Supporting Information

Hierarchical Graphene Oxide/Polyaniline Nanocomposites Prepared by Interfacial Electrochemical Polymerization for Flexible Solid-State Supercapacitors

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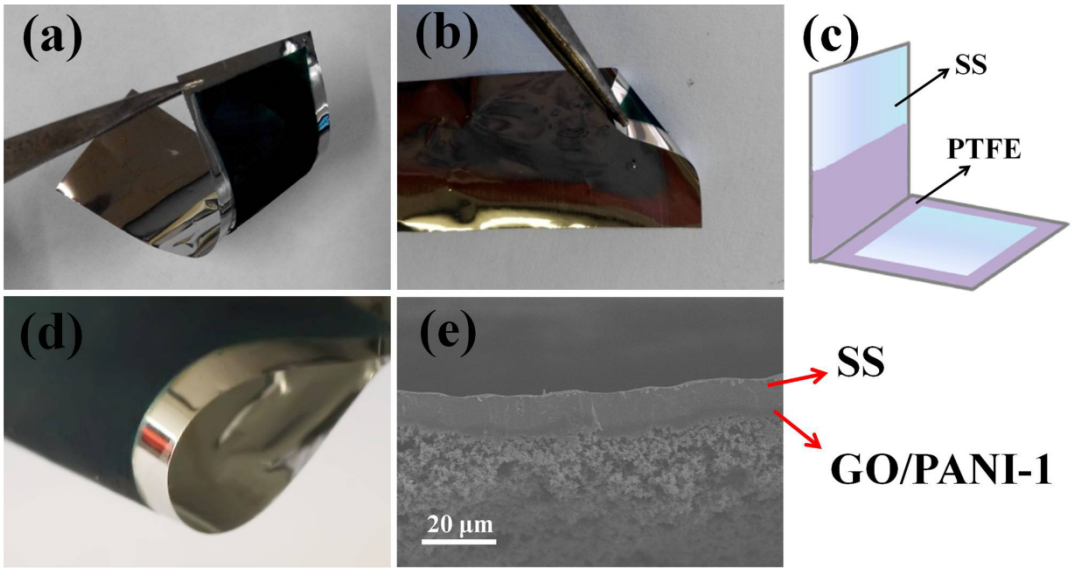

Fig. S1 Optical images of (a) one side (contact with water) and (b) the other side (contact with chloroform) of the electrode by interfacial electrochemical polymerization. (c) Scheme of the bended SS foil with Teflon layer cover. (d) The enlarged image of the bended electrode. (e) SEM image of the electrode at the cross-section.

After interfacial electrochemical polymerization, a uniform dark green GO/PANI-1 was obtained on only one surface side of the flexible SS. Good adhesion between GO/PANI and SS is also observed upon bending. SEM image of the electrode at the cross-section shows that a compact film of GO/PANI attached on the SS foil, indicating a high affinity.

Table S1 The mass weight of GO/PANI on each electrode of supercapacitors.

	Electrode A	Electrode B
1	0.96 mg	0.97 mg
2	1.02 mg	1.01 mg
3	1.03 mg	1.04 mg

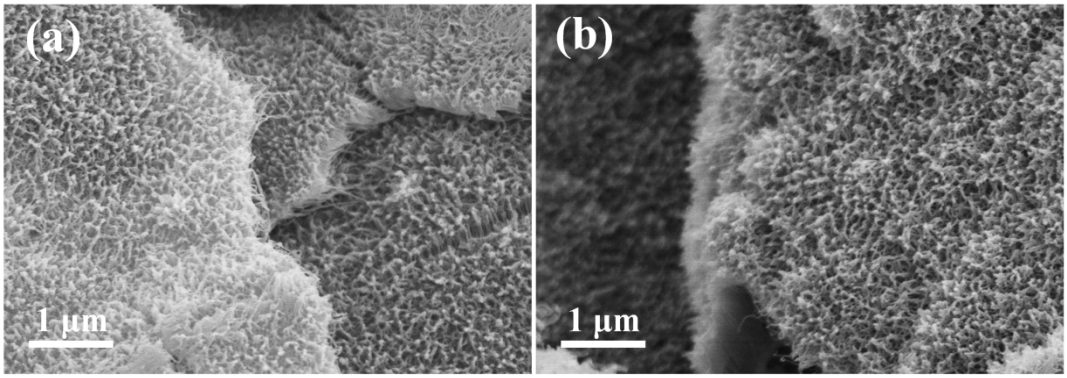

Fig. S2 SEM images of GO/PANI on two different electrodes under the same polymerization condition.

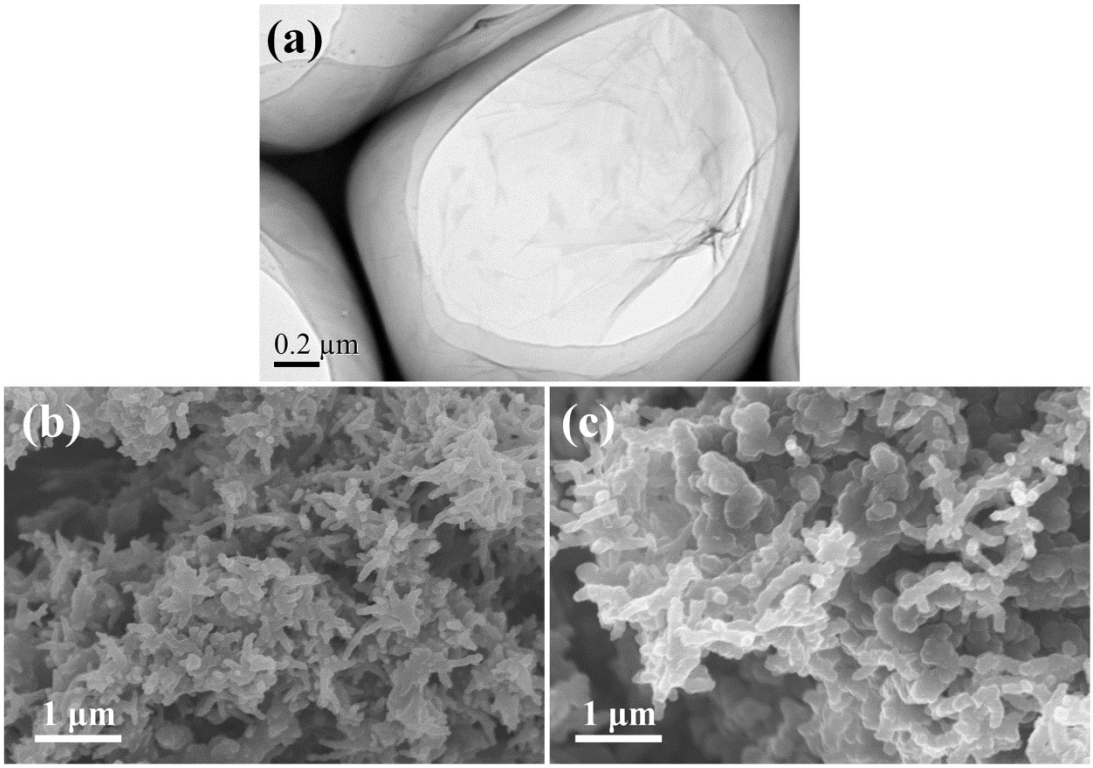

Fig. S3 (a) TEM image of GO. SEM image of (b) PANI-2 and (c) GO/PANI-2

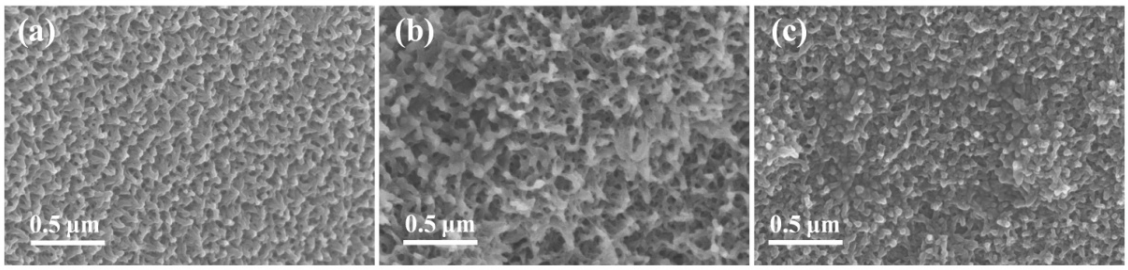

Fig. S4 SEM image of GO/PANI-1 prepared at the concentration of aniline (a) 0.05M, (b) 0.1M and (c) 0.5 M.

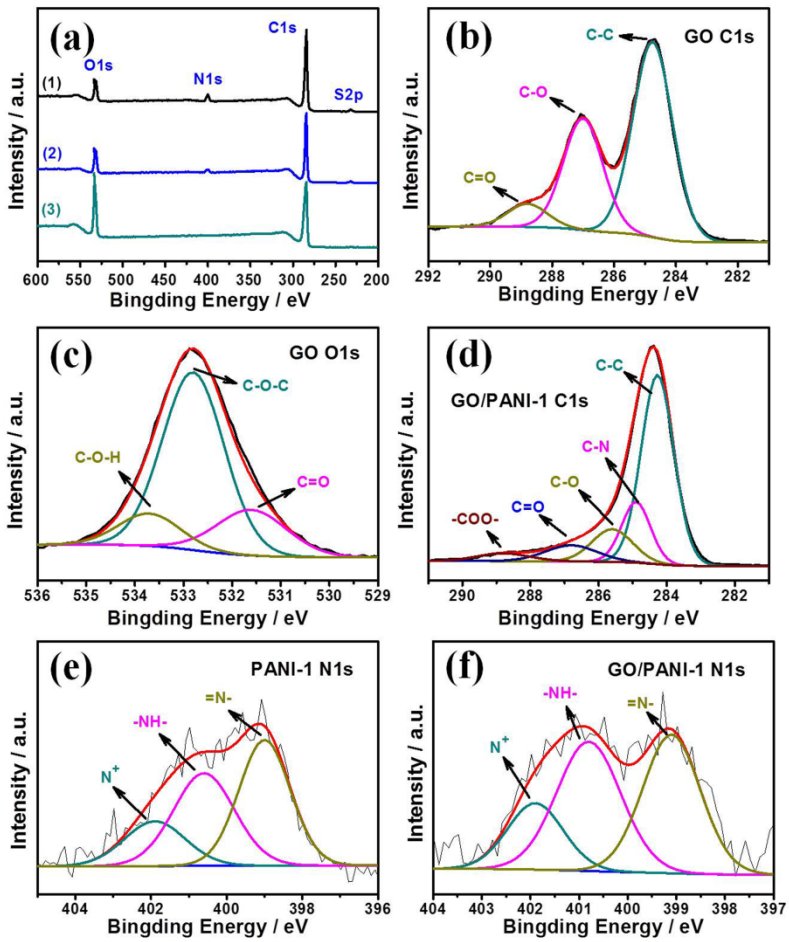

Fig. S5 (a) XPS spectra of (1) PANI-1, (2) GO/PANI-1 and (3) GO. (b) C 1s spectrum of GO. (c) O 1s spectrum of GO. (d) C 1s spectrum of GO/PANI-1. (e) and (f) N 1s spectrum of PANI-1 and GO/PANI-1, respectively.

Table S2 Elemental composition of PANI-1, GO/PANI-1 and GO in atomic percent.

sample	O (%)	N (%)	C (%)	S (%)
GO	22.91	0	76.57	0.52
PANI-1	8.02	6.81	82.17	3
GO/PANI-1	13.13	3.4	80.62	2.85

The atomic ratio of N/C and S/O in PANI-1 is 1/12.07 and 2.67, respectively. S/O in GO is 0.02. According to those ratios, C/O atomic ratio of GO component in GO/PANI-1 nanocomposite is calculated as 6.47.

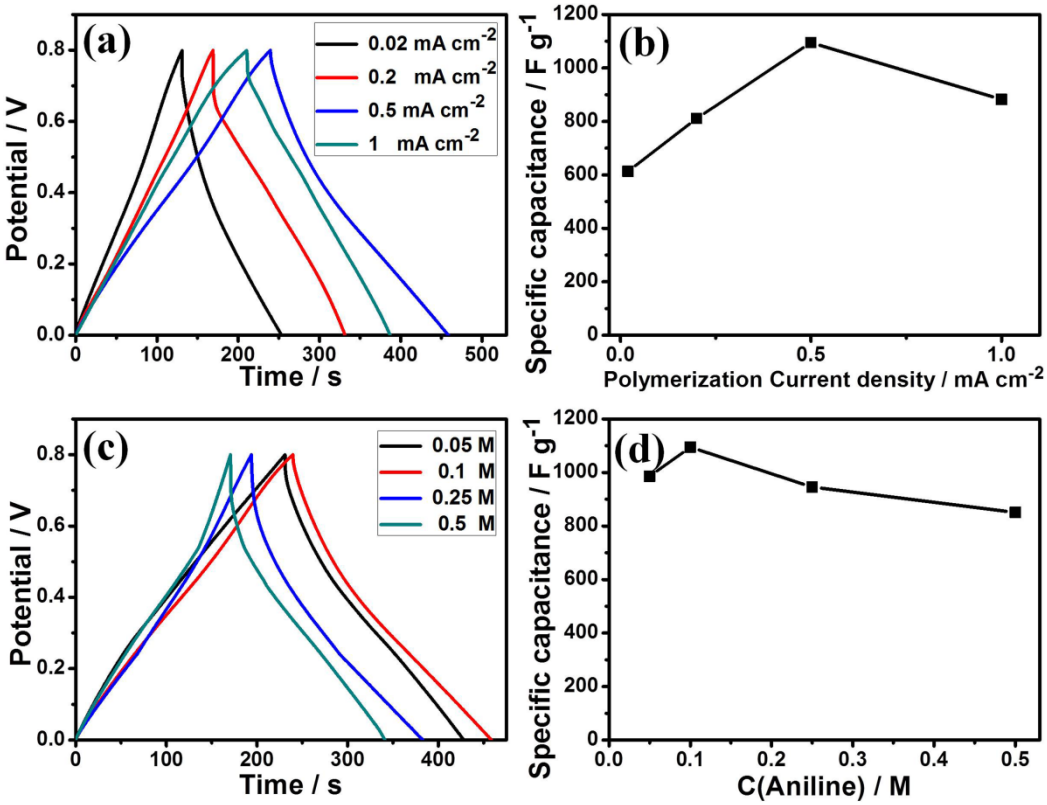

Fig. S6 (a) galvanostatic charge-discharge curves and (b) mass specific capacitance of GO/PANI-1 nanocomposites at various current densities of polymerization (0.02, 0.2, 0.5, 1 mA cm^{-2}), (c) galvanostatic charge/discharge curves and (d) mass specific capacitance of GO/PANI-1 nanocomposites at various concentrations of aniline (0.05, 0.1, 0.25, 0.5 M).

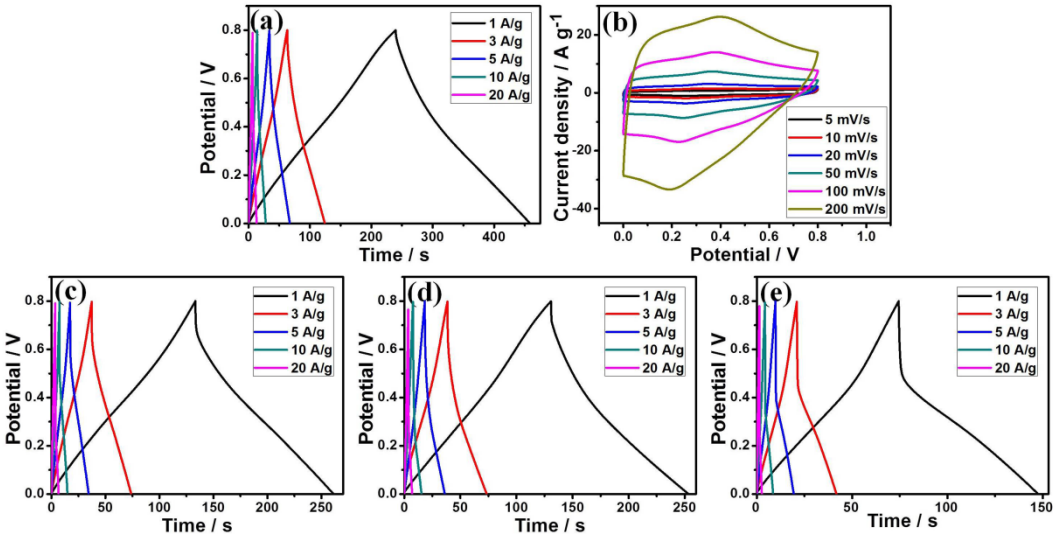

Fig. S7 (a) Galvanostatic charge-discharge curves of the GO/PANI-1 supercapacitor at different current densities. (b) CV cures of GO/PANI-1 supercapacitor at different scan rates. Galvanostatic charge-discharge curves of supercapacitors based on (c) GO/PANI-2, (d) PANI-1 and (e) PANI-2 at different current densities.

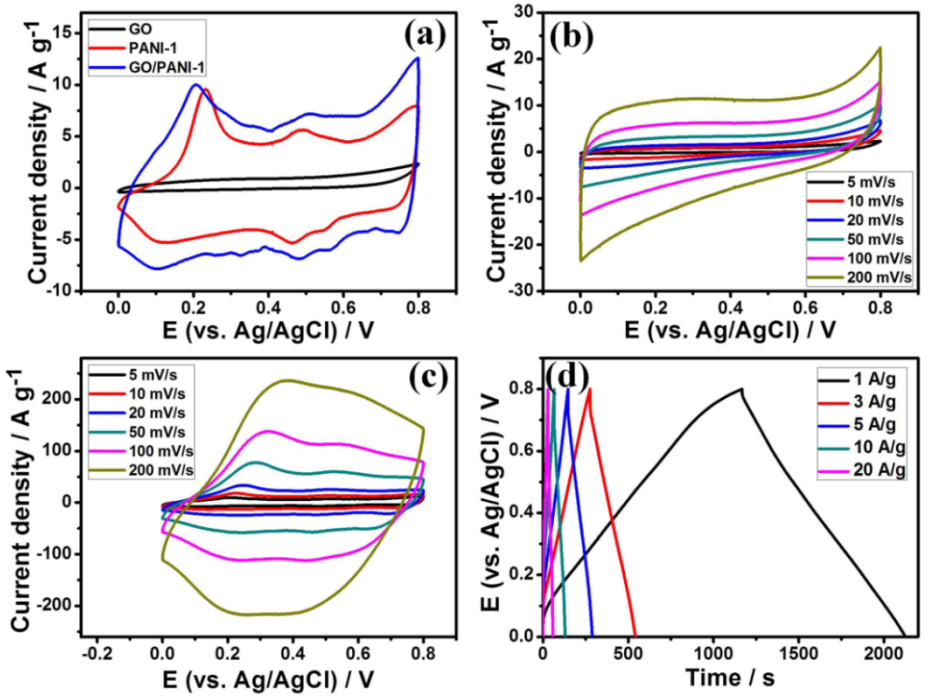

Fig. S8 (a) CV curves of GO, PANI-1 and GO/PANI-1 nanocomposite at 5 mV s^{-1}. (b) and (c) CV curves of GO and GO/PANI-1 nanocomposite at different scan rates. (d) Galvanostatic charge-discharge curves of GO/PANI-1 nanocomposite at different current densities.

The basic electrochemical properties of the electrode materials are evaluated in 1 M H_2SO_4 in a three-electrode configuration with a Pt foil as counter electrode and Ag/AgCl as reference electrode. In Fig. S8a, GO/PANI-1 and PANI-1 show two pairs of redox peaks and much higher current density than that of GO, indicating that the capacitance of GO/PANI-1 mainly arises from PANI-1. Besides, the increase in current with different scan rates demonstrates a good rate capability of GO and GO/PANI-1 (Fig. S8 b and c). According to Fig. S8d, a high specific capacitance of 1200 F g^{-1} of GO/PANI-1 was gained at 1 A g^{-1} according to the following equation $C_S = (I \times t) / (m \times \Delta V)$, Where m is the mass of active material of the electrode (g).

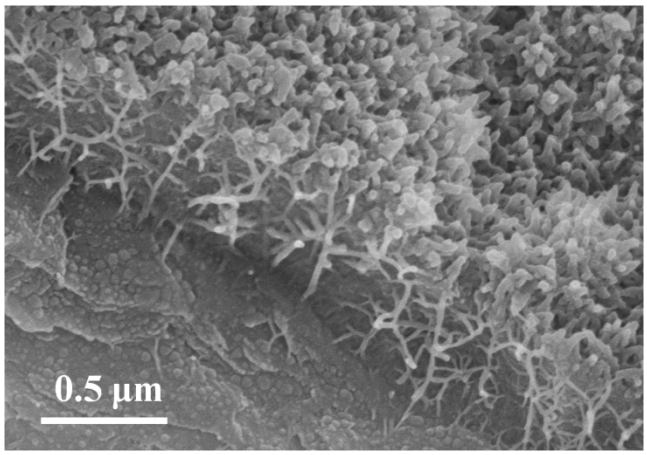

Fig. S9 A high-resolution SEM image of GO/PANI-1 showing a well contact between GO and PANI.

Calculations of energy density and power density.

The energy density (E/Wh kg^{-1}) and power density (P/kW kg^{-1}) of the supercapacitors were calculated according to the following equations:

$$E=CV^2/8 \quad \text{(S1)}$$

$$P=V^2/4R_{ESR}M \quad \text{(S2)}$$

Where C is the specific capacitance of electrode materials (F g^{-1}), V is the maximum potential of charge (V), Equivalent series resistance (R_{ESR}) was estimated according the formula $R_{ESR}=V_{drop}/(2I)$, where V_{drop} (V) is the voltage drop at the beginning of the discharge curve. M is the mass of active material of the whole device (g).

Table S3 The performance of GO/PANI-1 electrode material compared with previous studies based on two-electrode solid-state supercapacitors.

	electrolyte	C_S (F g^{-1})/I	Capacitance retention/I (1000 cycles)	E_{max} ($WhKg^{-1}$)	Ref
In this work	PVA-H_2SO_4	1095/1 Ag^{-1}	91.1%/1Ag^{-1}	19.3	
rGO/PANI	PVA-H_3PO_4	~3892/2.5Ag^{-1}	~90%/2.5Ag^{-1}		24
Graphene/PANI	PVA-H_3PO_4	~980/0.75Ag^{-1}	89%/1mAcm^{-2}	23.2	29
Carbon paper/PANI	PVA-H_2SO_4	~620/1 Ag^{-1}	102%/5Ag^{-1}	14.3	32
CNT/PANI	PVA-H_2SO_4	332/~1 Ag^{-1}	91.9%/1Ag^{-1}	7.1	33

The tests are conducted in a two-electrode system based on the symmetrical all-solid-state supercapacitors. Here the Cs is the specific capacitance of each electrode materials calculated according to equation $C_S = 4(I \times t) / (M \times \Delta V)$. The cycling number is 1000 for all the supercapacitor s.Energy densities (E_{max}) are calculated according to equation $E=CV^2/8$.