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Effect of a metallic coating on the thermal conductivity of carbon nanofiber–dielectric matrix composites

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ABSTRACT

A theoretical model is developed to evaluate the thermal conductivity of composites made of a dielectric matrix material containing randomly oriented and aligned carbon nanofibers coated with a metallic layer. The effect of the metallic coating on the phononic thermal conductivity of the matrix material and the electron–phonon coupling inside the metallic coating are both taken into account in this model. It is shown that: (1) the metallic coating has an extraordinary effect on the enhancement of the composite thermal conductivity. For a volume fraction of 30% of fibers with radius of 50 nm and 10 nm-coating of copper, the increase in the thermal conductivity is as high as 27%, which increases significantly with the fiber volume fraction. (2) Although the thermal conductivity of silver is 453% as that of indium, the composite thermal conductivity is only increased slightly by changing an indium coating to a much more expensive silver coating, due to the relatively high thermal conductivity of these metals in comparison with the one of the matrix. (3) The composite thermal conductivity increases with the volume fraction of the fibers when their radius and the radial thermal conductivity are greater than the Kapitza radius at the matrix-coating interface and the effective thermal conductivity of the matrix, respectively. The obtained theoretical results agree fairly well with experimental data reported in the literature for the thermal conductivity along the axis of aligned carbon fibers with copper coating and embedded in an epoxy matrix. This model is expected to be valid for composites in the absence of percolation with the length-to-radius aspect ratio of fibers in the range of 10–100, and it provides theoretical guides for optimizing cost-efficient high thermal conductivity composites.

1. Introduction

Nanostructured carbon materials such as carbon nanotubes (CNTs), graphene and carbon nanofibers (CNFs) are widely used as reinforcement materials for composites because of their outstanding mechanical, thermal and electrical properties, which can significantly improve the performance of various materials for industrial applications [1–3]. It is very common to optimize conflicting requirements on the material properties by combining the most useful properties of two or more phases, which does not ordinarily appear together in nature. Especially, carbon-fiber-reinforced polymers have become essential nowadays in aerospace and automobile industry. For example, using CNFs instead of steel can lower the weight of the involved components by up to 50%, which can improve fuel economy for vehicles by as much as 40% [2,4]. Typical aspect ratios (length/diameter) of these fibers are on the order of tens to hundreds, which facilitate the control on their random or aligned distribution inside the matrix, and the understanding and prediction of the physical properties of CNFs-based composites.

Significant research efforts have been dedicated to improve the thermal conductivity of composites made up of CNFs embedded in a dielectric matrix for thermal management applications [5–7]. The poor thermal conductivity of the CNFs in the radial direction, which can be as low as three orders of magnitude smaller than that along their axial direction, has limited the improvement. Metallic coating of the CNFs has been proposed recently to overcome this problem, with production techniques developed [8,9]. Taking into account the relatively high thermal conductivity of metals, the surface metallization of CNFs is expected to enhance the overall thermal conductivity of these composites and therefore extending their application spaces. However, the thermal performance of these composites is still not well understood. Given that the thickness of the coating layer can be on the order of the mean free path of the energy carriers (electron and phonons), which is typically of

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a few nanometers for metals at room temperature, the thermal conductivity of these fibers and therefore of the whole composite is expected to depends strongly on interactions of energy carriers and their scattering processes with interfaces [10–15]. Currently there are no theoretical models that can provide design guidelines.

The objective of this work is to develop a theoretical model to quantify the thermal conductivity of composites consisting of a dielectric matrix embedded with randomly oriented and aligned CNFs that are coated with a metallic layer. The effect of the metallic coating is determined by comparing the enhancement of the thermal conductivity of composites as a function of coating thickness for different metallic coatings. This work could shed some light on the design of cost-effective high thermal conductivity CNF–dielectric composites.

2. Theoretical model

Fig. 1(a) shows a composite consisting of CNFs with a metallic coating, embedded in a dielectric matrix. The effective thermal conductivity of these fibers is anisotropic and it has the values $k_\parallel$ and $k_\perp$ in the axial and radial directions, respectively. The coating and the matrix are assume to have the isotropic thermal conductivities k_c and k_m, respectively. Starting with the our previous thermal conductivity model valid in the dilute limit of particles (Section 2.1) [11], the theoretical model for the composite thermal conductivity is developed here, by taking into account the fiber–fiber interaction at the non-dilute limit (Section 2.2) and the size effect on the thermal conductivity of the matrix and the coating, due to the small sizes of the coating and the CNFs (Section 2.3).

The modeling of the composite thermal conductivity is done by assuming that: (1) the aspect ratio (length/diameter) of the fibers is small enough to warrantee their randomness inside the matrix and in the absence of percolation, at least within a wide range of volume fractions of the fibers. This could be easily achieved for aspect ratio on the order of or smaller than 100. (2) The aspect ratio of the fibers is big enough (much greater than the unity) to make sure that they can provide a preferential direction of conduction along their axis. The lower bound of this aspect ratio can be accurately determined through the geometrical factors, which define the geometry effect of particles [16]. Fig. 2 shows these factors along the major ($L_\parallel$) and minor ($L_\perp$) axes of an ellipsoidal particle. Note that as the aspect ratio of the ellipsoid increases, the geometrical factors $(L_\parallel, L_\perp) = (1/2, 0)$, which are the values for a very long cylindrical fiber of circular cross section. For an aspect ratio of $a_\parallel/a_\perp = 10$, the geometrical factors deviate from their corresponding values for an infinite fiber by just about 2%, and this deviation reduces for higher aspect ratios. This indicates that a fiber with an aspect radio equal or greater than 10 has a very similar geometrical effect than another one with an infinite aspect ratio. Thus, it is clear that our assumptions for the composite under consideration can be fulfilled when the aspect ratio of the fibers is greater than 10 but smaller than 100.

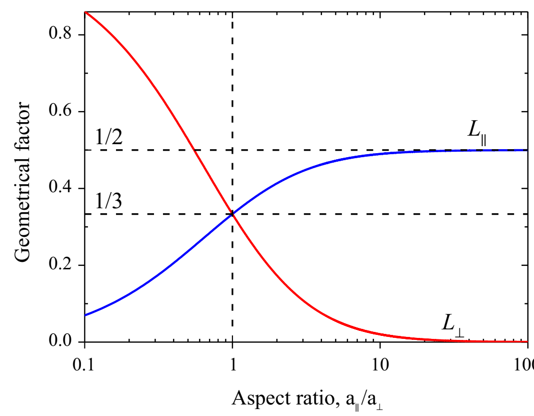

Fig. 2. Geometrical factors of an ellipsoidal particle as a function of its aspect ratio.

2.1. Tri-phase thermal conductivity model under the dilute limit

The description of the effective thermal conductivity of a tri-phase composite shown in Fig. 1(a) requires capturing the effects of the thermal and geometrical properties of the CNFs, its metallic coating, and the dielectric matrix. Based on the temperature profile inside a composite exposed to a constant heat flux and assuming that the volume fraction f of coated spheroidal particles is small enough ($f \ll 1$, dilute limit) to neglect their interactions, Ordonez-Miranda et al. [11] have derived a model for the effective thermal conductivity k of a tri-phase composite. For cylindrical particles with random spatial distributions, as is the case of CNFs shown in Fig. 1(a), the results by Ordonez-miranda et al. reduce to [11]

$$\frac{k}{k_m} = \frac{3 + (\beta_1 + \beta_3)f}{3 - \beta_1 f}, \tag{1a}$$

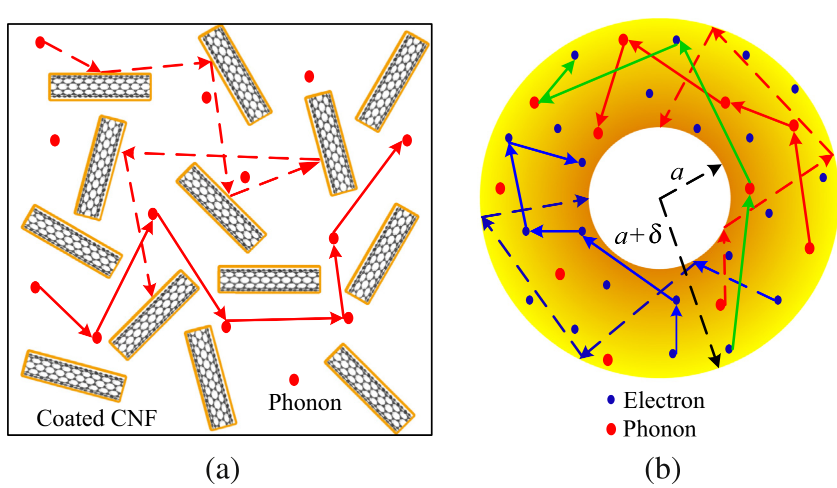

Fig. 1. (a) The phonon scattering processes inside the matrix, (b) the electron and phonon scattering processes in the δ-thickness metallic coating of a CNF with radius a (cross-section).