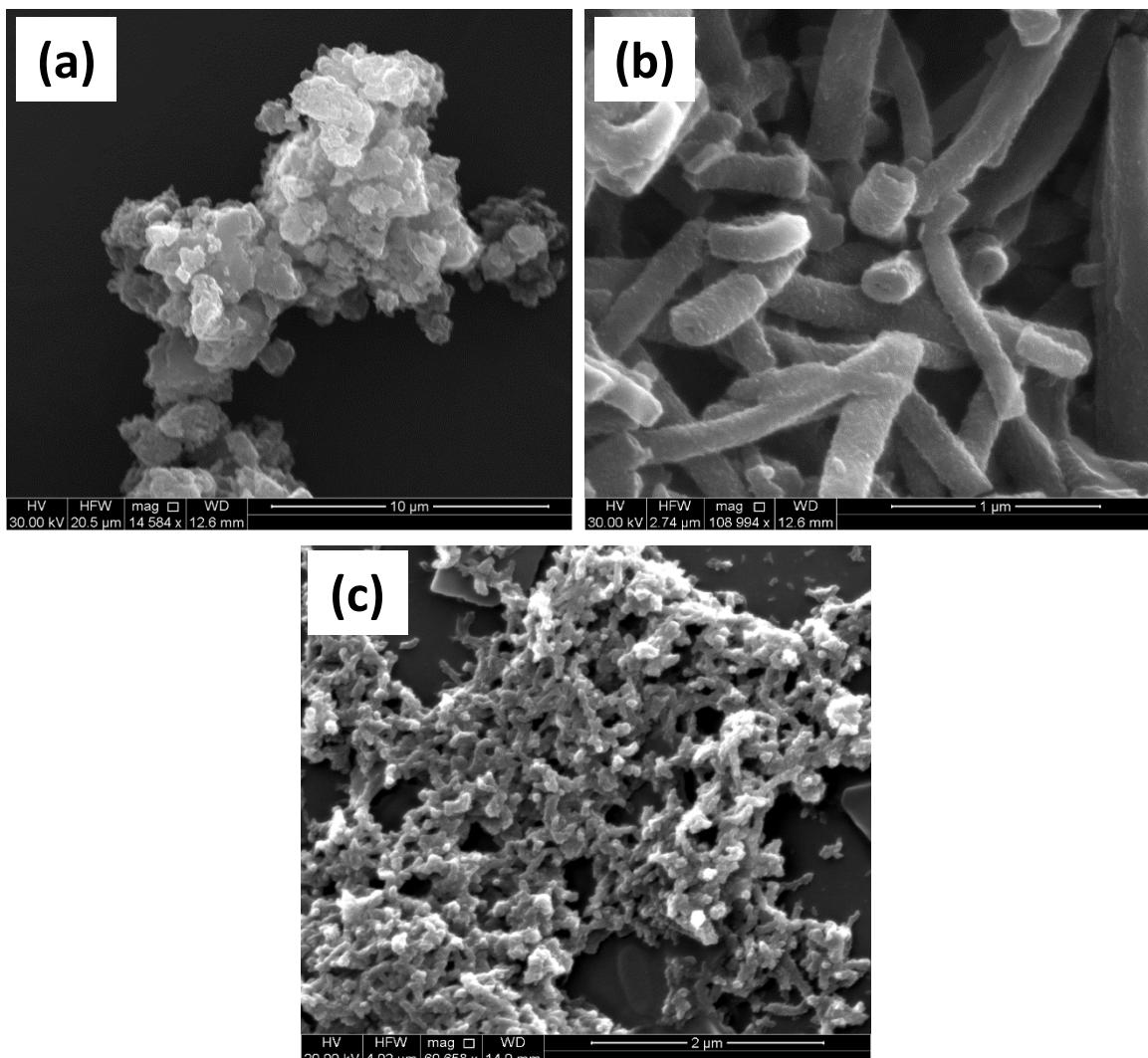
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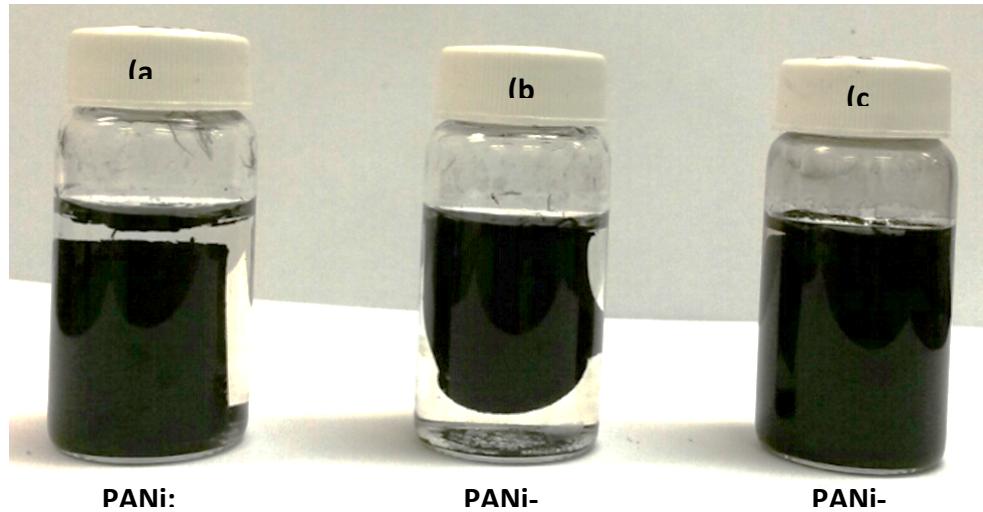
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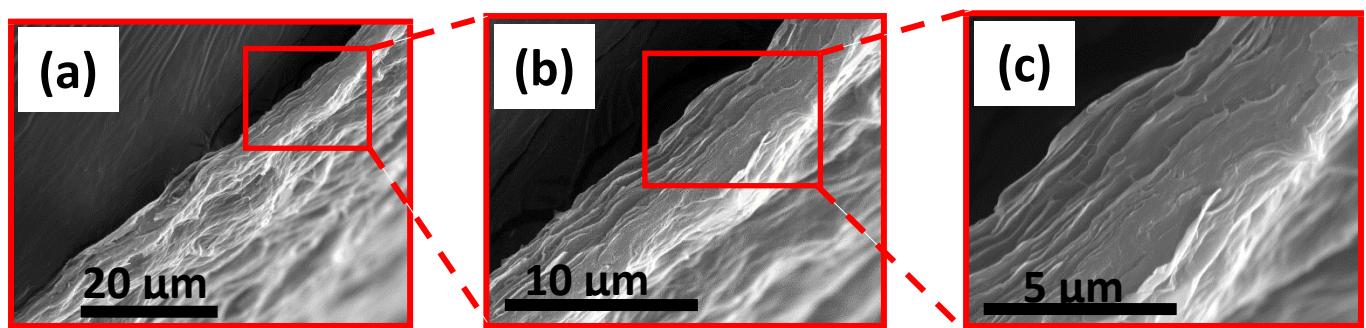
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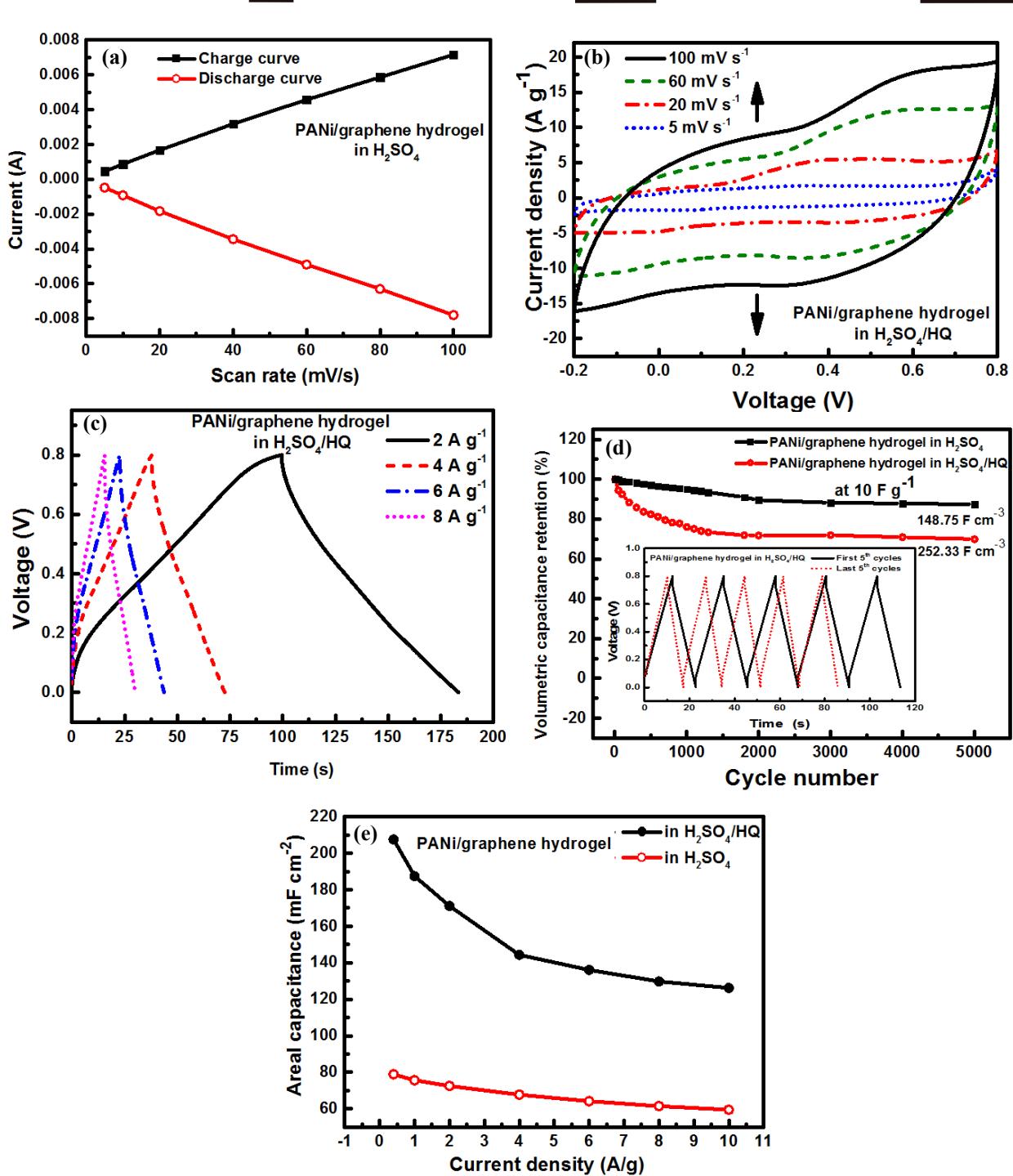
**Fig. S1.** SEM images of (a) a composite of polyaniline and poly (2-acrylamido-2-methyl-1-propanesulfonic acid (denoted PANi-PAMPA), (b) PANi-nanotubes and (c) PANi-nanofibers.



**Figure S2.** Photos of polyaniline/graphene hydrogel (PANI/ graphene hydrogel) in presence of (a) PANI- PAMPA, (b) PANI-nanotubes and (c) PANI-nanofibers.



**Figure S3.** SEM of free-standing PANi/graphene hydrogel film illustrates its layer structure at different magnifications.



**Figure S4.** Electrochemical performance of PANi/graphene hydrogel electrodes in  $\text{H}_2\text{SO}_4/\text{HQ}$ : (b) correlation between current and scan rate in  $\text{H}_2\text{SO}_4$ , (a) Cyclic voltammograms (CVs) at different scan rates in  $\text{H}_2\text{SO}_4/\text{HQ}$ , (b) The charge/discharge curves (CDs) at different current densities in  $\text{H}_2\text{SO}_4/\text{HQ}$ , (c) cycling performance in  $\text{H}_2\text{SO}_4/\text{HQ}$ , and (e) the areal capacitances at different current densities.

**Table S1.** Reported gravimetric and volumetric capacitances of some graphene materials.

Materials	Electrolyte	$\rho$ (g/cm <sup>3</sup> )	Electrode configuration	$C_{wt}$ (F/g)	$C_{vol}$ (F/cm <sup>3</sup> )	Ref.
<b>PANi/graphene hydrogel</b>	1 M H <sub>2</sub> SO <sub>4</sub>	1.02	Two	223.82	228.30	This work
<b>PANi/graphene hydrogel</b>	(1 M H <sub>2</sub> SO <sub>4</sub> /0.4 M HQ)	1.02	Two (0.4 A/g)	580.52 (0.4 A/g)	592.96 (0.4 A/g)	This work
<b>High density porous graphene</b>	6 M KOH	1.58	Two	238.00	376.00	[2]
<b>Carbon nanotubes-graphene fibres</b>	PVA/H <sub>3</sub> PO <sub>4</sub>	0.59	Two	-	300.00 (26.7 mA/cm <sup>3</sup> )	[3]
<b>Compact reduced graphene gel</b>	1 M H <sub>2</sub> SO <sub>4</sub>	1.25	Two (0.1 A/g)	191.70 (0.1 A/g)	255.5	[1]
<b>Holey graphene frameworks</b>	1 M H <sub>2</sub> SO <sub>4</sub>	0.71	Two (1 A/g)	310.00 (1 A/g)	220.10* (1 A/g)	[4]
<b>Porous carbon layer/graphene</b>	6 M KOH	-	Three (0.5 A/g)	481.00 (0.5 A/g)	212.00 (0.5 A/g)	[5]
<b>Graphene hydrogel</b>	1 M H <sub>2</sub> SO <sub>4</sub>	0.05	Three (0.3 A/g)	258.00 (0.3 A/g)	12.9* (0.3 A/g)	[6]
<b>Oriented graphene hydrogel</b>	1 M H <sub>2</sub> SO <sub>4</sub>	0.069	Two (0.1 A/g)	215.00 (0.1 A/g)	14.8* (0.1 A/g)	[7]
<b>Graphene hydrogel film</b>	1 M H <sub>2</sub> SO <sub>4</sub>	0.167	Two (1 A/g)	190.00 (1 A/g)	31.67* (1 A/g)	[8]
<b>Functionalized graphene hydrogel film</b>	1 M H <sub>2</sub> SO <sub>4</sub>	-	Two (1 A/g)	441.00 (1 A/g)	-	[9]
<b>Self-assembled graphene hydrogel</b>	5 M KOH	-	Two (1 A/g)	160.00 (1 A/g)	-	[10]

\* The volumetric capacitance ( $C_{vol}$ ) = gravimetric capacitance ( $C_{wt}$ ) × Packing density ( $\rho$ ) [1].

## References

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