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Thermal or electrical bulk properties of rod-filled composites

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The cell model (or self-consistent scheme) has been largely used to estimate the bulk thermal properties for dilute slender rods embedded in a matrix by applying a thermal gradient perpendicular to the main axis of the rod. This approach is then extended by considering a thermal gradient along the particle, or in other words a thermal flux through the end surfaces of the fiber. For a slender-body, this additional contribution is found to be negligible. Then, to circumvent the diluteness assumption, particle-particle contacts are considered when determining the effective thermal conductivity of the composite. This involves adding to the dilute result a new contribution depending on the magnitude of the Biot number (defined later) and on the micro-structures of the particles, and exhibiting a quadratic dependence on the particle concentration. Two assumed contact surface areas are investigating leading to the development of two micro-mechanical models, the difference being in the choice of the contact area. In addition, the model predictions are in good agreement with previously published data, where in-plane measurements of effective thermal conductivity are performed on highly conductive copper fibers impregnated with a poorly conductive PMMA matrix, for a wide range of fiber orientations and concentrations. When an appropriate cell size dimension for the dilute model is adopted, its predictions are also found to give an accurate fitting. Although this paper focuses on the thermal properties, the derivation can be carried over to the electrical properties with very minor modifications. This has been carried out for estimating the effective electrical conductivity of several CNTs (i.e., SWCNT and MWCNT) embedded in an epoxy matrix, leading again to good agreements.

1. Introduction

The determination of the bulk properties for composite materials from their microscopic structures and components is still a challenging problem in many branches of physical science and engineering, such as the thermal or electrical conductivities, dielectric constant or magnetic permeability. In many applications, the inclusion of fillers leads to innovative lightweight solutions, whilst not compromising on physical properties (i.e., lightning strike prevention, static charge accumulation prevention, electromagnetic immunity, dynamic damping, heat dissipation, etc.). Potential matrix materials involve plastics, ceramics and metals, and filler materials may include metal, carbon, ceramics and hybrid composites (Han & Fina, 2011; Radzuan, Sulong, & Sahari, 2017a).

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For polymer composites, the enhancement of effective transport properties can be achieved by using highly conductive particles since the corresponding properties for polymers are usually quite low and, therefore often neglected in the design of conductive composites. Amongst fillers, conductive slender rods (such as short carbon fibers (Keith, Hingst, Miller, King, & Hauser, 2006; Nithikarnjanatharn, Ueda, Tanoue, Uematsu, & Iemoto, 2012; Ram, Rahaman, & Khastgir, 2014), carbon nanotubes (Dalmas, Dendievel, Chazeau, Cavaill, & Gauthier, 2006; Kim et al., 2006; 2007) and metallic rods (Cheng, Sastry, & Layton, 2001; Danès, Garnier, & Dupuis, 2003; Danès, Garnier, Dupuis, Lerendu, & Nguyen, 2005; Tekce, Kumlutas, & Tavman, 2007)) are relevant owing to the formation of a potential connected network, even at very low filler concentration. This network is supposed to traverse the specimen and produces a significant increase in the thermal/electical conduction. Effects of particle shape and content (Bigg, 1986; Flandin, Verdier, Boutherin, Brechet, & Cavaill, 1999; Hussain et al., 2017; Kim et al., 2007; Mahdavi, Yousefi, Baniassadi, Karimpour, & Baghani, 2017; Néda, Florian, & Brechet, 1999; Nithikarnjanatharn et al., 2012; Ram et al., 2014; Sanada, Tada, & Shindo, 2009; Tekce et al., 2007; Zhang, Luo, Guo, & Wu, 2017; Zhu, Ma, Wu, Yung, & Xie, 2010) as well as inter-particle contact zones with hypothetical interfacial thermal barriers on the conduction properties (Bhatt, Donaldson, Hasselman, & Bhatt, 1990; Chekanov, Ohnogi, Asai, & Sumita, 1998; Dalmas, Cavaillé, Gauthier, Chazeau, & Dendievel, 2007; Géminard, Bouraya, & Gayvallet, 2005; Géminard & Gayvallet, 2001) of filled polymer composites have been explored. The orientation and spatial distributions of rods can also play a major role since both may evolved during processing (Agari, Ueda, & Nagai, 1994; Chekanov et al., 1998; Danès et al., 2005; Fu & Mai, 2003; Hussain et al., 2017; Orgéas et al., 2012; Qu, Nilsson, & Schubert, 2018; Qu & Schubert, 2016; Taipalus, Harmia, Zhang, & Friedrich, 2001).

From a modeling point of view, Shen, Cui, He, and Zhang (2011) proposed a review on theoretical and empirical models for predicting the thermal conductivity of polymer composites. Due to the high contrast between the two phases, most models based on the self-consistent field method, which give lower and upper limits, may not be useful for these composite materials (Carson, Lovatt, Tanner, & Cleland, 2005). To overcome this, several models have been dedicated to systems exhibiting a fibrous micro-structure (Berhan & Sastry, 2007a; 2007b; Cheng & Sastry, 1999; Cheng et al., 2001; Favier, Dendievel, Canova, Cavaille, & Gilormini, 1997; Mahdavi et al., 2017; Sanada et al., 2009; Sundararajakumar & Koch, 1997; Yu et al., 2013). Examination of particle volume fraction and aspect ratio have been performed but few of them investigated the effects of the micro-structure orientation state (Cheng et al., 2001; Herrmann, 2015; Mackaplow, Shaqfeh, & Schiek, 1994; Marcos-Gómez, Ching-Lloyd, Elizalde, Clegg, & Molina-Aldareguia, 2010; Pal & Kumar, 2016; Sundararajakumar & Koch, 1997) and particle-particle contacts (Dalmas et al., 2006; Pal & Kumar, 2016; Vassal, Orgéas, Favier, Auriault, & Le Corre, 2008a,b).

In this context, this research work attempts to derive an effective conductivity tensor of a non-deformed rod-filled material through which there is a steady transport of heat. The approach is based on the Batchelor ideas (Batchelor, 1974), which is summarized and applied for dilute slender bodies in the next section. Then in Section 3, we attempt to investigate the effect of considering the thermal flux through the flat edges of a rod on the conductive properties of the bulk medium. In Section 4, the case for non-dilute concentrations of rods are attempted by taking into account a heat flux density at the contact points between interacting particles. This last section ends by showing some model predictions against measurements of in-plane thermal conductivity of cooper-filled PMMA polymer composites. It is useful to note that the results obtained in this work maybe also applied for the transport of electricity (the governing equations are the same, only the terminology changes). In this framework, the model is updated to estimate the electrical conductivity for SWCNTs and MWCNTs embedded in an epoxy matrix.

2. Theoretical background

2.1. Average thermal conductivity

The effective thermal conductivity of a statistically homogeneous composite material is considered and can be defined in terms of volume-averaged quantities (Batchelor, 1974). Hence, the present problem involves relevant quantities such as the average temperature gradient $\langle \nabla T \rangle$ and the average heat flux density $\langle \mathbf{q} \rangle$, respectively defined by

$$\langle \nabla T \rangle = \frac{1}{V} \int_V \nabla T dV, \tag{1}$$

and

$$\langle \mathbf{q} \rangle = \frac{1}{V} \int_V \mathbf{q} dV, \tag{2}$$

where V is a volume large enough to contain many particles (representative volume), $\langle \nabla T \rangle$ is the constant temperature gradient of the imposed linear ambient field $\langle \nabla T \rangle \cdot \mathbf{r}$ and $\mathbf{r}$ is a location vector from a fixed coordinate system. Because of the linearity of the governing equation (i.e., Laplace equation) in both the matrix and particles, it follows that $\langle \mathbf{q} \rangle$ and $\langle \nabla T \rangle$ are linearly related such as Phan-Thien (1980a,b)

$$\langle \mathbf{q} \rangle = -\boldsymbol{k}_c \cdot \langle \nabla T \rangle, \tag{3}$$

where $\boldsymbol{k}_c$ is defined as the effective thermal conductivity tensor of the composite. The heat flux density $\langle \mathbf{q} \rangle$ is usually expressed as Batchelor and O'Brien (1974)

$$\langle \mathbf{q} \rangle = \frac{1}{V} \int_{V-\sum V_i} \mathbf{q} dV + \frac{1}{V} \sum_i \int_{V_i} \mathbf{q} dV = -k_m \langle \nabla T \rangle + \frac{1-\alpha^{-1}}{V} \sum_i \int_{A_i} \mathbf{r}\mathbf{q} \cdot \mathbf{n} dA. \tag{4}$$