

Effect of non-covalent functionalisation on thermal and mechanical properties of graphene-polymer nanocomposites



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ARTICLE INFO

Article history:

Received 9 October 2015

Received in revised form

11 January 2016

Accepted 22 February 2016

Available online 23 February 2016

ABSTRACT

Fast growing power densities of modern electronic devices demand high-performance thermal interface materials (TIMs). Owing to the superior thermal conductivity of graphene, composites with graphene fillers dispersed in polymer matrix are expected to be promising TIM candidates. However, the thermal conductivity of graphene-polymer composites is hindered by a high thermal resistance across the interface between graphene fillers and polymer matrix. This research focuses on modulating the thermal transport across the graphene-polymer interface by employing a non-covalent functionalisation technique. Using molecular dynamics simulations, the effects of different non-covalent functional molecules on the graphene-paraffin interfacial thermal resistance are investigated systematically. It is found that the interfacial thermal resistance can be considerably reduced by non-covalent functionalisation and the reduction depends on the coverage of functional molecules. The thermal transport properties of the composites are improved without compromising their mechanical properties. Different functional molecules including 1-pyrenebutyl, 1-pyrenebutyric acid and 1-pyrenebutylamine can produce similar reductions in the interfacial thermal resistance. Based on the effective medium theory, it is demonstrated that the overall thermal conductivity of graphene-paraffin composites increases when the interfacial thermal resistance decreases, which can be achieved by using the non-covalent functionalisation technique.

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1. Introduction

Efficient heat dissipation has become a critical requirement for the design of modern electronic packages, not only because of the rapidly increasing power densities of current electronic devices, but also due to the fast development of next generation devices, such as 3-dimensional integrated circuits (ICs), microelectromechanical systems and light emitting diodes, etc. [1–3]. In a modern high-power IC package, thermal interface materials (TIMs) are widely employed to manufacture the most critical parts in the heat dissipation system. TIMs are the materials used between two contacting solid surfaces, i.e., between the IC chip and the heat sink. Their function is to enhance the heat transfer from the IC to the heat sink by eliminating the interstitial air gaps between the contacting solid surfaces. TIMs are mostly composed of a polymer-based material

matrix and dispersed thermally conductive fillers. Till now, the thermal conductivity of commonly used composite TIMs spreads from 1 to 10 Wm^{−1}K^{−1} [4]. It is widely recognised that the thermal conductivity of current TIMs is one of the main bottlenecks hindering the heat dissipation efficiency of modern electronic packages, and thus, the development of superior TIMs has attracted great research interest [4,5].

Owing to the extremely high thermal conductivity of graphene (i.e., 3080–5300 Wm^{−1}K^{−1} at room temperature) [6,7], it is expected that composites composed of polymer matrix and graphene fillers should be excellent TIM with a significantly improved thermal conductivity. Extensive research has thus been carried out to synthesise and characterise graphene-polymer composite TIMs. For example, in 2012, Shahil and Balandin [8] synthesised nanocomposite TIMs using graphene fillers and epoxy resin matrix. At a filler loading fraction of 10 vol.%, the thermal conductivity of the nanocomposite was measured to be 5.1 Wm^{−1}K^{−1}. Similarly, Yu et al. [9] found that the graphene-epoxy composites with 25 vol.% graphene loading yielded a thermal conductivity of 6.44 Wm^{−1}K^{−1}.

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At a graphene loading of 5 vol.%, it was experimentally measured that the graphene-paraffin nanocomposites had a thermal conductivity of $2.41 \text{ W m}^{-1} \text{ K}^{-1}$ [10]. Based on the research studies so far, the reported thermal conductivity of graphene-polymer composite TIMs spreads from 1 to $7 \text{ W m}^{-1} \text{ K}^{-1}$, which is far below those predicted by theoretical studies [8–16]. One of the main reasons for these unsatisfactory results was because of the high interfacial thermal resistance (or Kapitza resistance) between the graphene fillers and the polymer matrix [8–17].

To reduce the graphene-polymer interfacial thermal resistance, various techniques have been proposed. Two most investigated are non-covalent functionalisation and covalent functionalisation [18–25]. Non-covalent functionalisation is more advantageous than covalent ones, because it does not introduce defects into the graphene basal plane, and thus does not destruct the intrinsic thermal transport properties of graphene [23,24]. In 2013, Lin and Buehler [23] conducted molecular dynamics (MD) simulations to investigate the effect of non-covalent functionalisation on the interfacial thermal transport between graphene and octane. Using C₈-pyrene as functional molecules, it was demonstrated that the interfacial thermal conductance can be enhanced by about 22% compared to that without functional molecules. In 2011, Teng et al. [24] carried out experiments to functionalise graphene non-covalently using poly (glycidyl methacrylate) containing localised pyrene groups as the functional molecules. The non-covalently functionalised graphene fillers were then dispersed in an epoxy matrix to synthesize composite TIMs. At a filler loading of 4 wt.%, the thermal conductivity of the composites with non-covalently functionalised graphene fillers was measured to be 20% higher than that of the composites with pristine graphene. Clearly, previous research findings have indicated that non-covalent functionalisation is promising in reducing the graphene-polymer interfacial thermal resistance, thereby improving the thermal conductivity of composite TIMs. However, there are still some critical uncertainties towards the effective and reliable application of the non-covalent functionalisation technique in graphene-polymer composites. For example, what are the effects of different types and coverage of non-covalent functional molecules? How does non-covalent functionalisation affect the overall thermal conductivity of the composite TIMs at different filler loadings? The reliable application of composite TIMs under a high temperature environment (i.e., inside the IC packages) also depends on the mechanical integrity of the composites. Hence, it is also of great importance to understand how the mechanical properties of graphene-polymer composites can be influenced by non-covalent functionalisation.

In this work, firstly, large-scale MD simulations are conducted to investigate the thermal transport across graphene-paraffin interfaces with graphene decorated by various non-covalent functional molecules including 1-pyrenebutyl, 1-pyrenebutyric acid and 1-pyrenebutylamine. Secondly, the effect of non-covalent functionalisation on the overall thermal conductivity of graphene-paraffin composite TIMs is evaluated by the effective medium theory. Finally, using MD simulations again, the effect of non-covalent functionalisation on the mechanical behaviours of graphene-paraffin composites subjected to uniaxial tensile loading is examined.

2. Computational methods

In this work, the thermal transport across graphene-paraffin interfaces was investigated by employing the reverse nonequilibrium molecular dynamics (RNEMD) simulation method based on Muller-Plathe's approach [26]. Large-scale atomic/molecular massively parallel simulator (LAMMPS) [27] was used to

carry out all the simulations. While simulating the graphene-paraffin composites, the adapted intermolecular reactive empirical bond order (AIREBO) potential was chosen to describe the interactions between the carbon atoms in graphene [28]. The polymer consistent force field (PCFF) was used to model the paraffin matrix and non-covalent functional molecules [29,30]. No chemical bond exists between the graphene, paraffin matrix and non-covalent functional molecules, hence the interactions between these different components are purely van der Waals (vdW) interactions, which were described by Lenard-Jones (LJ) potential. The LJ potential is expressed as: $V(r_{ij}) = 4\epsilon[(\sigma/r_{ij})^{12} - (\sigma/r_{ij})^6]$, where r_{ij} is the distance between atoms i and j ; ϵ and σ are the energy and distance constants, respectively. The parameters (ϵ and σ) were adopted from the vdW term in the PCFF from *Material Studio*. The LJ potential parameters across different types of atoms were calculated by applying the Lorentz-Berthelot mixing rules, i.e., $\epsilon_{ij} = \text{sqrt}(\epsilon_i \epsilon_j)$; $\sigma_{ij} = (\sigma_i + \sigma_j)/2$. The detailed parameters used in the present simulations can be found in our previous work [20].

Paraffin (C₃₀H₆₂) was used as the matrix material of the composite TIMs. The paraffin blocks were initially built in *Material Studio* using the Amorphous Cell module, which generates bulk disordered systems of paraffin molecules. Then, the composites were constructed by sandwiching the paraffin blocks with other components including graphene sheets and non-covalent functional molecules. As shown in Fig. 1a, the composite models were designed to have a cross-section area of $29 \text{ \AA} \times 29 \text{ \AA}$ and a length of $160 \text{ \AA}$. Periodic boundary conditions were applied in all three directions. The simulation models have more than 16,128 atoms. A small time step of 0.25 fs was used in all the simulations due to the presence of light hydrogen atoms. At the beginning of the simulations, an energy minimisation was performed using the conjugate gradient algorithm. The composite model was then annealed in a canonical NVT ensemble (i.e., constant number of atoms, volume and temperature) for 0.5 ns. During annealing, the system temperature was raised from 300 K to 1000 K, and then cooled down to 300 K at a rate of 7 K/ps. Finally, the composite model was fully relaxed in an isothermal-isobaric NPT ensemble (i.e., constant number of atoms, pressure and temperature) at 300 K and 1 atm for

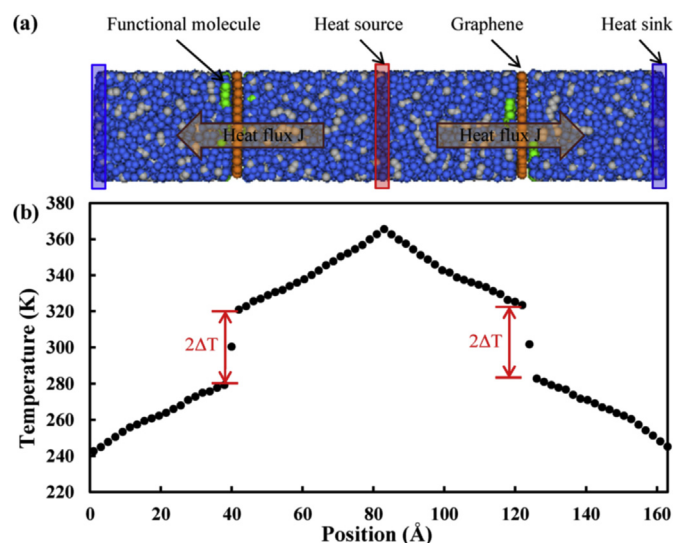


Fig. 1. (a) Composites composed of paraffin, graphene and non-covalent functional molecules in RNEMD simulation (Blue balls are hydrogen atoms, grey balls are carbon atoms in paraffin, orange balls are carbon atoms in graphene and green balls are carbon atoms in functional molecules); and (b) the resultant temperature gradient along the length of the composites. (A colour version of this figure can be viewed online.)

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