

Supplementary information

**Bi-functional sulphate-coupled reduced graphene oxide as efficient dopant for
conducting polymer with enhanced electrochemical performance**

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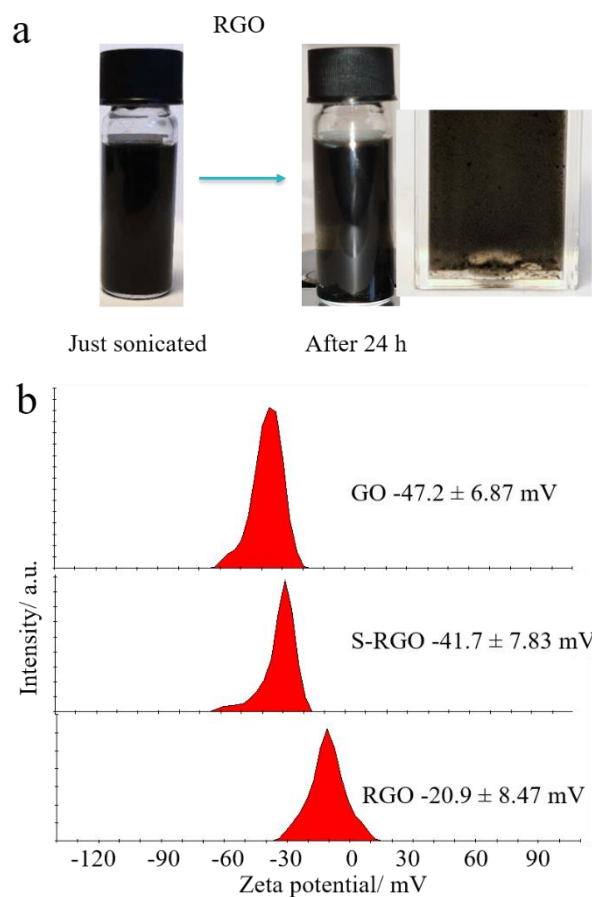


Figure S1. (a) Digital images of RGO aqueous suspensions in 1 mg mL^{-1} just sonicated and 24 h later. The magnified image of RGO in a cuvette showed that RGO tends to aggregate together and form sediments at the bottom of the cuvette after storing at room temperature for 24 h. (b) comparison of the zeta potential for GO, RGO and S-RGO aqueous suspensions.

Table S1. The atomic percentage of C, S, N and O, and the carbon-containing functional groups in S-RGO and GO.

Samples	Element content/at%					Carbon-containing functional groups/at%				
	C	O	S	N	O/C (at%/at%)	C-C	C-O ¹	C=O	C-S	C-N ²
S-RGO	80.26	13.97	4.00	1.77	0.17	82.86	1.89	1.05	9.42	4.68
GO	51.9	48.1	-	-	0.93	42.01	53.01	4.98	-	-
RGO	74.83	19.54	-	-	0.26	68.35	27.16	4.48	-	-

Note: ¹C-O: C-OH/C-O-C, ²C-N: C-N/C=N.

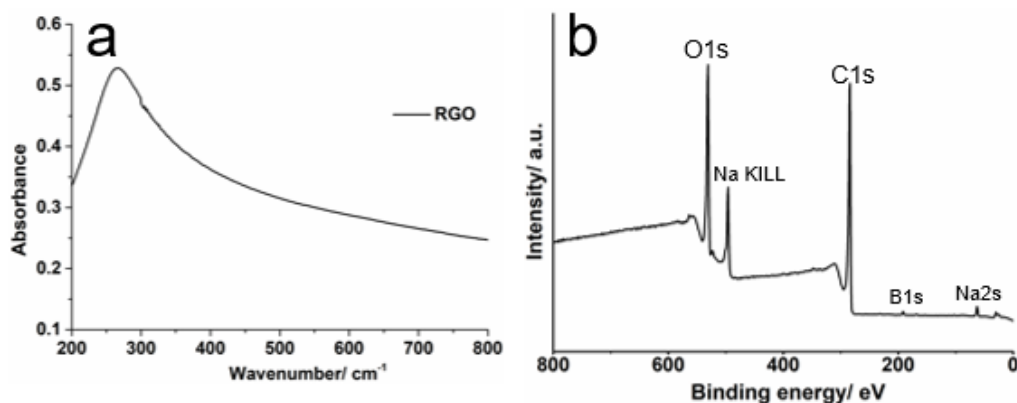


Figure S2. (a) UV-vis spectra of RGO; (b) XPS full survey of RGO; (c) atomic percentage of RGO calculated by XPS.

In Figure S2a, the absorption band is located at 264.7, which is close to S-RGO (263 nm) and red-shifted compared to GO (230 nm), indicating the successful reduction of the GO to RGO. The XPS full survey shows two main characteristic peaks at 284 and 531 eV, which are attributed to C1s and O1s. Small peaks were observed at 64 and 192 eV and can be ascribed to the residue of sodium borohydride. The O/C automatic ratio of RGO is 0.26, which is much smaller than that of GO (0.93) and slightly larger than that of S-RGO (0.17). The lower content of oxygen in S-RGO via 4-HBS reduction compared to RGO via sodium borohydride reduction, indicates that 4-HBS might be better reductant than sodium borohydride for GO reduction.

Table S2. Peak current and peak separation for GO and S-RGO.

Probe	Material	$I_{p,oxi}/\mu A$	$I_{p,red}/\mu A$	E_{oxi}/V	E_{red}/V	$\Delta E_p/mV^1$
$[Ru(NH_3)_6]^{2+/3+}$	GO	4.62	5.56	-0.160	-0.290	130
	S-RGO	87.50	71.15	-0.200	-0.295	95
$[Fe(CN)_6]^{3-/4-}$	GO	1.97	2.05	0.305	0.135	170
	S-RGO	23.4	19.6	0.330	0.115	215

Note: $^1\Delta E_p = E_{oxi} - E_{red}$

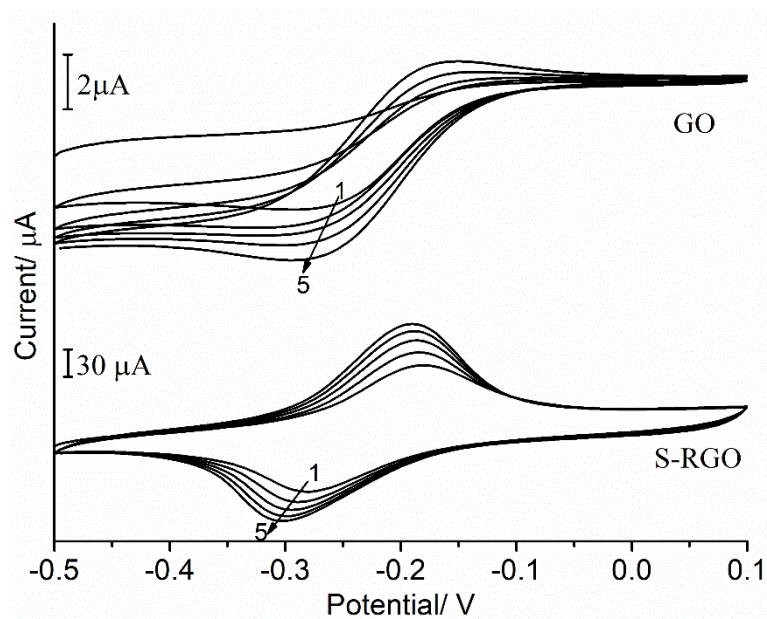


Figure S3. Successive 5 cycles of CV scans of GO and S-RGO modified electrodes in 0.1 M KCl containing 2 mM $[\text{Ru}(\text{NH}_3)_6]^{2+/3+}$.



Figure S4. Digital image of free-standing GO, RGO, and S-RGO prepared by vacuum filtration.

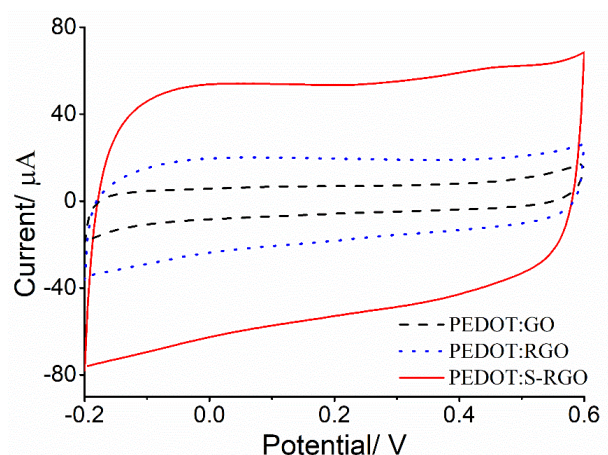


Figure S5. CVs of the corresponding PEDOT:GO, PEDOT:RGO and PEDOT:S-RGO in 0.1 M KCl, scan rate 50 mV s⁻¹.

The CVs of the corresponding PEDOT:S-RGO, PEDOT:GO and PEDOT:RGO electrodes were then examined in 0.1 M KCl. As shown in Figure S5, PEDOT:S-RGO displayed a better quasi-rectangular shaped CV curve compared to those of PEDOT:GO and PEDOT:RGO, indicating the better ideal electrical double-layer supercapacitor characteristic of the PEDOT:S-RGO. The interfacial capacitance (calculated from the voltammetric curves by summing the charge current density in the forward and reverse scans and dividing the sum by twice the scan rate) for PEDOT:S-RGO was estimated to be 14.4 mF cm⁻², which is 2.8 and 7.8 times higher than that of PEDOT:RGO (5.1 mF cm⁻²) and PEDOT:GO (1.8 mF cm⁻²), respectively. Thus the S-RGO resulted in improved electropolymerisation efficiency of PEDOT compared to GO and RGO.

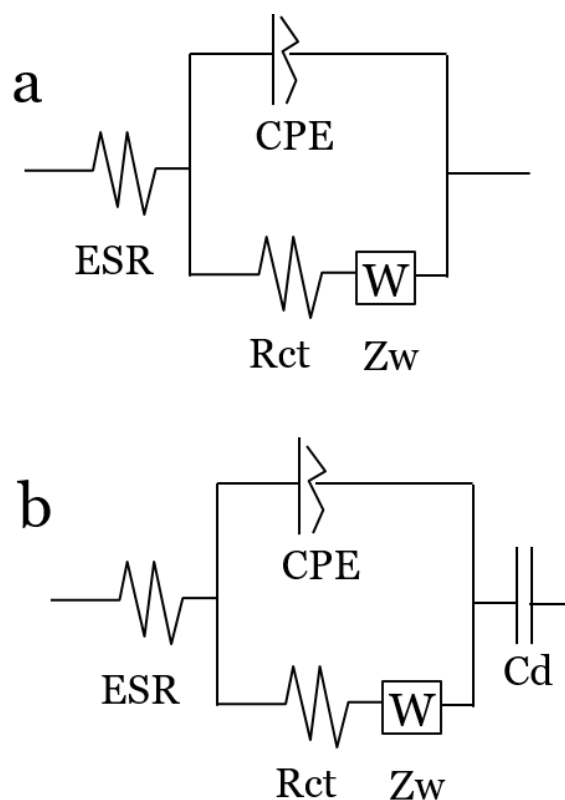


Figure S6. (a) Equivalent circuit for bare electrode; (b) Equivalent circuit for PEDOT:GO, PEDOT:RGO and PEDOT:S-RGO modified electrode.

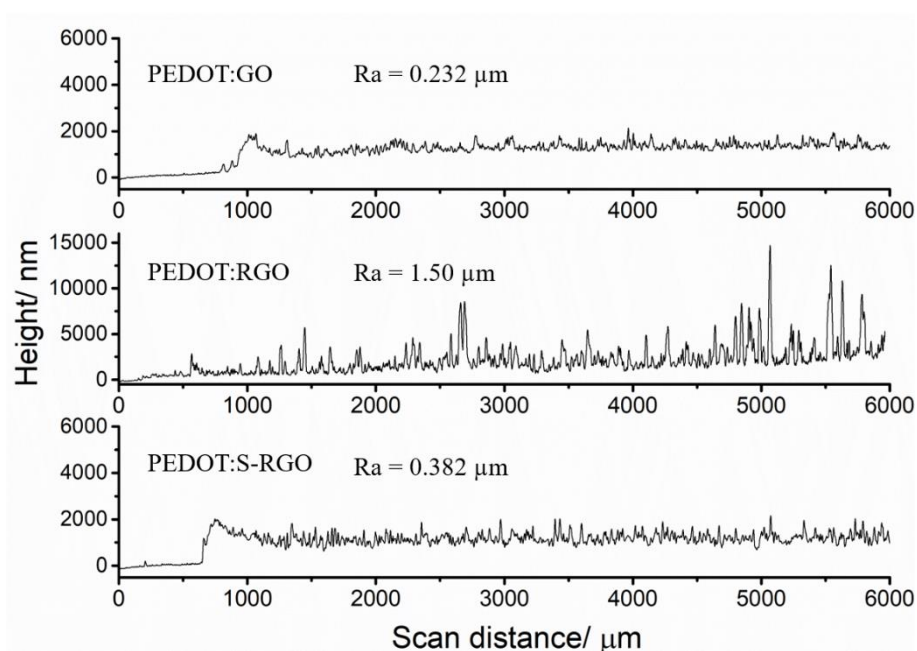


Figure S7. Surface profiles of PEDOT:GO, PEDOT:RGO and PEDOT:S-RGO by a Dektak 6 M surface profilometer (Veeco Inc. USA) with a scanning range of 6000 μm.

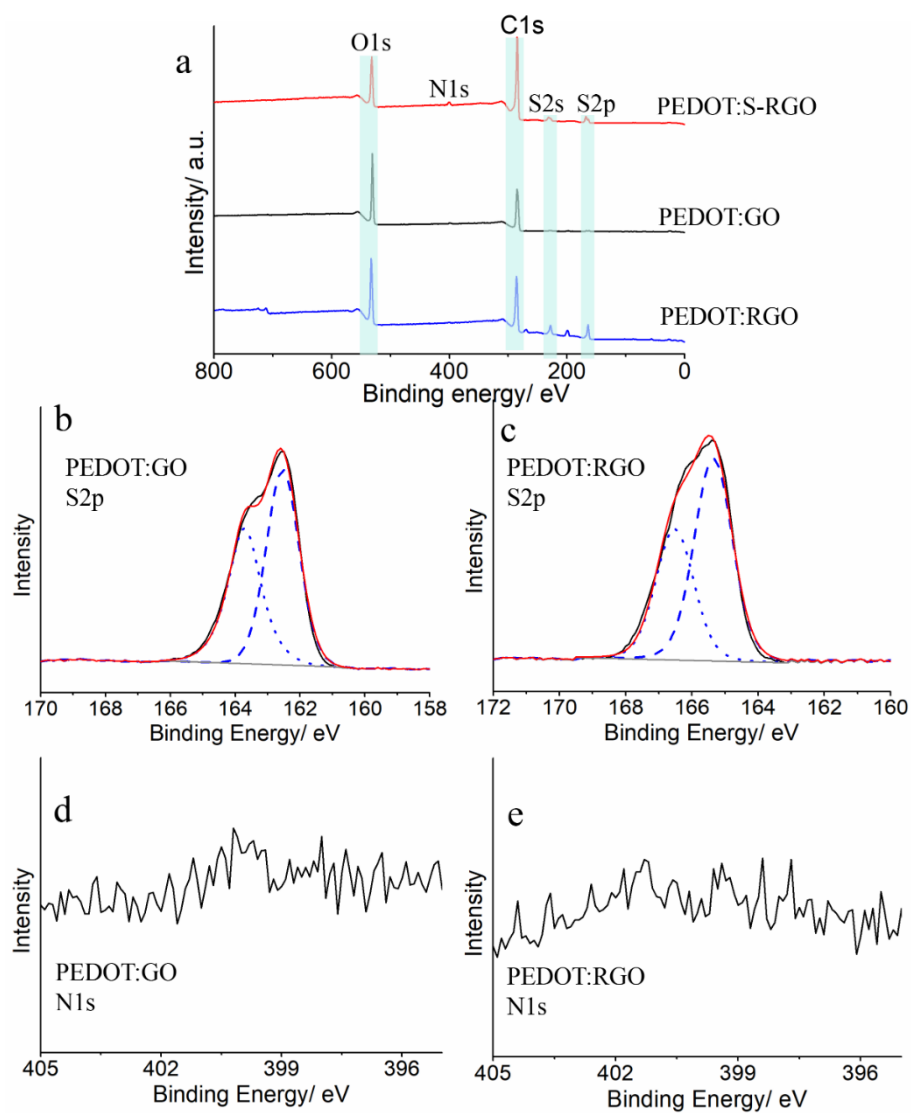


Figure S8. (a) XPS full survey of PEDOT:S-RGO, PEDOT:GO and PEDOT:RGO; S2p spectrum of PEDOT:GO (b) and PEDOT:RGO (c); N1s spectrum of PEDOT:GO (d) and PEDOT:RGO (e).

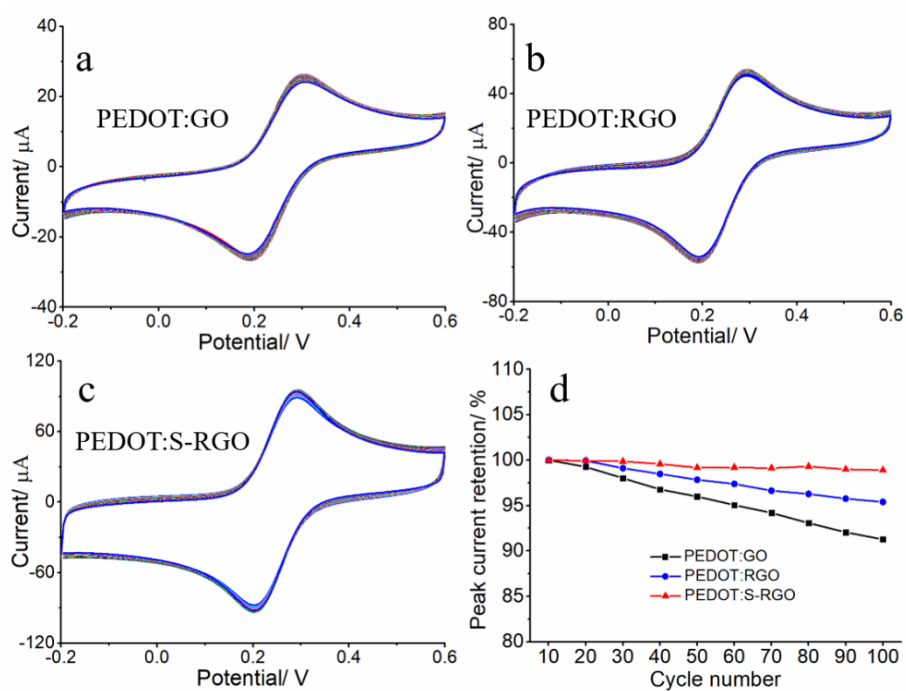


Figure S9. Cyclic stability test by CVs of PEDOT:GO (a), PEDOT:RGO (b) and PEDOT:S-RGO (c) for successive 100 cycles in 0.1 M KCl containing 5 mM $\text{Fe}(\text{CN})_6^{3-/4-}$, scan rate 50 mV s^{-1} ; (d) Plotting of oxidation peak current changes (every 10th cycles) with the function of cycle number.