Numerical simulation of thermal conductivity of graphene filled polymer composites

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ABSTRACT

In this work, the thermal behavior of graphene foam (GF) filled polymer composite is investigated using the finite element method. Owing to the interconnected structure of GF, which forms effective heat pathways, GF filled polymer composite is endowed with good thermal properties. The effect of contact thermal resistance, interfacial thermal conductance, as well as GF strut length and diameter on thermal conductivity is simulated and compared with experimental results. It is found that the contact thermal resistance is more important than interfacial thermal conductance in terms of thermal conductivity of composite. The contact thermal resistance between GF filled polymer composite and copper block is about 10^{-4} to $10^{-3} m^2 KW^{-1}$. The shorter length and larger radius of GF struts are beneficial for heat dissipation. The results prove that the GF filled polymer composite is a good candidate material for heat management of electronic devices.

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1. Introduction

Rapidly increasing power densities in electronics make efficient heat removal a crucial issue for progress in information, communication and energy storage technologies [1]. Development of next generation of integrated circuits, three-dimensional integration, and ultrafast high-power density communication devices makes the thermal management demands be urgent and necessary. Thermal interface materials (TIMs) applied between heat source and heat sink, are essential element of thermal management for electronic devices [2]. Conventional TIMs filled with thermally conductive particles require high volume fraction of fillers (~ 50 vol %) to achieve thermal conductivity of ~1—5 W $m^{-1}K^{-1}$ at room temperature [3]. Earlier attempts utilizing highly conductive nanomaterials, for example, carbon nanotubes (CNTs) [4], as fillers in TIMs, have not led to practical applications due to mainly weak thermal coupling at CNTs/matrix interface.

Owing to high thermal conductivity and large surface-to-volume ratio of graphene [5], it has been used as nanofillers to enhance the thermal conductivity of lightweight polymeric composites [6—11]. Conventional methods for preparing graphene sheets filled composites include melt-blending, solution blending and in-situ polymerization [12—16]. However, when graphene is combined into polymer by such approaches, its exceptional properties cannot be fully exploited. This is because the strong π-π interactions trigger agglomeration or restacking of the graphene sheets, leading to their poor dispersion in the matrix. Fortunately, three-dimensional graphene foam (GF) was fabricated by Chen et al. [17], using chemical vapor deposition (CVD) technique. In the GF filled polymer composite, the unique interconnected structure of GF makes heat transfer easier along the GF struts in one hand, and also avoids the dispersion problem of graphene sheets in other hand. Pettes et al. [18] found that the thermal conductivity of GF consisting of few-layers of graphene and ultrathin graphite was from 0.26 to 1.70 W m^{-1} K^{-1} depending on synthetic conditions. Ji et al. [19] demonstrated that loading a paraffin wax phase change material with annealed ultrathin-graphite foam can significantly increase thermal conductivity by as much as a factor of 18. Zhang et al. [20] reported that thermal interfacial resistance of GF at the Si—Al interface was as low as 0.04 cm² K W^{-1}, which is an order of magnitude lower than those achieved with conventional thermal grease and thermal paste based TIMs.

As a new composite, graphene foam filled polymer composite has attracted widespread attention. Because of interconnected network of GF, the effect of interfacial thermal resistance between

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GF and polymer matrix on thermal conductivity may be greatly reduced in the co-continuous composite and the thermal conductivity enhancement seems to be much better than that of particle or fiber filled polymer composites. However, experimental results are not as satisfactory as expected [21–23].

In this work, a finite element model is developed to predict the thermal conductivity of GF filled polymer composite. The effect of GF size, interfacial and contact resistance on the thermal conductivity is investigated. The numerical results are compared with experimental ones.

2. Finite element model

Fig. 1a shows the morphology of graphene foam according to our previous work [21,22]. Experimental details is shown in supported materials. Three dimensional interconnected structure of GF is well shown, of which the single cell diameter is several hundred micrometers, hollow strut length is 300—600 μm, and effective radius (the radius of circumcircle of triangular cross-section) is about 50 μm. By using equation proposed in Ref. [24], the void density of GF is calculated to be about 4.8×10^3voids/cm^3. Fig. 1b shows a fractured surface of GF filled polydimethylsiloxane (PDMS) composite. It reveals that the PDMS penetrated into not only the space of interconnected GF network, but also the interior of GF struts.

In order to numerically simulate the GF/PDMS composite, a representative volume element (RVE) is constructed as given in Fig. 2 with its geometry approximating that of real composite. The side length of cubic RVE is L. The temperatures at the top and bottom surfaces are set to be T_t and T_0, respectively.

The RVE was meshed and imported in the commercial finite element software ABAQUS. Two material sections are defined for GF and PDMS matrix. In the model, the following assumptions are made:

1. The heat conduction is one dimensional (1D);
2. The thin shell element is used to simulate graphene sheets composing GF;
3. The solid element is used to simulate isotropic PDMS matrix;
4. The thermal conductivity is independent of temperature.

The thermal conductivities of PDMS and graphene sheets are listed in Table 1 according to the literature [18]. PDMS is isotropic, while few-layer graphene sheet is transversely isotropic. The in-plane thermal conductivity of graphene sheets is much higher than the out-of-plane one. In the plane, covalent sp2 bonds between adjacent carbon atoms are among the strongest in nature. By contrast, the adjacent graphene sheets (out-of- plane direction) within a graphite crystal are linked by weak van der Waals
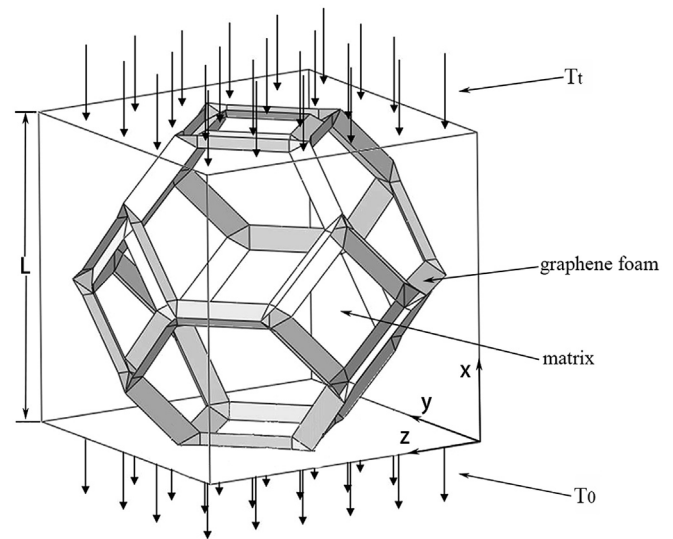

Fig. 2. A representative volume element (RVE) for modeling the composite.

Table 1
Thermal conductivity of graphene sheets and PDMS matrix.

Constituents	Symbol	Wm^{-1}K^{-1}
Graphene sheets [27]	$k_{11} = k_{22}$	596
	k_{33}	6
PDMS [21]	k_{PDMS}	0.2

interactions [25]. The GF is consisted of a large number of graphene sheets connected by nodes.

Considering the transverse isotropy of graphene sheets, two in-plane directions are defined as 1 and 2, while the out-of-plane direction as n, as shown in Fig. 3. Each GF strut consists of three pieces of large graphene sheets. However, some struts of GF have defects due to the hydrochloric acid etching of Ni and incomplete growth of graphene. Through those defects such as microcracks, the liquid PDMS matrix can infiltrate into hollow struts to form solid composite.

3. Calculation of effective thermal conductivity

Thermal conductivity of composite sample (diameter: 30 mm, thickness: 1.5 mm) was measured by the heat flux method (DRL-III, Hunan Xiangyi) in accordance to ASTM D5470 [22]. Steady-state measurement is a one-dimensional testing method for which the sample is placed between two co-planar blocks, just as shown in
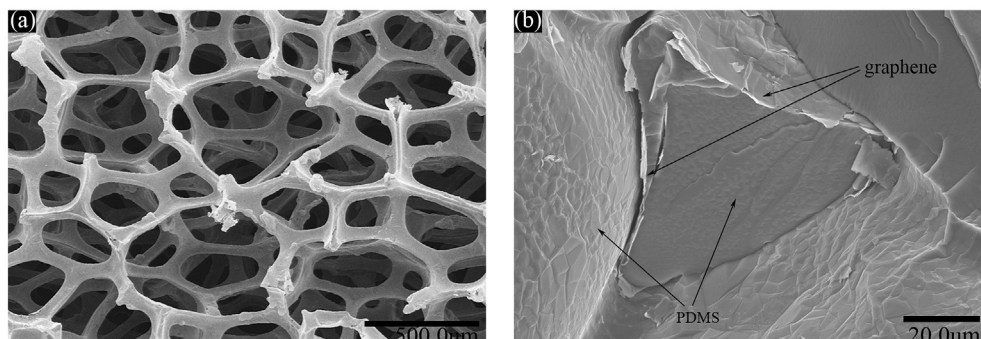

Fig. 1. SEM micrographs of (a) graphene foam and (b) fractured surface of GF/PDMS composite.