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Electronic Supplementary Information

Facilely prepared polypyrrole reduced graphene oxide composite with crumpled surface for high performance supercapacitor electrodes

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Experimental Section

GO was synthesized using a modified Hummer's method and dispersed in water with the concentration of 1.0 mg/1.0 g H_2O by ultrasonication. The monodisperse PPy spheres were prepared according to reported method with a few modifications. In a typical synthesis, 0.20 g of ferrous chloride ($FeCl_2$) to 88.0 mL of deionized water, where 2.0 mL of pyrrole has been pre-dispersed. After the addition of 10.0 mL of H_2O_2 to the pyrrole/$FeCl_2$/H_2O mixture, pyrrole polymerization was initiated and lasted for 6 h, leading to a deep dark precipitate. The products were concentrated by centrifugating the solution and sequentially washed with water several times to remove unused reactants and reaction byproducts, and then were dispersed in distilled water. PPyRGO composites with different ingredient mass ratios from 4:1 to 1:2 for PPy and RGO are signed as PPyRGO ratio. Here we take the PPyRGO11 composite as an example, indicating that the mass ratio of PPy and RGO is 1:1. The procedure are the follows: PPy spheres were dispersed in water with the concentration of 0.05 g PPy/ 50.00 g H_2O; the solution was adjust to pH=2, and heated to 70 ˚C; 50 mL GO disperision was dropped into the solution by constant flow pump at the speed of 10.0 mL/h; the resulting PPyGO composite was then reduced by 0.50 mL hydrazine hydrate at 90 ˚C for 6 h. The PPyRGO11 composite was obtained.

Raman analysis was performed with a Jobin Yvon HR800. High-resolution Transmission electron microscopy (HRTEM) images were obtained with a FEI Tecnai F30 operated at 200 kV. X-ray diffraction (XRD) analysis was performed on a Shimadzu XRD 6000 diffractometer. Scanning electron microscopy (SEM) imaging was performed on a JEOL-JSM-7600F SEM. Zeta potential was recorded on a Malvern Nano-Z Instrument. UV-Vis spectra were obtained with a Lambda 35 UV-vis spectrometer. The electrical conductivity of all synthesized materials at room temperature was determined using the four-probe system connected to a Keithley 2400 source meter. All electrochemical measurements were carried out with a CHI660d (Shanghai CH Instrument Company, China). Cyclic voltammetry (CV) and galvanostatic charge-discharge were performed with a conventional three-electrode system consisting of a bare or modified glassy carbon electrode (GCE; diameter=3mm) as the working electrode, a saturated calomel electrode (SCE) as the reference electrode, and a platinum wire as the counter electrode in 1 M KCl solution.

Zeta potential tests

Zeta potential could reflect the surface properties of colloids, and is strongly affected by the pH of the dispersion. We have measured the zeta potential of PPy and GO in a series of pH ranging from 1 to 7 (see Fig. S1, ESI). The pH of the dispersion in the synthesis process was 2. It can be found in the figure that at pH=2, PPy and GO are oppositely charged. Hence, electrostatic interaction could be a contributing factor in the coating process. However, after the reduction of GO into RGO, π-π accumulation has replaced it to be the dominating interaction. Therefore, the product can still be very stable at pH=7.

UV-Vis spectra tests

UV-Vis spectra (see Fig. S2, ESI) of PPy, GO and PPy-GO are collected to further confirm the interaction between GO sheets and PPy spheres. The curve of GO dispersion exhibits a peak at 228 nm and a tiny shoulder at 300 nm, which are attributed to π-π* transitions of aromatic C=C bonds and n-π* transitions of C=O bonds28. The figure illustrates that the absorption peak at 228 nm for GO shifts to 236 nm with the addition of PPy. That means the energy of π-π* transitions of aromatic C=C bonds in GO has changed significantly by the addition of PPy, which is very likely caused by the π-π stacking between PPy and GO.

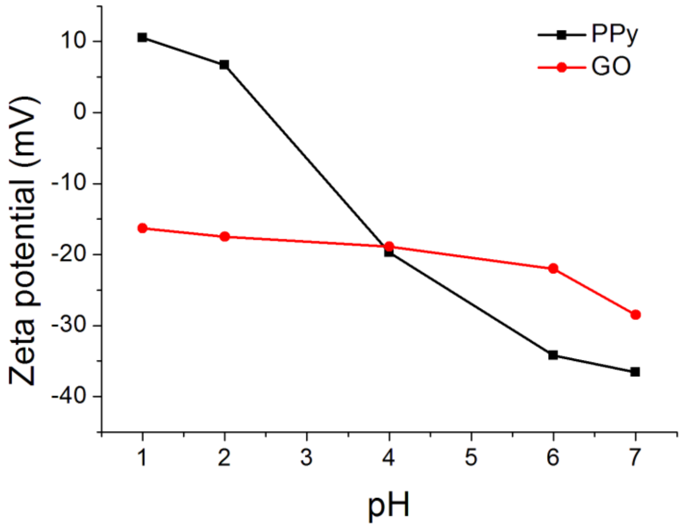

Fig. S1 Zeta potential of PPy and GO in a series of pH ranging from 1 to 7.

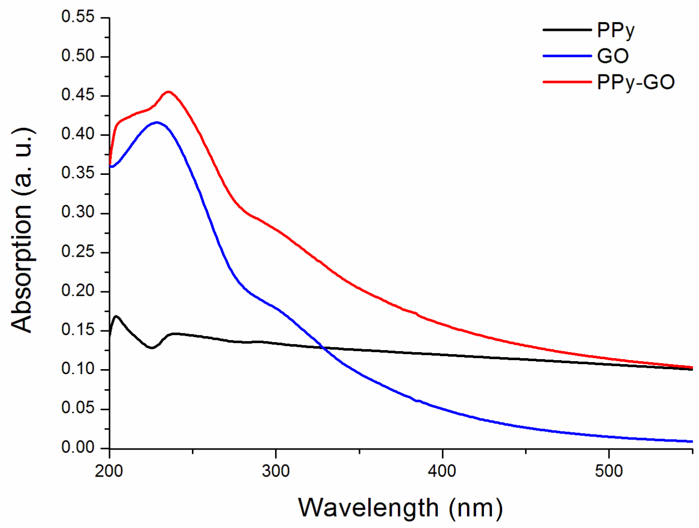

Fig. S2 UV-Vis spectra of PPy (72 mg L^{-1}), GO (8 mg L^{-1}), and PPy-GO (80 mg L^{-1}).

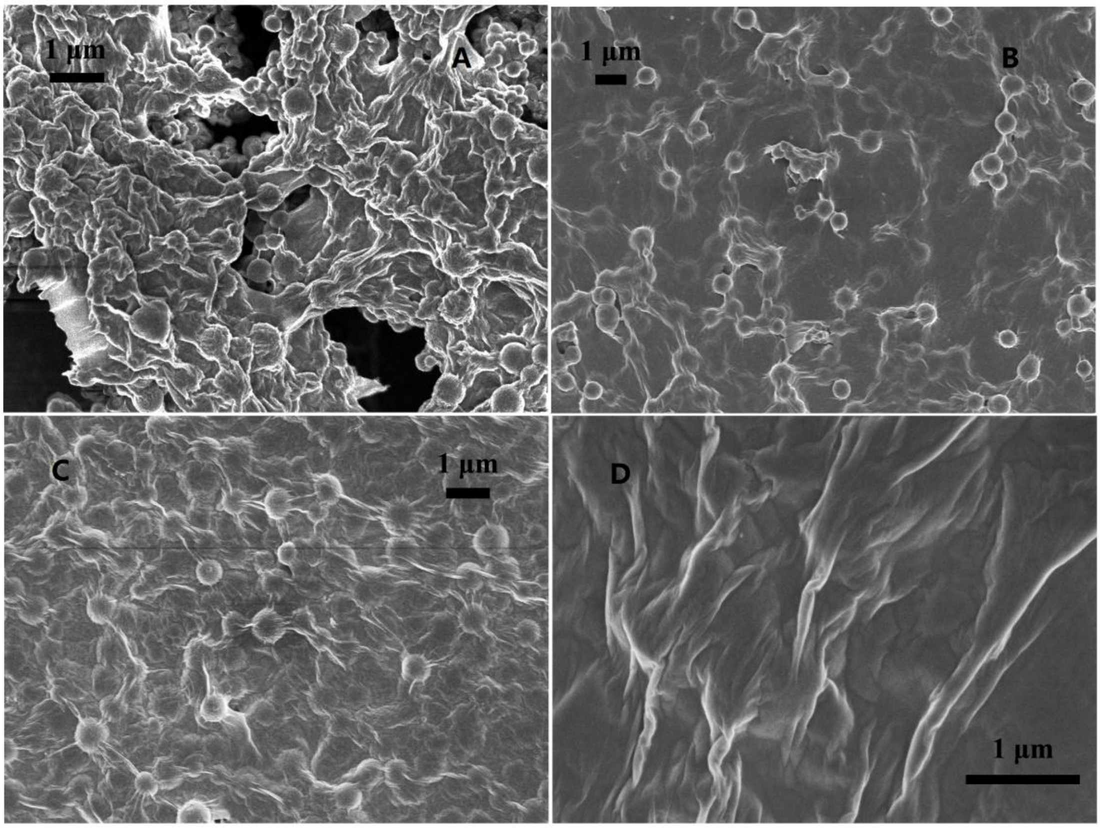

Fig. S3 SEM images of PPyRGO composite films with weight ratio of (A) 4:1, (B) 2:1, (C) 1:1, and (D) 1:2 respectively.

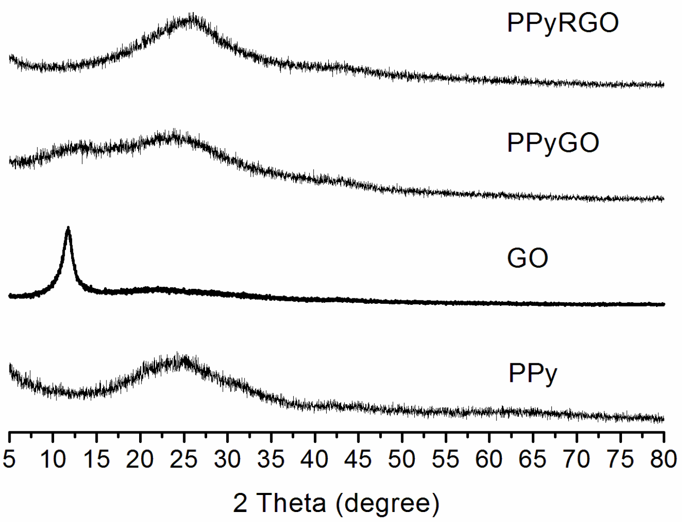

Fig. S4 XRD patterns of PPy, GO, PPyGO and PPyRGO composites.

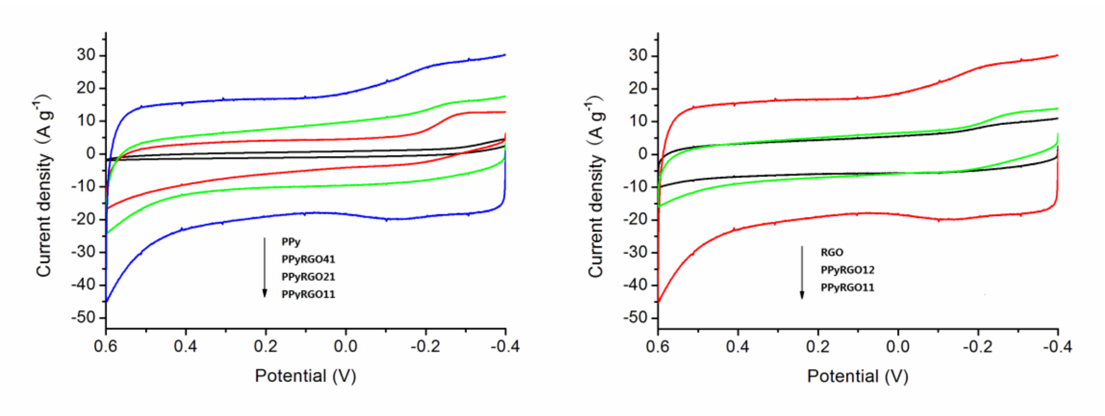

Fig. S5 CV curves of different materials modified electrodes in 1.0 M KCl in a three-electrode system.

Table S1 Electrical conductivity data of different materials.

	PPyRGO41	PPyRGO21	PPyRGO11	PPyRGO12
Conductivity (S/cm)	0.00036	0.029	5.4	4.6