



Extremely high thermal conductivity of nanodiamond-polydopamine/thin-layer graphene composite films

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ABSTRACT

The hybrid composite films containing nanodiamonds (ND) coated with polydopamine (pDA) (ND-pDA) and reduced graphene oxide (rGO) converted from graphene oxide (GO) films are designed for achieving extremely high thermal conductivity. The ND-pDA/rGO films are successfully fabricated using the vacuum-filtration process followed by a heat treatment at 800 °C. The thermal conductivities of the films in the in-plane ($K_{//}$) and through-plane (K_{\perp}) directions are measured by the laser flash method to better understand how the addition of dopamine, the amount of ND-pDA, and the test temperature affect the thermal properties of the hybrid films.

The experimental results show that the addition of dopamine results in dense structure of the ND-pDA/rGO hybrid films, which is favorable for phonon transport, and thus remarkably increase the thermal properties of the film. Additionally, films with higher ND-pDA loading possess lower in-plane but higher through-plane thermal conductivity. $K_{//}$ and K_{\perp} of 1406 and 0.677 W m⁻¹ K⁻¹, respectively, for 20ND-pDA/20rGO measured at 25 °C are achieved. Both the $K_{//}$ and K_{\perp} of 20ND-pDA/20rGO increase with test temperature. Maintaining such high thermal conductivities at high temperature, the hybrid films are believed to be suitable for lightweight thermal management materials with high heat transfer properties in a specific direction.

1. Introduction

Owing to the miniaturization of modern electronic devices, effective thermal management is becoming a key role in preventing devices from overheating. Therefore, fabrication of lightweightness and high thermally conductive thin films has attracted much attention recently. Carbon nanomaterials such as graphene, carbon nanotubes, and nanodiamond possess outstanding properties such as low mass density and high mechanical and thermal properties, which make them promising candidates for many applications [1–6]. Graphene films exhibit anisotropic thermal properties with high theoretical in-plane thermal conductivity ($K_{//}$) of 5000 W m⁻¹ K⁻¹ [7–9], and ultralow through-plane thermal conductivity (K_{\perp}), over two orders of magnitude smaller than $K_{//}$. High $K_{//}$ is attributed to strong sp² or sp³ bonding, which is favorable for lattice vibration, while low K_{\perp} is ascribed to the weak van der Waals force between each graphene layer. Therefore, graphene films with 2D structure can transfer heat in the in-plane direction while impede the phonon transport in the through-plane direction, as a result, protect the beneath electronic devices from overheating. Numerous papers have reported on the topics related to the high thermal anisotropy of graphene paper and applied them to heat-dissipating materials for thermal management [10–23]. Results have shown that the

interfacial thermal resistance of the films is remarkably influenced by the size, orientation, and the degree of crystallinity of the graphene sheets. For example, Kumar et al. prepared graphene oxide (GO) film by vacuum filtration of the GO solution containing variable size GO fabricated by the modified Hummers method. Graphene paper was produced after reduction by hydroiodic acid. The thermal properties measured by laser flash analysis (LFA) revealed that the thermal conductivities of reduced large size GO (around 23 μm²) and reduced small size GO (around 1 μm²) were 1390 W m⁻¹ K⁻¹ and 900 W m⁻¹ K⁻¹, respectively. Heat transfer in solid can be explained by phonon transport, typical phonon mean free path around the room temperature is approximate 800 nm [24,25]. It indicated that the larger sheet size of reduced graphene oxide (rGO) resulted in an increase in the wavelength of acoustic phonons, which is favorable for heat transfer [18]. Song et al. investigated the thermal conductivities of graphene paper fabricated from GO paper after thermal annealing at varied temperature. The graphene paper annealed at 1200 °C exhibited a high thermal conductivity of 1043 W m⁻¹ K⁻¹, while the graphene paper annealed at 800 °C has the thermal conductivity of only 5.1 W m⁻¹ K⁻¹. The study showed that a critical temperature of approximately 1000 °C was required to reconnect the sp² domain and substantially enhance thermal conductivity [16]. Higher annealing temperatures contributed to the

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restoration of sp^2 bonding and the effective removal of oxygen-containing functional groups, significantly lowering the phonon scattering effect and enhancing the in-plane thermal conductivity [20].

Nevertheless, the high thermal anisotropy of graphene paper may restrict its application on thermal management. For example, a heat spreader is required to have better heat conduction in the through-plane direction. To meet the demand for highly thermally conductive thin films with tunable thermal anisotropy, Zhang et al. constructed a 3D bridged carbon nanoring (CNR)/graphene hybrid paper with CNRs covalently bonded to the graphene sheets, enhancing the heat conduction in the through-plane direction. Hence, an improvement in the through-plane thermal conductivity from 2.0 to $5.8 \text{ W m}^{-1} \text{ K}^{-1}$ was observed, while in-plane thermal conductivity decreased from 946 to $890 \text{ W m}^{-1} \text{ K}^{-1}$ because of the scattering points formed by bonding along the in-plane direction; therefore, lowered the thermal anisotropy [26]. Additionally, Hsieh et al. used carbon nanotubes (CNTs) and graphene nanosheets (GNs) to fabricate a CNT/GN composite, and demonstrated that the in-plane and through-plane thermal conduction were contributed to by both GN and CNT, respectively. The dense structure of the CNT/GN composite also reduced the interfacial thermal resistance. Very high in-plane thermal conductivity ($1991 \text{ W m}^{-1} \text{ K}^{-1}$) and through-plane thermal conductivity ($76 \text{ W m}^{-1} \text{ K}^{-1}$) at 323 K were reported [22,27].

In this study, to design rGO-based composite films with high thermal conductivity and tunable thermal anisotropy, we introduced nanodiamond (ND) and polydopamine (pDA) into the rGO-based material system. Nanodiamond provide bridges between the graphene layers, and create more phonon transport paths in the through-direction; as a result, the ration of in-plane/through-plane thermal conductivity can be tunable. In addition, dopamine acted as a binder to enhance the density of the hybrid films while being favorable for phonon propagation. Furthermore, the carbon shell formed by pDA after high temperature annealing enhanced the heat transfer in the through-plane direction.

Because of inherent 2D structure, graphene films exhibit anisotropic thermal properties with high in-plane thermal conductivity and ultralow through-plane thermal conductivity. High in-plane thermal conductivity is attributed to strong sp^2 or sp^3 bonding, which is favorable for lattice vibration which refers to phonon transport. However, oxygen-containing functional groups of graphene impede phonon transport. rGO is obtained after high temperature annealing. The effective removal of oxygen-containing functional groups increase crystallinity and lower the phonon scattering effect. It also enhance phonon transport in graphene layers, as a result, increase the in-plane thermal conductivity. Thus, fabrication of a highly thermally conductive thin films with tunable thermal anisotropy by controlling the amount of ND-pDA can be achieved. The ND-pDA/rGO hybrid films were fabricated using the vacuum filtration method followed by the thermal reduction process under an argon atmosphere (Fig. 1) to form a dense structure with high thermal conductivities and adjustable thermal anisotropy. The thermal properties of the hybrid films in the in-plane and through-plane directions were measured by the LFA method. In addition, the influence of the addition of ND or ND-pDA, ND-pDA loading, and

temperature-related effects on the thermal performance of the composite films were also investigated.

2. Experimental section

2.1. Material preparation

Preparation of GO suspension was made according to the modified Hummers method [28] using natural graphite flakes (Alfa Aesar, 325 mesh, 99.8%) as the raw material. In a typical process, 1.0 g of natural graphite flakes was slowly added to 100 ml of concentrated sulfuric acid (H_2SO_4) solution and then stirred in an ice bath for 1 h, followed by the addition of 4.0 g of potassium permanganate (KMnO_4) with successive stirring for 6 h at 40°C . After the oxidation process, 50 ml of de-ionized water was slowly added to the solution and stirred for 1 h. Subsequently, another 100 ml of de-ionized water was added and the solution was stirred for 30 min. Finally, 40 ml of hydrogen peroxide (H_2O_2 , 35%) was gradually added to the solution under stirring until the color of the mixture changed from brown to bright yellow. The solution was kept overnight, after which the precipitated mixture was washed with hydrochloric acid to eliminate the metal ions and impurities. The mixture was then washed with de-ionized water by centrifugation until the neutral pH was reached. Nanodiamond with a powder size of 100 nm used in this work was purchased from HonWay Co. Ltd.

2.2. Preparation of rGO film, ND/rGO hybrid film, and ND-pDA/rGO hybrid films

A schematic process flow diagram is shown in Fig. 1. To obtain the homogeneous ND-pDA/GO solution, different weights of ND (20, 40, 60, and 80 mg) were dispersed in pH 8.5 tris-buffer solution by sonication for 60 min, followed by the addition of dopamine hydrochloride under stirring for 18 h. After the self-polymerization process, polydopamine-coated ND (ND-pDA) was washed with de-ionized water by centrifugation and then dispersed in ammonia solution. In addition, GO solution (20 mg) sonicated for 1 h was mixed with the ND-pDA solution under stirring for 1 h. The GO film, ND/GO hybrid film, and ND-pDA/rGO hybrid films were fabricated by vacuum filtration of the GO solution, ND/GO solution, and GO/ND-pDA solution, respectively, using membrane filters (Mixed Cellulose Esters, 47 mm in diameter, $0.2 \mu\text{m}$ in pore size, Advantec). After drying, the free-standing films were subjected to a thermal reduction under argon atmosphere at 800°C for 1 h to reduce the graphene oxide. Thereby, rGO film, ND/rGO hybrid film, and ND-pDA/rGO hybrid films were formed. The ND/rGO hybrid film containing 20 mg GO and 20 mg ND was denoted 20ND/20rGO and the ND-pDA/rGO hybrid films with varied amounts of ND were denoted xND-pDA/20rGO where x represents the weight of ND in the hybrid film. A critical issue for the preparation of ND-pDA-rGO is that the polymerization reaction of dopamine occurred in alkaline condition. However, the alkaline condition is not favorable for GO expansion even though with the assistance of sonication, leading to the loose and fragmented pDA-rGO film. This is the reason why we didn't show the thermal conductivity of the pDA-rGO sample.

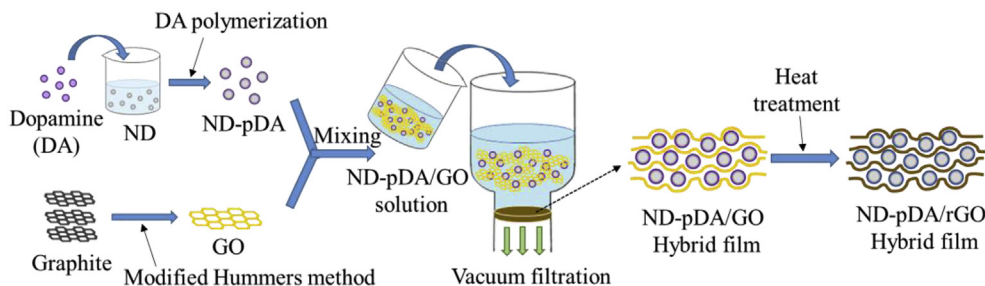


Fig. 1. Schematic figure showing the preparation of ND-pDA/rGO hybrid films.

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