Carbon 126 (2018) 319–327

Contents lists available at ScienceDirect

Carbon

journal homepage: www.elsevier.com/locate/carbon

Enhanced thermal conductivity of graphene/polyimide hybrid film via a novel “molecular welding” strategy

Haoliang Li, Sichang Dai, Jie Miao, Xian Wu, Navya Chandrasekharan, Hanxun Qiu**, Junhe Yang*

School of Material Science and Engineering, University of Shanghai for Science and Technology, Shanghai 200093, China

ARTICLE INFO

Article history:
Received 8 July 2017
Received in revised form
10 October 2017
Accepted 14 October 2017
Available online 15 October 2017

ABSTRACT

With the development of high-power electronics and portable devices at a noticeable rate, thermal dissipation becomes challenging and critical to the performance of these devices. It greatly promotes the research and development of thermal dissipation materials. Due to its extremely high in-plane thermal conductivity, graphene have shown great potential in thermal management. In this work, a subtle “molecular welding” strategy was realized for the fabrication of flexible, ultrathin, but highly conductive graphitized-graphene/polyimide (g-GO/PI) hybrid films. PI was applied as a solder to weld the micron-sized graphene platelets up through the covalent bonding and to fill up the voids between the graphene layers. This is believed as the key to the considerable enhancement of the in-plane thermal conductivity of g-GO/PI hybrid films. It was worth noting that an enhancement of 21.9% in the in-plane thermal conductivity was achieved for the “molecular welded” g-GO/PI hybrid film as compared to the pristine g-GO film. The proposed “molecular welding” conception provides not only a promising way to the development of the next-generation graphene-based film for thermal management, but also an effective technique to fabricate functional materials with GO or graphene nanoplatelets in other fields.

© 2017 Elsevier Ltd. All rights reserved.

1. Introduction

Heat dissipation has become a critical issue determining the work efficiency and long-term reliability of high power electronic devices [1,2]. With the development and increasing integration level of microprocessors, the currently used graphite film with the thermal conductivity only about 300 W m^{-1} K^{-1} can hardly afford the sufficient heat removal [3]. Though the commercial thermal conducting papers made by polyimide (PI) exhibits a much higher in-plane thermal conductivity, the poor mechanical properties and the complicated preparation have greatly restricted its application in thermal management [4]. Thus, it is essential to search for a novel thermal conducting materials for the more efficient heat removal from electronic devices.

It is always a question until the discovery of graphene, a typical 2-dimensional structure of carbon atoms. Due to the outstanding mechanical and thermal properties [5,6], especially its extraordinary thermal conductivity, graphene has been regarded as one of the promising alternative material serving as thermal conducting film for heat dissipation. The reported thermal conductivity of suspended single-layer graphene at room temperature reaches $(5.30 \pm 0.48) \times 10^3$ W m^{-1} K^{-1} [7], which is much higher than that of the bulk graphite limit (~2000 W m^{-1} K^{-1}) [8].

Although graphene is a promising alternative material in heat dissipation, the excellent performances of a single-layer graphene sheet cannot be directly applied as a lateral thermal spreader. On one hand, many efforts have been devoted to improve the cross-plane thermal conductivity of polymers or greases matrix, using the exfoliated few-layered graphene flakes [9] or functionalized graphene-metal hybrid particles [10,11] as fillers in the thermal interface materials. On the other hand, as an intermediate in preparing graphene by widely used Hummers method, graphene oxide (GO) is considered as the most accessible precursor to fabricate graphene materials because of its good dispersibility in water and organic solvents [12,13]. Elaborate works so far about the free-standing graphene papers for thermal management made by GO solution has been carried out, while these current researches were mainly focused on the preparation method of graphene oxide film, such as vacuum filtration [14], electro-spray deposition [15], mild

* Corresponding author.
** Corresponding author.
E-mail addresses: hxqiu@usst.edu.cn (H. Qiu), jhyang@usst.edu.cn (J. Yang).

https://doi.org/10.1016/j.carbon.2017.10.044
0008-6223/© 2017 Elsevier Ltd. All rights reserved.

evaporation-induced self-assembly [16], and wet-spinning [17–19] and so on, followed by the high temperature graphitization. Moreover, great efforts have been made to improve the performance of graphene films or fibers by using large-sized GO sheets [18,19]. More recently, Peng and co-workers reported a debris-free graphene film with extremely high thermal conductivity as well as excellent flexibility due to the micro-folds of graphene sheets [20]. The superior performances are attributed to the fact that the graphitization treatment can completely remove the functional groups and restore the defects in GO sheets. However, at the same time, the annealing treatment will give rise to the voids or "air pockets" in GO films, and these macroscopic gaps are irreparable during graphitization, resulting in grain boundaries in graphene films [21–23]. It has been proved that the existence of grain boundaries will lead to a higher extent of phonon scattering and thermal resistances leading to the decline in the thermal and electrical conductivity dramatically [24–26]. An alternative way to reduce the grain boundaries in graphene films is using large sized graphene sheets [22], but efficient methods about how to enlarge the grain sizes of GO and how to fill up the voids in GO films is still a big challenge so far.

Herein, we propose a novel "molecular welding" strategy to fabricate a flexible and free-standing graphene/polyimide (g-GO/PI) hybrid film with enhanced in-plane thermal conductivity. PI, as a solder, is employed to connect the adjacent graphene sheets via covalent bondings and evolved into oriented graphitic structure after the graphitization treatment. The in-plane thermal conductivity of g-GO/PI film achieved a considerable improvement of 21.9% with only 1% of PI addition. The "molecular welding" conception contributing to the enhancement in the thermal conductivity shows not only a promising way to develop the next-generation graphene-based film for thermal management, but also an efficient method to utilize GO or graphene nanosheets as a body material in preparation of functional materials.

2. Experimental

2.1. Preparation of GO and polyamide acid (PAA)

GO was prepared by a modified Hummers method which have been reported in our previous work [27,28]. PAA was synthesized through a traditional solution blending method. Briefly, 5 g pyromellitic dianhydride (PMDA) and 5 g 4, 4′−diaminodiphenyl ether (ODA) were dissolved in 30 mL N, N−Dimethylformamide (DMF), respectively. (All reagents are purchased from Sinopharm Chemical Reagent Co, Ltd., Shanghai, China) Then the PMDA was slowly added to the ODA with the continuously stirring under an atmosphere of N_2 for 4 h at room temperature. The polymer content of the PAA solution was 15%.

2.2. Fabrication of GO/PAA and g-GO/PI film

The preparation of GO/PAA and g-GO/PI film were described as four steps as illustrated in Fig. 1. In a typical process, GO/PAA film was fabricated by an evaporation induced self-assembly method. Concretely, 20 mL GO (5 mg/mL) was dispersed in DMF with ultrasonication. After that, 6.7 μL PAA solution was added into the GO solution and the mixture was transferred into a Teflon plate and self-assembled at 70 °C for 6 h. The as-obtained film was denominated as GO/PAA-1%.

The annealing treatment of GO/PAA films were conducted in a hot-pressing furnace (ZT-40-20Y, Shanghai Chen Hua Electric Furnace Co. Ltd) that were heated to 250 °C at a rate of 2 °C/min for imidization and to 1000 °C at a rate of 5 °C/min for further reduction under nitrogen flow. Finally, the reduced films, named as g-GO/PI-1%, were graphitized at 2800 °C at a rate of 20 °C/min for 45 min with the protection of argon. (KGP-100, Zhuzhou Chenxin Induction Equipment Co. Ltd) In the same manner, we fabricated the GO/PAA-x% and g-GO/PI-x% samples, where x represented the weight percentage of PAA to GO.

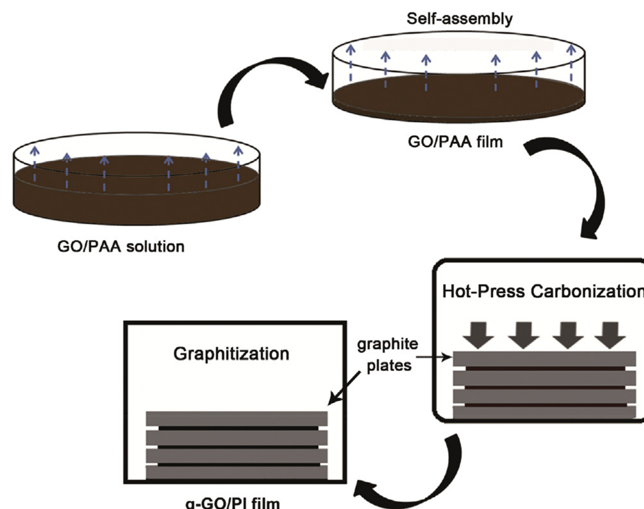

Fig. 1. Schematic procedures of the synthesis of the g-GO/PI composite film. (A colour version of this figure can be viewed online.)

2.3. Characterization

The surface and cross-section morphology of film samples were observed by scanning electron microscopy (SEM) (FEI Quanta FEG, USA) with an accelerating voltage of 20 kV. Energy dispersive spectrometer (EDS, JEOL-2010) was employed to investigate the distribution of polymer in the film. The cross-section of g-GO and g-GO/PI-1% films were carved and observed via focused ion beam (FIB) technology at the tilt angle with 52.5°. X-ray diffraction (XRD) patterns were measured using a Bruker D8 ADVANCE diffractometer with Cu Kα radiation (λ = 0.154 nm) at a generator voltage of 40 kV and a current of 40 mA. Raman spectroscopy was excited using a LabRAM spectrometer (Horiba, LabRAM HR Evolution, France) with a laser of 532 nm. X-ray photoelectron spectroscopy (XPS) measurements were performed using a Physical Electronics PHI-5300 spectrometer with a monochromatic Mg Kα radiation at a power of 250 W and a voltage of 14 kV.

2.4. Measurement of thermal conductivity

The in-plane thermal diffusivity (α) of film samples was measured using a laser flash analyzer (LFA 467/Ins Nanoflash, Netzsch) at room temperature. As shown in Fig. S1, for the measurement of α, an energy pulse (5 J) enabled by a Xenon lamp heats the sample from bottom and the induced temperature rise is determined by the InSb IR detector on the top side. The sample holder has four holes around the center for temperature detection. The width of each hole is much larger than the sample thickness, which enable an in-plane thermal transportation model. All film samples were carved to a diameter of 25 mm for the measurement. The α and the thermal conductivity (κ) was calculated by the following equations:

$\alpha = 0.1388^{*}d^{2*}t_{1/2}^{-1}$ (1), where d was the thickness of sample and $t_{1/2}$ was the half of the diffusion time, respectively.

Download English Version:

https://daneshyari.com/en/article/5431515

Download Persian Version:

https://daneshyari.com/article/5431515

Daneshyari.com