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Facile synthesis of graphene sheets for heat sink application

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ABSTRACT

A mechanical cleavage (MC) approach has been demonstrated to synthesize graphene nanosheets (GNs) as heat sink materials from artificial graphite paper (GP). The facile MC method is composed of three main steps: GP isolation, GP exfoliation, and GN collection. The method is capable of preparing few layers of GNs repeatedly without using chemical oxidizing agents and costly deposition apparatus. The as-prepared GN powders are well characterized by X-ray diffraction and Raman spectroscopy. On the basis of the experimental results, the MC method shows a great feasibility to synthesize high-quality GN products with high repeatability and environmental friendliness. We also report that the addition of GN onto Cu foil induces an improved capability for heat dissipation, as compared with original GP and Cu heat foil. According to the calculations of Fourier's law, the thermal conductivity of the GN/Cu composite heat sink can reach as high as 2142 W/m K, leading to 26% increase of thermal conductivity compared to the GP heat sink.

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1. Introduction

Graphene nanosheet (GN), a single or few layers of graphite, has become a rising star due to its two-dimensional (2D) honeycomb lattice [1–3]. The unique 2D architecture exhibits excellent electrical conductivity, high optical transparency, large specific surface area, good thermal stability, high thermal conductivity, and superior mechanical strength [1,4]. Based on the fascinating properties, a number of research groups have devoted themselves to apply GNs in a variety of technological applications including catalyst support [5,6], lithium-ion battery [7–9], solar cell [10,11], electrochemical capacitor [12], transparent conducting film [13], and so on. Although, some progress has been made in recent years, the reliable fabrication of large-area GNs still remains a challenge. Some of the fabrication methods that are being explored for mass production of GNs consist of chemical oxidation of graphite powders (e.g., Hummers' method) [14], growth by chemical vapor deposition (CVD) [15], sublimation of Si from SiC [16], and solution-phase exfoliation of graphite [17]. One traditional method, Scotch tape method, is also capable of producing individual sheets of graphene with dimensions approaching several hundred micrometers [18]. This approach shows a great potential for scalability, low thermal budget, inexpensive raw materials, and good compatibility with additive coating techniques. However, there are few reports focusing on large-scale production of GNs by using facile mechanical cleavage method.

Herein the present work utilizes a mechanical cleavage (MC) approach to produce exfoliated GNs, prepared from artificial graphite paper (GP). The MC route to fabricate GNs could be performed at ambient temperature without any chemical oxidizing agents. The schematic diagram for describing the MC method, as illustrated in Fig. 1, is composed of the following steps: (i) polymeric coating (e.g., polyethylene terephthalate (PET)) onto GP, (ii) isolation of PET-coated GP by cutting tool, (iii) vertical GNs from GP exfoliation, (iv) collection of GNs from tape, and (v) repeated GN isolation procedure. The exfoliated GNs could be prepared as GN suspension in organic solvents (e.g., N-methyl-2-pyrrolidone (NMP) and acetone) after appropriate ultrasonic bathing. The GN suspension shows uniform dispersion without any precipitate sediment for 10 days in a row. This sheds some lights on how to design an efficient method of obtaining high-quality GNs by using MC approach.

Theoretically, thermal conductivity of graphene falls into the region of 4840–5300 W/m K based on the analysis of shift in the Raman *G* peak with increasing incident laser power [19,20]. Such high magnitude of GNs is much higher than that of Cu (i.e., 385 W/

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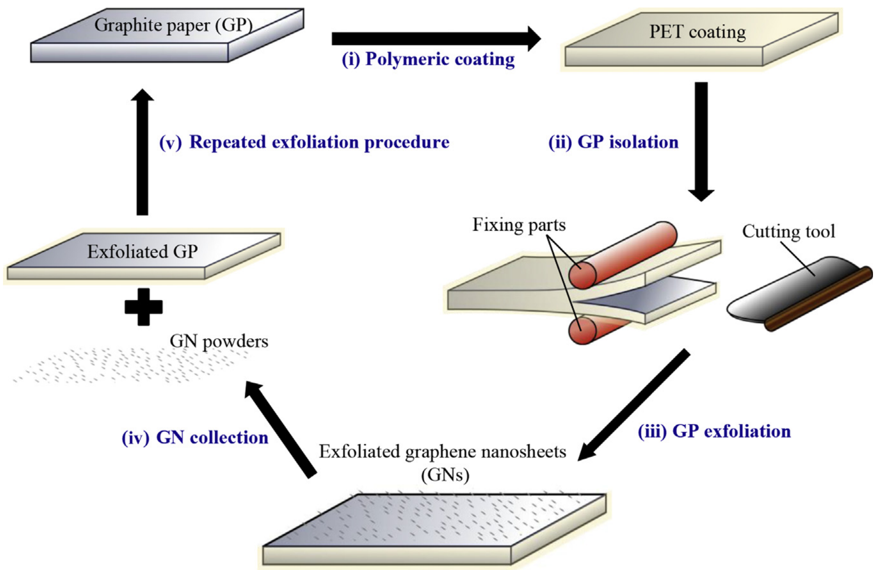

Fig. 1. The schematic diagram of the MC approach for growing graphene sheets, consisting of (i) polymeric coating, (ii) GP isolation, (iii) GP exfoliation, (iv) GN collection, and (v) repeated isolation procedure.

m K) [21] and multi-layered carbon nanotubes (i.e., 650–830 W/m K) [22]. Accordingly, the GNs are promising heat sink materials that can be used in various electronic applications, e.g., motherboard and chip. To exploit the effect of GNs, we introduce the GN coating layer on Cu substrate, forming a GN/Cu heat sink. On the basis of the experimental results, the GN/Cu heat sink delivers superior thermal releasing ability with high thermal conductivity, as compared with Cu and GP layers.

2. Experimental

2.1. Formation of exfoliation graphene nanosheets

The MC approach concerning the fabrication of GNs (see Fig. 1) could be briefly described in the following section. Herein an artificial GP, which was highly graphitized at 2800 °C, served as matrix. The GP had a thickness of 200 μm and an area of 30 × 30 cm^2. Without any further treatment, the GP was totally coated with PET thin layer, with a thickness of approximately 100 μm. After that, the PET-coated GP substrate was placed into a home-made exfoliation test plane, equipped with two fixing parts on both sides of the test plane. A cutting tool with a unique design (i.e., a polished stainless foil) was employed to exfoliate the GP substrate, otherwise The GP exfoliation procedure would destroy well-ordered sp^2 graphite layer, forming vertical graphite fragments onto the GP substrate. The highly-adhesive tape was applied to collect a large number of graphite fragments. Then, the fragments were dispersed in organic solvent, e.g., NMP and acetone. An ultrasonic bathing technique enabled the well dispersion of GN slurry at 50 °C after 1.5 h. The MC approach could be operated repeatedly to collect more GNs.

2.2. Materials characterization

The micro-structural morphology of as-grown GNs was examined using field-emission scanning electron microscope (FE-SEM, JEOL JSM-5600) and a high-resolution transmission electron microscope (HR-TEM, JEOL, JEM-2100). An X-ray diffraction (XRD, Shimadzu Labx XRD-6000) spectroscope, equipped with Cu-Kα radiation emitter, was used to characterize the crystalline structures of GN samples and GP substrates. The crystalline structure of GN samples was characterized by Raman spectroscopy (Renishaw Micro-Raman spectrometer).

To examine thermal conductivity, the GN powders were first coated over Cu foil (purity: 99.9%) with a thickness of 10 μm. The GN/Cu films were fabricated by mixing GN powders, binder (polyvinylidenefluoride) and conducting media (KS-4) with the weight ratio of 85:10:5. The organic solvent, NMP, was employed to prepare the GN slurry. The mixture was blended with a three-dimensional mixer using zirconium (Zr) balls for 3 h to get well-dispersed slurry. The resultant slurry was uniformly pasted on Cu substrate with a doctor blade, followed by evaporating the solvent, NMP, with a blow dryer. Herein the GN/Cu films were adjusted to control a thickness of *ca*. 100 μm. The prepared GN/Cu films were then dried at 135 °C in a vacuum oven for 12 h and pressed under a pressure of ~200 kg cm^{-2}. The thermal conductivities were measured with infrared (IR) filament thermal-conductivity instrument. The GP, Cu, and GN/Cu samples were carefully cut into an area of 10 × 10 cm^2. One thermal-imaging system was used to detect the temperature distribution of all heat sinks. The measurements were carried out at ambient temperature.

3. Results and discussion

The physical structure of original and exfoliated GP substrates has been investigated using FE-SEM, as shown in Figs. 2 and 3, respectively. The FE-SEM images of Fig. 2(a) and (b) correspond to the original GP with flat graphite sheets. After 1st exfoliation, the resulting GP films are found to have lots of waved graphite sheets (see Fig. 3(a) and (b)). As observed from the top-view of the FE-SEM images, the edge of GP still maintains well-arranged stacking graphite layers, and the treated graphite surface is composed of wrinkled and vertical layers. This result demonstrates that the MC approach is capable of yielding isolated single or few layers of GNs if the broken graphite sheets are well treated using appropriate dispersion methods. Fig. 3(c) and (d) depict FE-SEM images of 2nd exfoliated GP substrates, focusing on the edge- and top-views, respectively. The exfoliated GP substrate is still rough, consisting of a large number of graphite fragments. This finding also reveals that the peeling force by the MC route exceeds the van der Waals bonding force between the face-to-face adhering multilayers. Such

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