Electronic Supplementary Information

Fluoro-polymer Functionalized Graphene for Flexible Ferroelectric Polymer-based High-*k* Nanocomposites with Suppressed Dielectric Loss and Low Percolation Threshold

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Calculation Method of Density

In this work, the density of the PDA-RGO and PF-PDA-RGO can be calculated based on the results of TGA (Fig. 1B).

For PDA-RGO, its density can be calculated by the following equation:

$$\rho_{PDA-RGO} = 1/\{\frac{W_{PDA-RGO} - W_{H-RGO}}{\rho_{PDA}} + \frac{1-(W_{PDA-RGO} - W_{H-RGO})}{\rho_{RGO}}\} \quad (1)$$

For PF-PDA-RGO, its density can be calculated by the following equation:

$$\rho_{PF-PDA-RGO} = 1/\{\frac{W_{PF-PDA-RGO} - W_{H-RGO}}{\rho_{PF-PDA}} + \frac{1-(W_{PF-PDA-RGO} - W_{H-RGO})}{\rho_{RGO}}\} \quad (2)$$

where $\rho_{PDA-RGO}$ is the density of PDA-RGO, $\rho_{PF-PDA-RGO}$ is the density of PF-PDA-RGO, ρ_{PDA} is the density of PDA (≈ 1.5 g/cm^3), ρ_{PF-PDA} is the density of PF-PDA (≈ 1.5 g/cm^3), ρ_{RGO} is the density of RGO (≈ 2.2 g/cm^3), W_{H-RGO} is the weight loss of H-RGO (12.49%, line b in Fig. 1B), $W_{PDA-RGO}$ is the weight loss of PDA-RGO (43.18%, line c in Fig. 1B), and $W_{PF-PDA-RGO}$ is the weight loss of PF-PDA-RGO (54.03%, line d in Fig. 1B), respectively. Therefore, according to the equation (1) and (2), $\rho_{PDA-RGO}$ is about 1.92 g/cm^3 and $\rho_{PF-PDA-RGO}$ is about 1.84 g/cm^3.

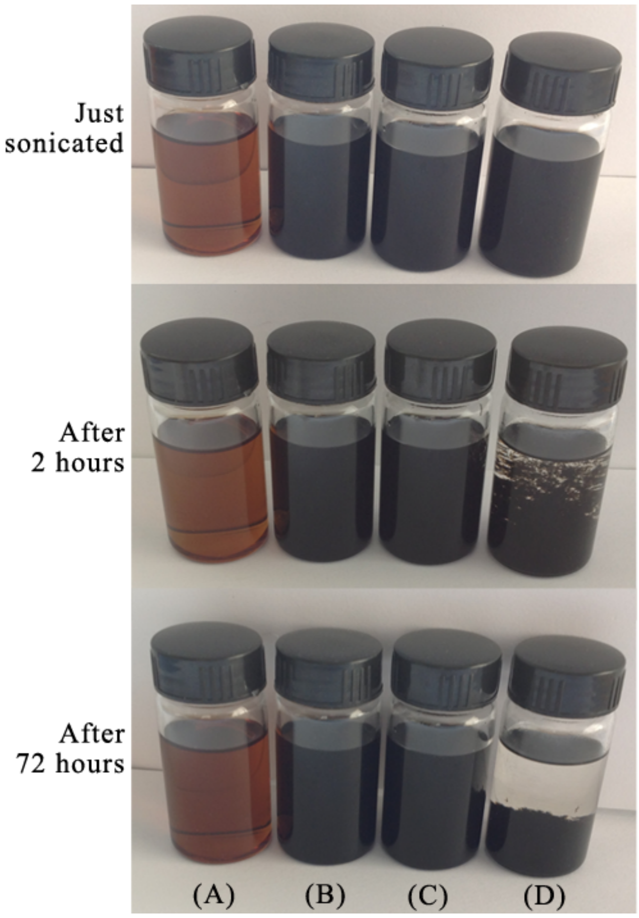

Fig. S1 Digital photographs of suspensions of (A) GO, (B) PDA-RGO, (C) PF-PDA-RGO, and (D) H-RGO nanosheets in DMF. Top: dispersions immediately after sonication. Middle: dispersions 2 h after sonication. Bottom: dispersions 72 h after sonication.

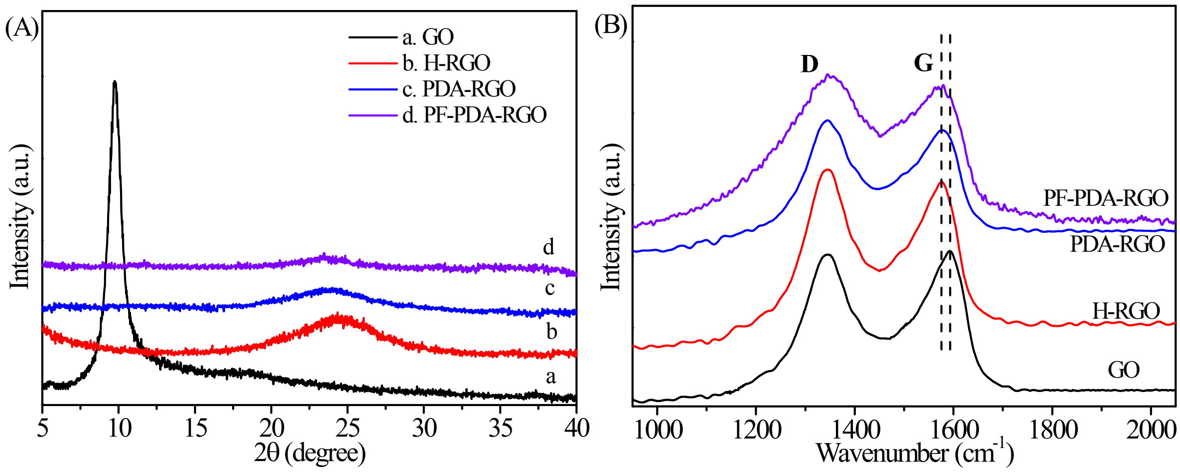

Fig. S2 (A) XRD patterns and (B) Raman spectra of GO, H-RGO, PDA-RGO and PF-PDA-RGO.

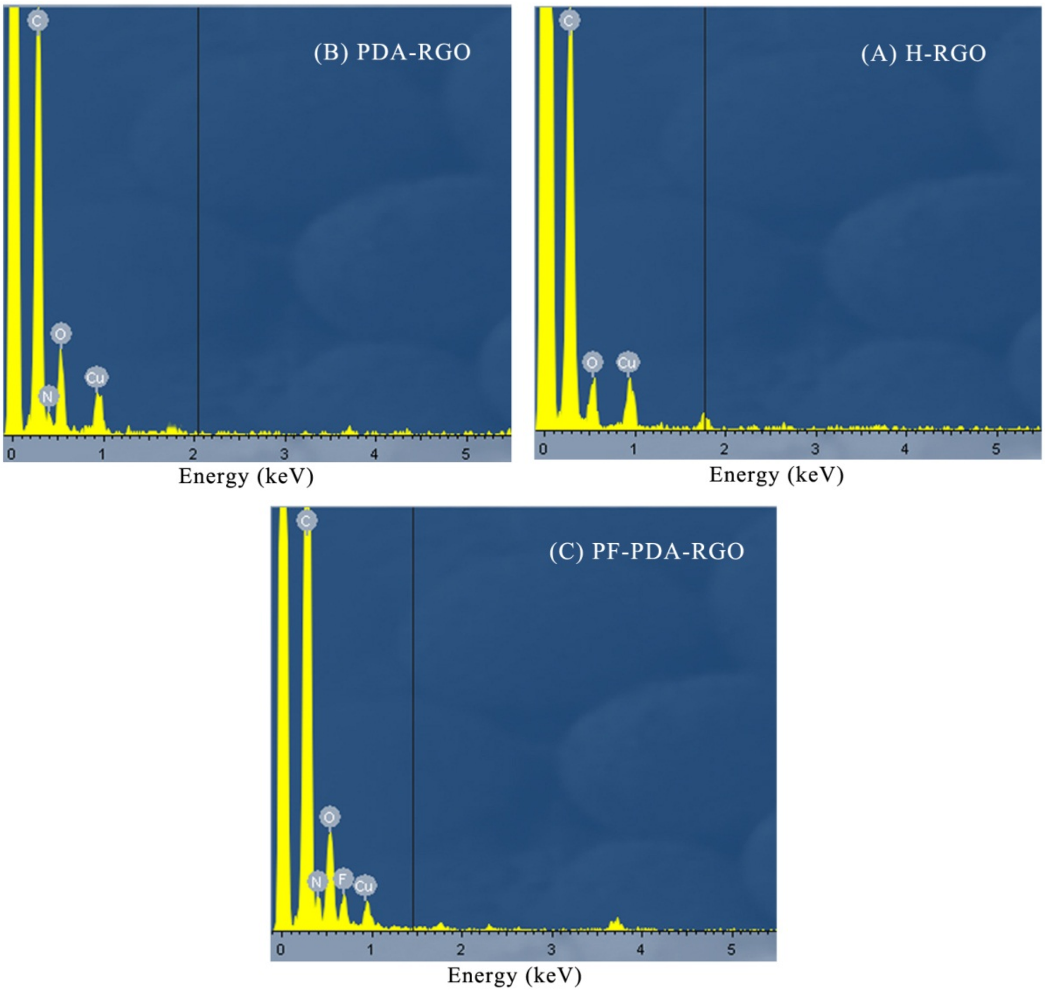

Fig. S3 The energy-dispersive X-ray (EDX) analysis of (A) H-RGO, (B) PDA-RGO, and (C) PF-PDA-RGO nanosheets.

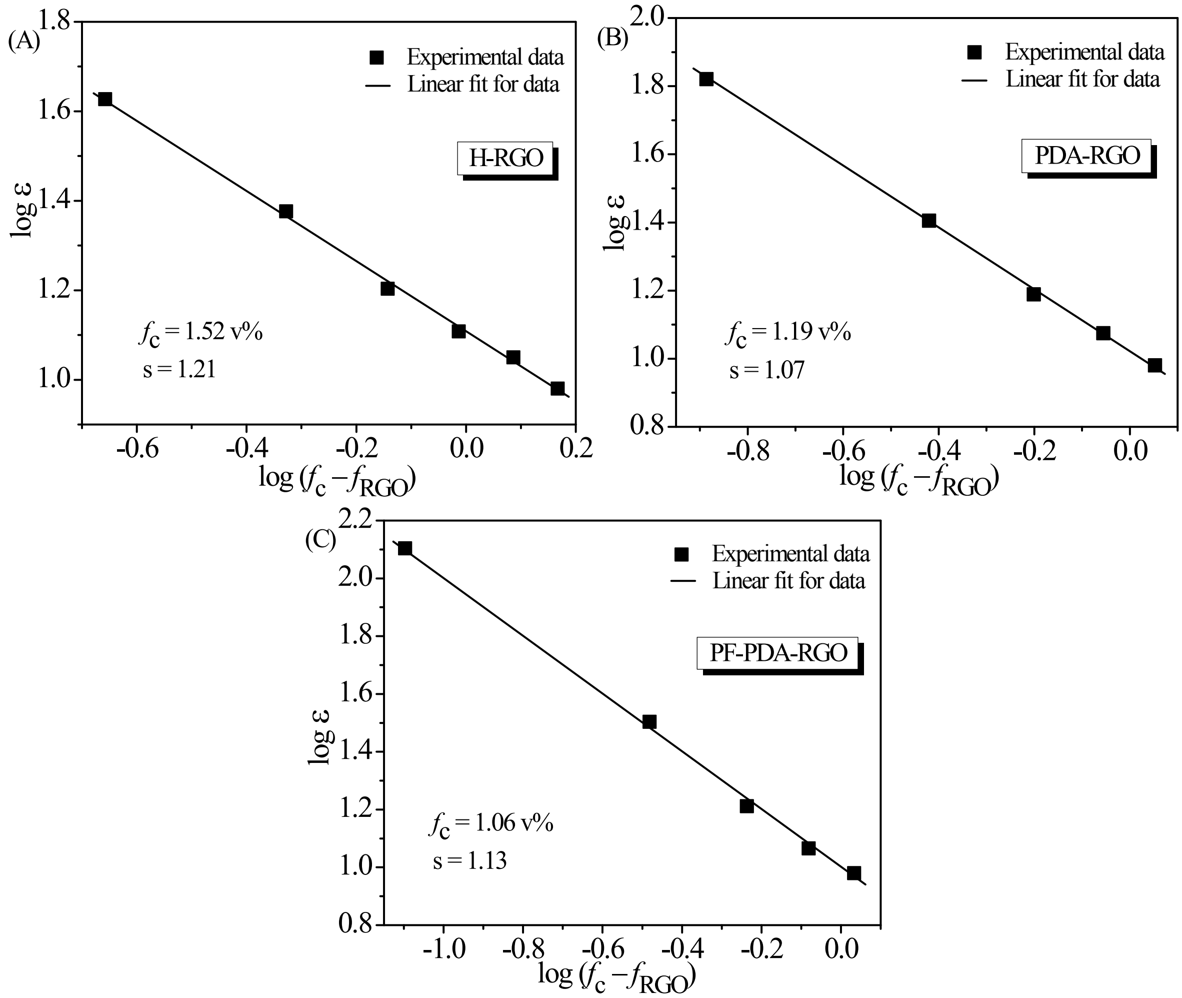

Fig. S4 The determination of percolation threshold of P(VDF-HFP)/RGO nanocomposites filled with different RGO nanosheets: (A) H-RGO, (B) PDA-RGO, and (C) PF-PDA-RGO. The results were calculated based on the best linear fit for experimental dielectric constant data acquired at 1 kHz.

Sample	P(VDF-HFP)/H-RGO				P(VDF-HFP)/PDA-RGO				P(VDF-HFP)/PF-PDA-RGO			
Number	1	2	3	error	1	2	3	error	1	2	3	error
@10^{-1} Hz	38.6	39.9	37.4	±1.3	117.9	115.7	116.8	±1.1	190.8	189.6	189.3	±0.9
@10^{3} Hz	23.8	24.5	23.2	±0.7	65.5	64.1	64.7	±0.7	107.9	106.5	106.6	±0.9
@10^{7} Hz	16.2	16.9	15.4	±0.8	38.8	37.9	38.3	±0.6	65.7	65.2	64.6	±0.6

Table S1. Dielectric constants of P(VDF-HFP)/RGO nanocomposites with 1.0 v% of RGO nanosheets. Three different capacitors for each sample were adopted.

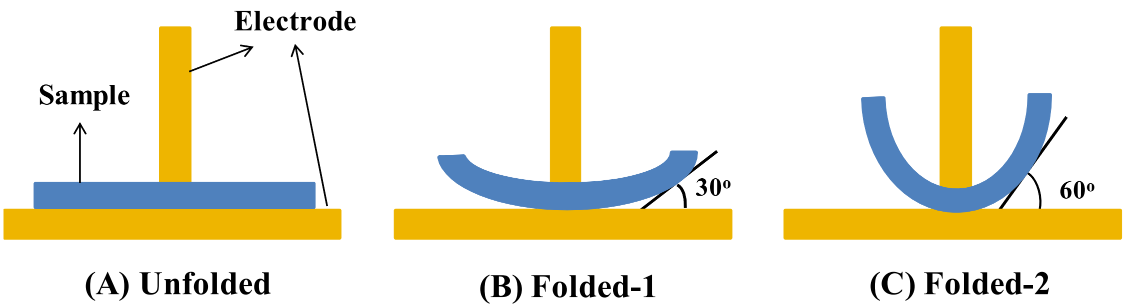

Figure S5. Schematic illustration for the dielectric properties measurement of samples under different folding states: (A) Unfolded, (B) Folded-1 and (C) Folded-2. A layer of gold evaporated on both surfaces to serve as electrodes, the actual electrode size of each sample is 10 mm in diameter.

Sample	P(VDF-HFP)/H-RGO				P(VDF-HFP)/PDA-RGO				P(VDF-HFP)/PF-PDA-RGO			
State	(A)	(B)	(C)	error	(A)	(B)	(C)	error	(A)	(B)	(C)	error
@10^{-1} Hz	38.6	39.1	39.7	±0.6	117.9	118.3	118.8	±0.5	190.8	191.3	191.7	±0.5
@10^{3} Hz	23.8	24.3	24.8	±0.5	65.5	65.8	66.1	±0.3	107.9	108.4	108.8	±0.5
@10^{7} Hz	16.2	16.6	17.1	±0.5	38.8	39.2	39.3	±0.3	65.7	66.2	66.4	±0.4

Table S2. Dielectric constants of P(VDF-HFP)/RGO nanocomposites with 1.0 v% of RGO nanosheets. Three different folding states of each sample were adopted: (A) Unfolded, (B) Folded-1 and (C) Folded-2.