Supporting Information

Synthesis of Graphene by Low-temperature Exfoliation and Reduction of Graphite Oxide under Ambient Atmosphere

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Materials

Fuming HNO₃ (65-68%), concentrated H₂SO₄ (95-98%), concentrated HCl (36-38%) were obtained from Sinopharm Chemical Reagent. KClO₃ was bought from Aladdin Reagent. All the materials are AR and used without further treatment except concentrated HCl was diluted to 10%. Natural graphite with a mean size of 35µm was kindly supplied by Qingdao Huatai Lubricant Sealing S&T. The PS pellets were provided by Shanghai SECCO Petrochemical Company Limited (China) and dried at 80 °C under vacuum before use.

Preparation of GO

The Staudenmaier method was utilized to oxidize natural graphite for the synthesis of graphite oxide. In a typical procedure, a 500 mL flask was immersed in an ice bath and purged with nitrogen. Then, 175 mL of concentrated H₂SO₄ and 90mL of fuming HNO₃ were mixed in the flask and stirred for 5 min to release the heat. Then 10g natural graphite was added. After the forming of uniform graphite dispersion, 110 g KClO₃ was added slowly to avoid the danger of explosion. The reaction lasted for the following 120 h at room temperature to achieve sufficient oxidation of graphite. After reaction, the slurry resultants were filtered and washed with 10% HCl solution to remove the sulfuric ions. The production was dried in an oven at 80°C for 3d and kept in a desiccator before use.

Exfoliation and reduction of GO

Graphene sheets (G-130) were produced from GO by thermal treatment at low temperature. A small GO bulk was placed in a flask pre-heated at 130 °C in ambient atmosphere for ~ 8 min. Explosion and dramatic volume expansion of GO accompanied by orange flash flame were observed. When visible expansion of GO stopped, the flask was cooled to room temperature.

As previously reported, the rapid decomposition of oxygen groups always occurs above 150-200 °C. However, the exfoliation temperature was decreased to 130 °C with the assistance of inner pressure generated by HCl in this study. To prove that such exfoliation can occur at 130 °C, the TG analysis was conducted and the result is shown in Fig. S2. The heating rate was kept at 1 °C/min to avoid exfoliating the GO during the scan. As shown in Fig. S2, we can see that there was a slight mass loss at ~ 130 °C, which indicating the initial decomposition of oxygen-containing groups in the GO at this temperature.
Since the reduction of GO is violent exothermic due to its highly energetic nature and the thermal diffusivity of GO is low, the heat released from the initial decomposition of GO at 130 °C would heat the inner GO, leading to the increased temperature (> 130 °C) at the inside of bulk GO. After a few minutes, we inferred that continuous cycle of this process would cause the inner temperature rising to the onset of rapid decomposition of oxygen-containing groups in the GO. That’s why the successful exfoliation of GO with the aid of HCl at 130 °C was achieved after ~ 8 min.

**Preparation of PS/LTEG composites**

PS/LTEG composites were prepared by solution blending. The obtained G-130 was dispersed in DMF by ultrasonication for 15 min. PS pellets were dried at 80 °C for 12 h, followed by dissolution in DMF by vigorous stirring. The G-130 dispersion and the PS solution were then mixed together and stirred for 1 more hour. After that, the above mixture was coagulated with methanol under vigorous stirring. The black solid products (PS/G-130) were filtered, washed with abundant methanol, and dried successively in a vacuum oven at 120 °C for 24 h. Finally, PS/G-130 composites were compression-molded to 1 mm thick plates using a hot-press at 180 °C under a pressure of 15 MPa for 10 min.

**Characterizations**

The diffraction behavior of the samples was studied using a Bruker AXS X-ray diffractometer with CuKa radiation at a generator voltage of 40 kV and a generator current of 40 mA. Scanning electron microscopy (SEM) observation was performed with a Hitachi S-4800 field emission SEM at an accelerating voltage of 4 kV. Transmission electron microscopy (TEM) was conducted on a Tecnai G2 F20 transmission electron microscope with an accelerating voltage of 100 kV. Typical tapping-mode atomic-force microscopy (AFM) measurements were performed using Multimode SPM from Digital Instruments with a Nanoscope V Controller from Veeco. Specific surface area analysis was conducted with a Micromeritics ASAP 2010 Analyzer (Norcross, GA) in terms of the Brunauer, Emmett and Teller (BET) method using nitrogen adsorption. XPS analysis was carried out a Kratos AXIS ULTRA Multifunctional X-ray Photoelectron Spectroscope using Al (mono) Kα radiation (1488.6 eV) under 1.2×10⁻⁹ Torron. Raman spectra were excited with a laser of 633 nm and record with Labram spectrometer (Super LabRam II system). The volume conductivity of the moderately conductive samples (>1×10⁻⁶ S/m) was measured using a standard four-probe method on a Physical Property Measurement System (Quantum Design, US). The samples with low conductivities (≤1×10⁻⁶ S/m) were measured with a three-terminal fixture on an EST121 ultrahigh resistance and micro current meter (Beijing EST Science & Technology CO. Ltd.) according to ASTM D257. Circular plates with 7 cm in diameter were fabricated for conductivity measurements. The sample surfaces were coated with silver paste to reduce contact resistance between the sample and the electrodes.
Fig. S1 XPS curves of GO and G-130.

Fig. S2 The TGA curve of GO with a heat rate of 1 °C/min in the N\textsubscript{2} atmosphere.
Fig. S3 The images of GO exfoliation process. Explosion and violent volume expansion was triggered by simple thermal treatment at 130°C in air. The exfoliation was rapidly finished over the entire films in about 0.3s.

Fig. S4 Electrical conductivity of the polystyrene/G-130 composites as a function of filler volume fraction. Right inset, log $\sigma_c$ plotted against log $(\phi-\phi_c)$, where $\phi_c$ is the percolation threshold.
Fig. S5 SEM images of G-160, G-180, and G-200.

<table>
<thead>
<tr>
<th>Sample</th>
<th>C/O Ratio</th>
<th>C=C/C-C (eV) %</th>
<th>Localized alternant hydrocarbon</th>
<th>C-O/C-O-C (eV) %</th>
<th>C=O/O-C=O (eV) %</th>
</tr>
</thead>
<tbody>
<tr>
<td>G-130</td>
<td>7.4</td>
<td>284.6 (65.0%)</td>
<td>285.3 (14.3%)</td>
<td>286.3 (14.2%)</td>
<td>288.0 (6.5%)</td>
</tr>
<tr>
<td>GO</td>
<td>2.8</td>
<td>284.6 (54.8%)</td>
<td>—</td>
<td>286.5 (38.1%)</td>
<td>288.6 (7.1%)</td>
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