Electronic Supplementary Information for:

Strain-Induced Delamination of Edge-Grafted Graphite

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Experimental

Materials. All reagents and solvents were purchased from Aldrich Chemical Inc. and used as received, unless otherwise specified. The graphite (Aldrich Cat#: 7782-42-5, type: powder, particle size: < 45μm, purity: 99.99+%) was purchased from Aldrich Chemical Inc. The monomer, 4-phenoxybenzoic acid, was purified by recrystallization from a toluene/heptane (50/50, v/v) mixture solvent (mp 162-164 °C).

Instrumentation. Elemental analysis (EA) was performed with a CE Instruments EA1110. The field emission scanning electron microscope (FE-SEM) used in this work was a LEO 1530FE. The field emission transmission electron microscope (FE-TEM) employed in this work was a FEI Tecnai G2 F30 S-Twin with an operating voltage of 200 kV. Dispersed PEK-g-graphite sample solution in ethanol was prepared. Then, sample on TEM copper grids were prepared by dipping into the solution and taken out to dry. The surface area was measured by nitrogen adsorption-desorption isotherms using the Brunauer-Emmett-Teller (BET) method using Micromeritics ASAP 2504N. Wide-angle X-ray diffraction (WAXD) powder patterns were recorded with a Rigaku RU-200 diffractometer using Ni-filtered Cu Kα radiation (40 kV, 100 mA, λ = 0.15418 nm).

Poly(ether-ketone, PEK) grafted graphite. A reaction flask equipped with a high-torque mechanical stirrer, nitrogen inlet and outlet was charged with 4-phenoxybenzoic acid (4.5 g, 21.0

mmol), graphite (0.5 g), PPA (83% P_2O_5 assay: 100 g) and P_2O_5 (25 g). The reaction mixture was stirred under a dry nitrogen purge at 130 °C for 72h. Some portion of the reaction mixture was poured into distilled water to form a light yellowish-tan-brown curly wire. The other portion was divided into four portions and placed in coagulation setup. All samples prepared were then Soxhlet-extracted with water for three days to remove residual reaction mixture and then with methanol for three more days to remove low molar mass impurities, if any. Finally, the samples were freeze-dried under reduced pressure (0.05 mmHg) at -120 °C for 72h to give a quantitative yield of the product. Anal. Calcd. for $C_{14.98}H_8O_2$: C, 81.78%; H, 3.67%. Found: C, 80.61%; H, 3.69%.

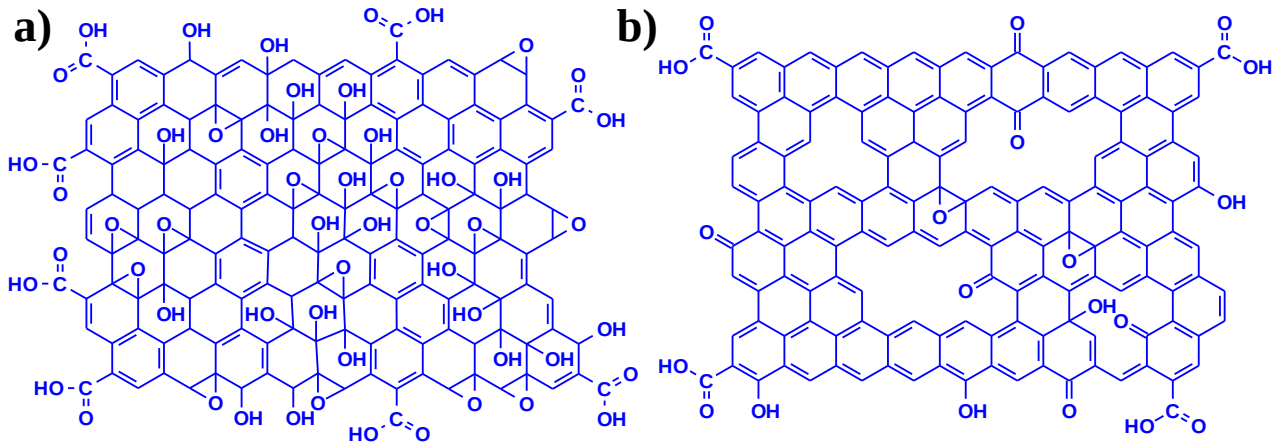

Fig. S1 Proposed structures: (a) graphene oxide (GO) reported by Lerf, *et al.*[1]. (b) reduced graphene oxide (rGO) reported by Gao, *et al.*[2]

[1] A. Lerf, H. He, M. Forster, J. Klinowski, *J. Phys. Chem. B* **1998**, *102*, 4477-4482.
[2] W. Gao, L. B. Alemany, L. Ci, P. M. Ajayan, *Nat. Chem.* **2009**, *1*, 403-408.

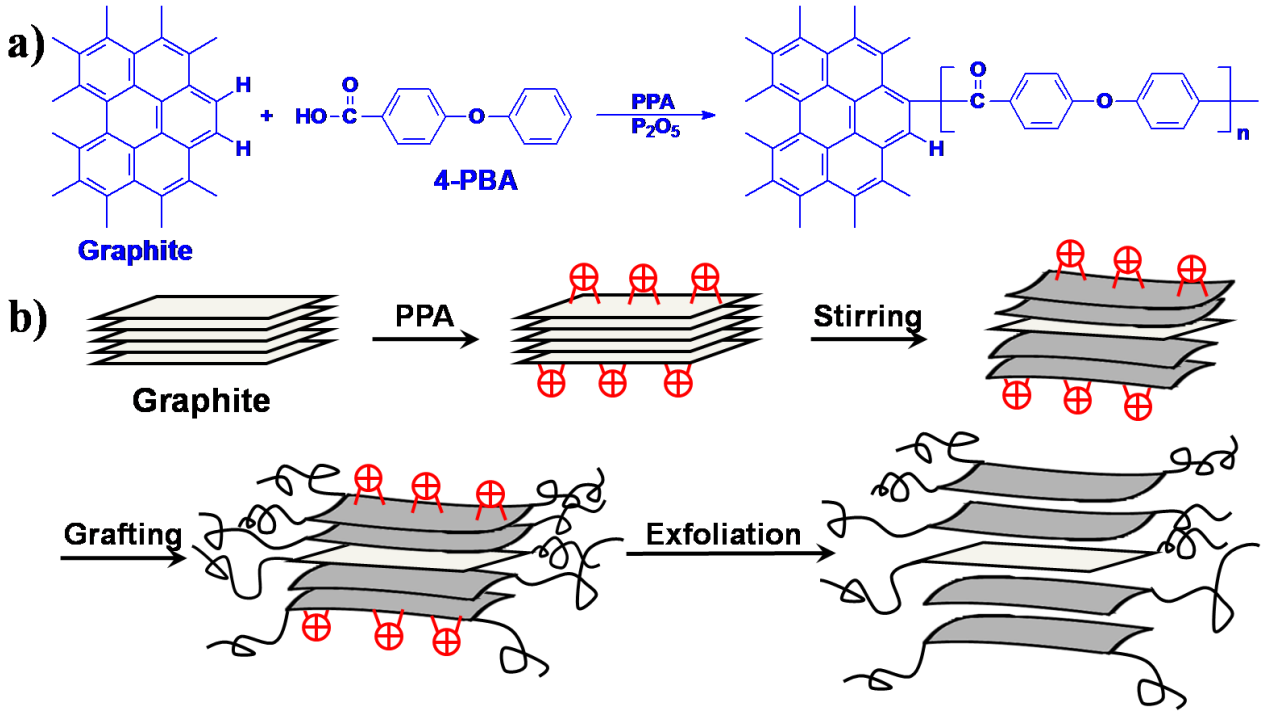

Fig. S2 (a) The reaction between pristine graphite and 4-phenoxybenzoic acid (4-PBA) in polyphosphoric acid (PPA)/phosphorous pentoxide (P_2O_5). The structure of graphite is simplified for the reason of clarity and the edges of graphite consist of sp^2C-H bonds, which is eligible for Friedel-Crafts acylation reaction. (b) The schematic presentation for the edge-grafting of PEK at the edges of graphite by a "direct" Friedel-Crafts acylation between the acylium ion (Ph-C^+=O) of 4-PBA and edge sp^2C-H sites of graphite to produce PEK-g-graphite.

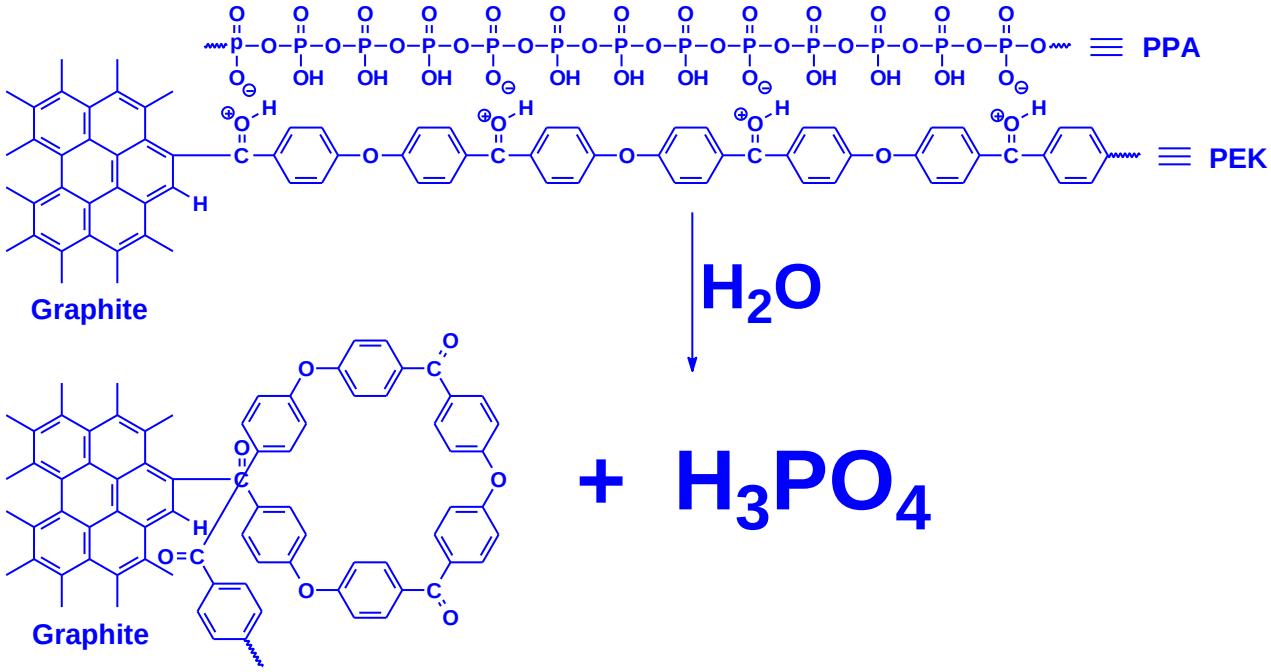

Fig. S3 Conformation changes of PEK-g-graphite before and after coagulation by air moisture. The PEK-g-graphite was homogeneously dispersed in PPA/P_2O_5 medium before coagulation. The homogeneous reaction mixture was phase-separated and coagulated by water (air moisture) as poor solvent.

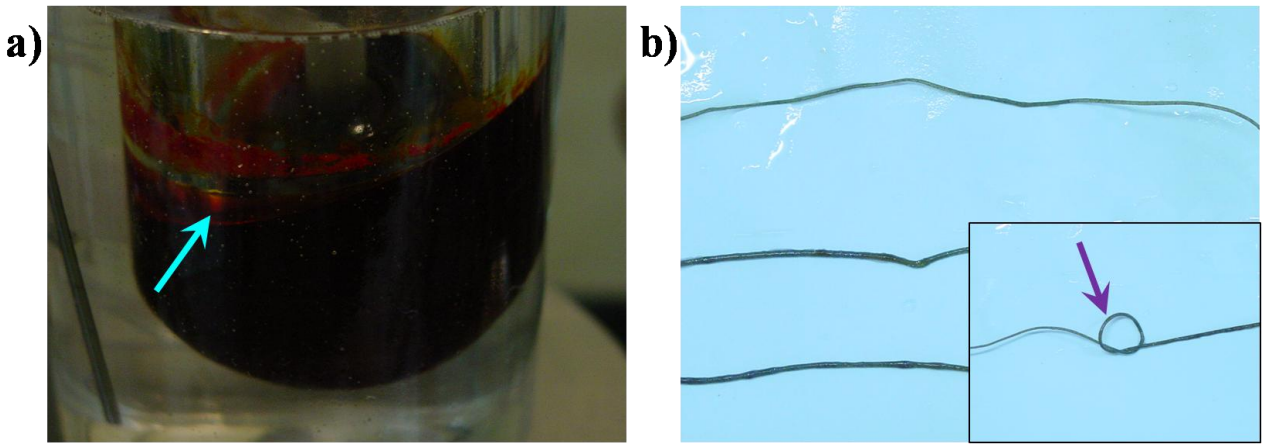

Fig. S4 Digital photographs: (a) reaction flask showing a shiny deep-red transparent color indicates the homogeneous dispersion of PEK-g-graphite into the reaction medium. (b) Fast coagulated PEK-g-graphite wire, after the homogeneous mixture was directly poured into distilled water. Inset is a tied wire demonstrating mechanical integrity, after the reaction medium PPA/P_2O_5 was soaked out into water leaving porous PEK-g-graphite.

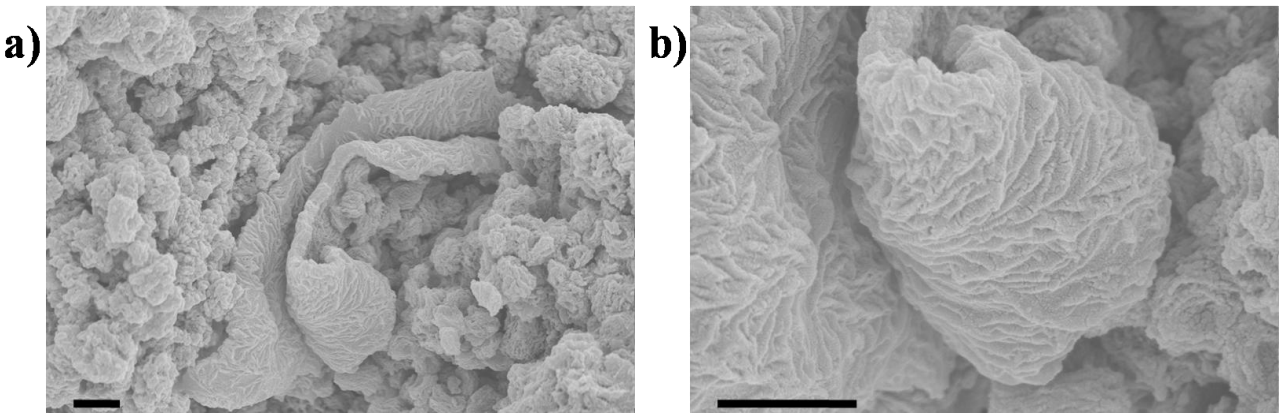

Fig. S5 SEM images of PEK-g-graphite coagulated at 60 °C: (a) at ×10 000. (b) ×30 000. Scale bars are 400 nm.

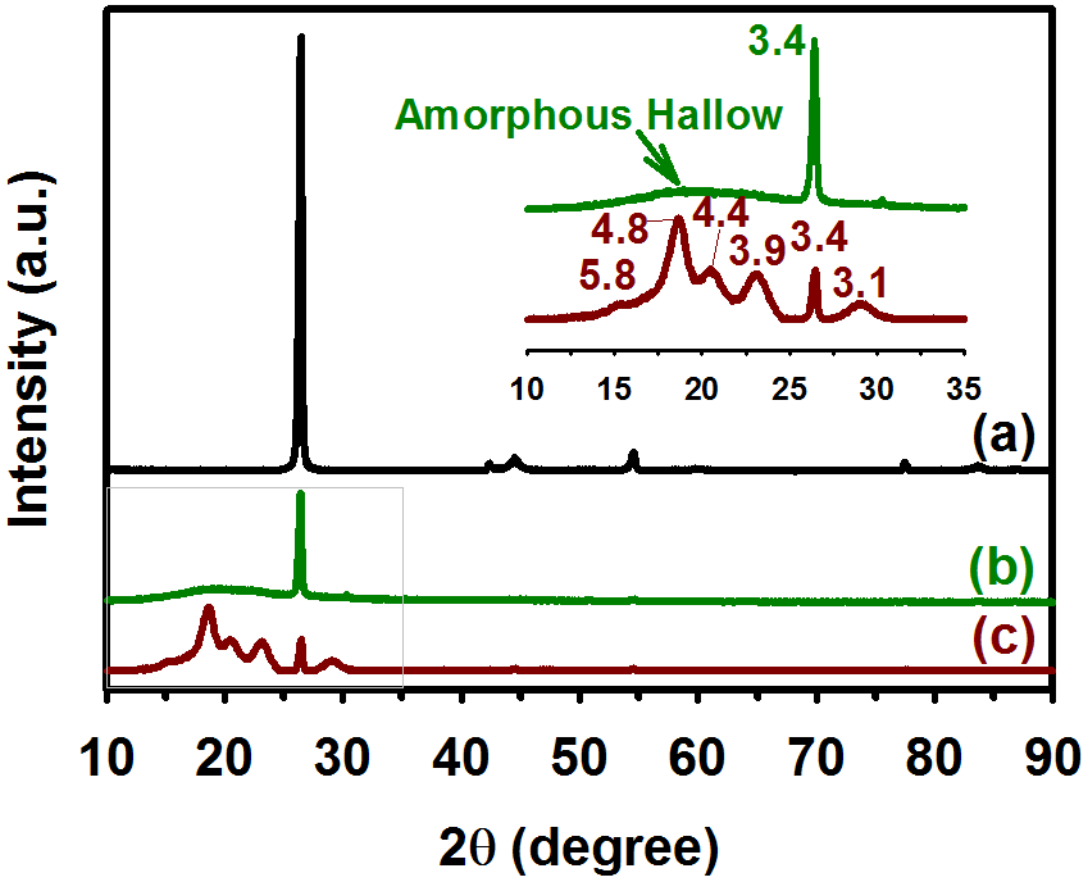

Figure S6 XRD diffraction patterns: (a) pristine graphite; (b) *p*PEK-g-graphite after fast coagulation; (c) *p*PEK-g-graphite controlled coagulation at 25 °C for 48h. Inset is a magnification of the gray rectangle area.

Table S1. BET surface areas, pore volumes and pore sizes of pristine graphite and PEK-g-graphite as a function of coagulation rate

Sample	Surface Area (m^2/g)	Pore Volume (ml/g)	Pore Size (nm)
Pristine Graphite	2.5	0.000019	38.2
PEK-g-graphite (fast)	30.9	0.002756	16.2
PEK-g-graphite (80 °C)	68.2	0.000110	7.5
PEK-g-graphite (40 °C)	96.6	0.005515	21.7
PEK-g-graphite (25 °C)	149.6	0.003784	24.6