

Electronic Supporting Information

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Toward Highly Compressible Graphene Aerogels of Enhanced Mechanical Performance with Polymer

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1. Materials used in this study

Natural graphite flake (with particle size of 70-80 μm) was obtained from Qingdao Henglide Graphite Co., Ltd. (China). Poly(acrylic acid) (PAA, $M_v = 1,250,000$), and poly(ethylene oxide) (PEO, $M_v = 4,000,000$) were purchased from Sigma-Aldrich Company. Reagent grade phosphorus pentoxide (P_2O_5), potassium persulfate ($\text{K}_2\text{S}_2\text{O}_8$), potassium permanganate (KMnO_4), L-ascorbic acid, sulfuric acid (H_2SO_4 , 98%), hydrochloric acid (HCl, 38%), and hydrogen peroxide (H_2O_2 , 30%) were purchased from Sinopharm Chemical Reagent Beijing Co., Ltd., and were used without further purification. The dialysis tubing used in this study (14K MWCO) was obtained from Jingke Biotechnology Co., Beijing.

2. Preparation of graphene oxide (GO)

Graphite powder (5.0 g) was mixed with P_2O_5 (2.5 g), $\text{K}_2\text{S}_2\text{O}_8$ (2.5 g), and 98% H_2SO_4 (100 mL) under stirring at 80 $^\circ\text{C}$ for 6 h. The pre-oxidized graphite oxide was then obtained by repeated washing and filtering with deionized (DI) water and drying at room temperature under vacuum. The obtained pre-oxidized graphite oxide (5.0 g) was mixed with 98% H_2SO_4 (115 mL) and the mixture was cooled in an ice water bath under stirring. KMnO_4 (15.0 g) was added slowly to maintain the temperature of the mixture lower than 20 $^\circ\text{C}$. The reaction system was transferred to a 35 $^\circ\text{C}$ water bath and heated for another 2 h. After that, 230 mL of DI water was slowly added to maintain the temperature of the mixture less than 98 $^\circ\text{C}$. The mixture was stirred slowly for another 15 min. The reaction was terminated by the addition of ~ 700 mL

of DI water, and then 30 mL of H₂O₂ (30%), which caused the color of the mixture to turn from brown to bright yellow. the mixture was filtered and washed with excess 1:10 HCl aqueous solution (~ 1 L) to remove metal ions followed by washing with DI water until the pH of the dispersion was approximately 5-6. The obtained dispersion was subjected to dialysis to completely remove the unwanted metal ions and acid. It was further concentrated by using vacuum distillation at a temperature lower than 40 °C, and then sonicated for 2 h. The resulting GO aqueous dispersion with a concentration of ~ 8 mg mL⁻¹ was then purified by centrifuging at 4000 rpm (~1500 g) for ~10 min to remove the impurities.

3. The high magnification SEM images of GA, GPA-PAA, and GPA-PEO

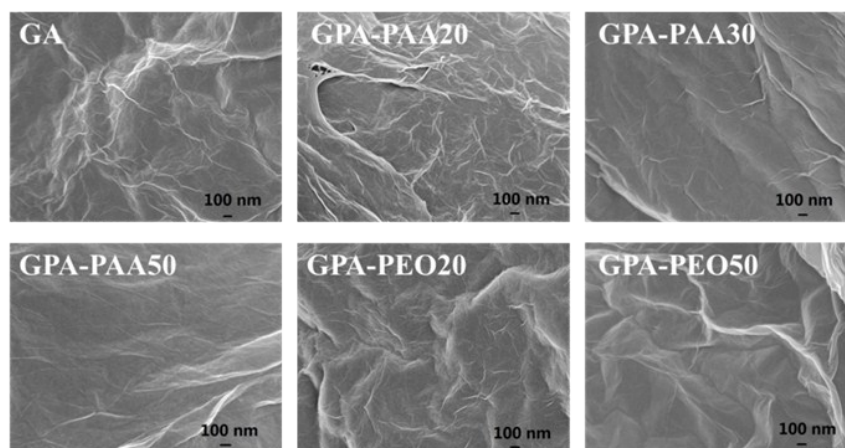


Fig. S1 The high magnification SEM images of GA, GPA-PAA (20, 30, 50), and GPA-PEO (20, 50) (with the magnification of ×30, 000)

4. TGA measurement results of graphene aerogel (GA) and graphene-polymer aerogels (GPAs)

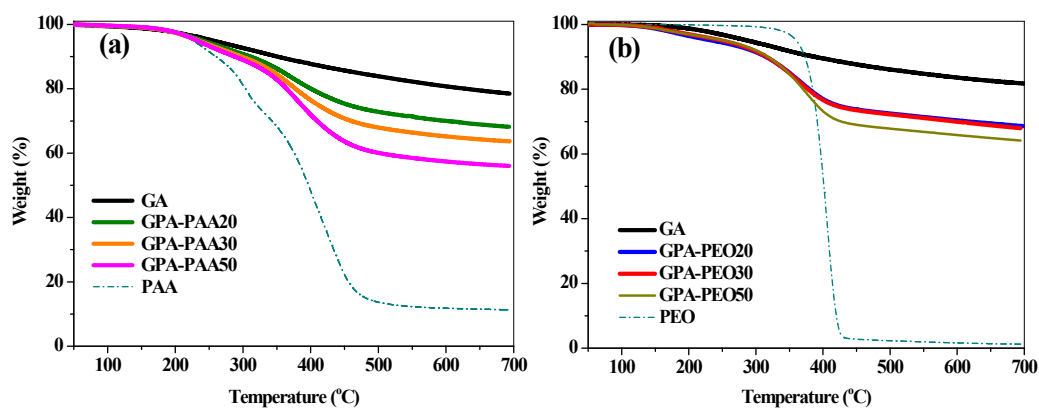


Fig. S2 TGA curves of GA and GPAs

5. Compressibility of GPAs

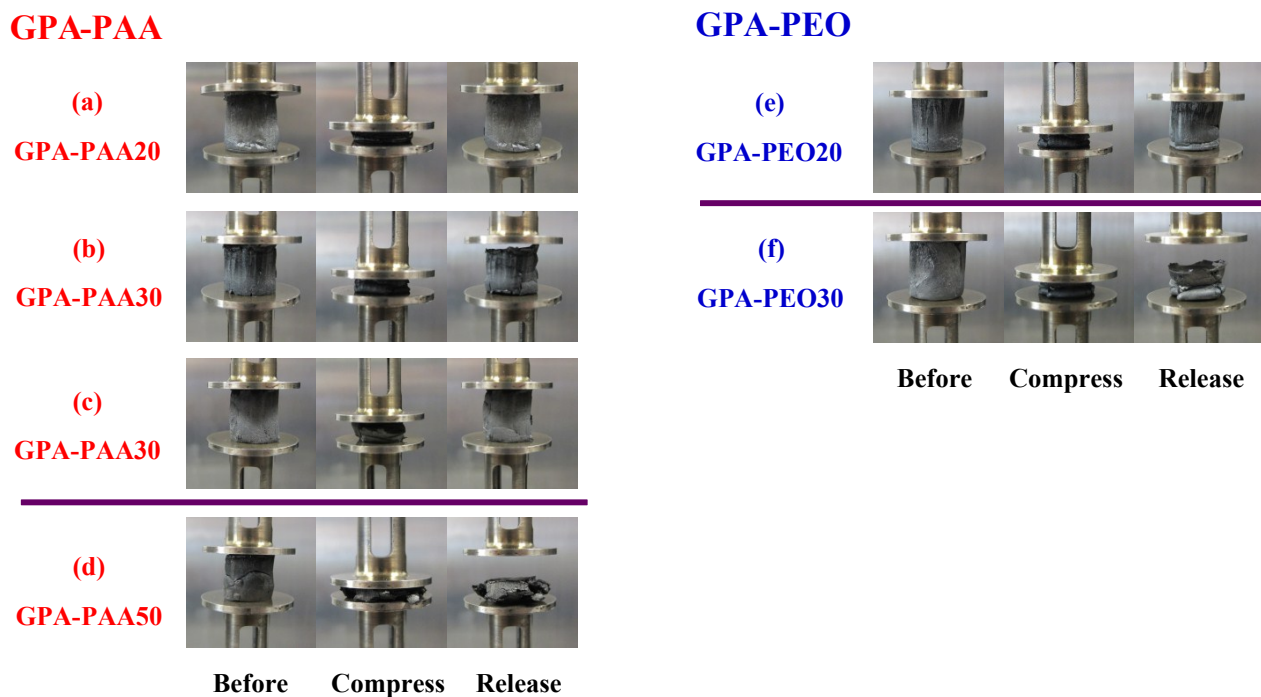


Fig. S3 Digital images showing the compressibility of (a) GPA-PAA20 (10th cycle, 80%), (b) GPA-PAA30 (10th cycle, 80%), (c) GPA-PAA30 (10th cycle, 70%), (d) GPA-PAA50 (1st cycle, 80%), (e) GPA-PEO20 (10th cycle, 80%), and (f) GPA-PEO30 (1st cycle, 80%).

6. DSC measurement results of GA and GPA-PEO

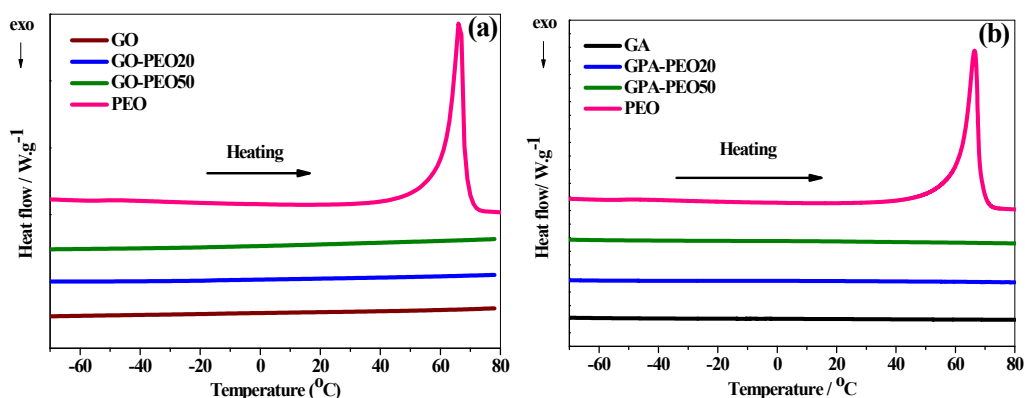


Fig. S4 DSC traces for (a) GO, neat PEO, GO-PEO20, and GO-PEO50; (b) GA, neat PEO, GPA-PEO20, and GPA-PEO50

7. Raman measurement results of GA and GPA-PAA

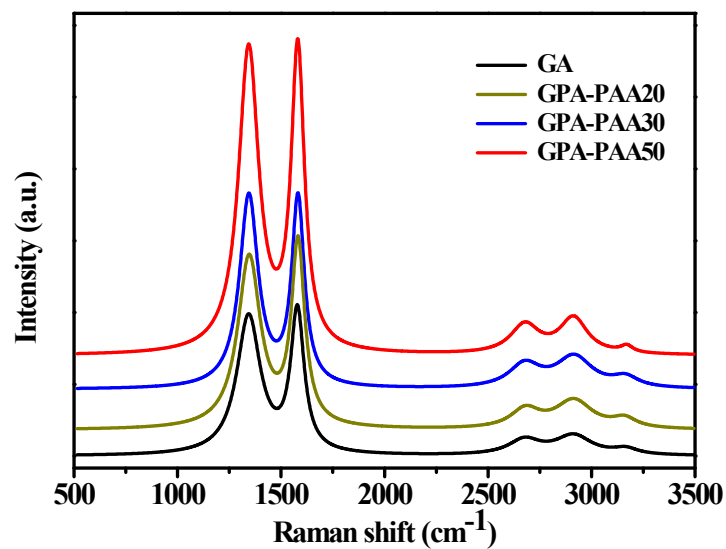


Fig. S5 Raman spectra at 532 nm excitation of GA, GPA-PAA20, GPA-PAA30, and GPA-PAA50

Table S1 Raman band parameters of GA and GPAs.

Samples	Polymer content (wt %)	I _D /I _G	FWHM _G
GA	0	0.94	83.6 ± 0.4
GPA-PAA20	19	0.92	78.4 ± 1.0
GPA-PAA30	32	1.00	75.1 ± 1.5
GPA-PAA50	52	0.98	75.6 ± 2.1

Noted: FWHM_G were acquired by Lorentzian fit curves for the G and D peaks in Raman spectra of GA and GPAs