



Research Paper

Thermal conductivity of filled composite materials considering interactions between fillers

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HIGHLIGHTS

- The 10% is the inflection point and percolation threshold for cube fillers with ideal filler contacts.
- The contacts and shapes of fillers are considered.
- A series of new generalized models are proposed.
- 3-dimensional, 2-dimensional modeling and experimental tests are compared.

ARTICLE INFO

Keywords:

Composite materials
Filler contacts
Thermal conductivity
Generalized model
Isolated model

ABSTRACT

Adding high thermal conductive fillers to base materials has been recognized as an efficient way to increase the thermal conductivity of composites materials. The heat conductive path is formed by filler contacts, this is a key point. In this paper, the effects of filler contacts are investigated based on generalized model and isolated model. The results indicate that the thermal conductivity is always linearly increased with the sphere filler contents. However, for cube filler, when the volume content exceeds 10%, the growth of thermal conductivity is accelerated along with filler contents. The 10% filler content is the inflection point and percolation threshold for cube filler. It is much lower than previously reported. The effect of filler contacts is very small for sphere fillers. Meanwhile, the deviation of thermal conductivity between two-dimensional and three-dimensional model is also very small for sphere filler. The cube fillers have substantial effects on the consequently thermal conductivity than sphere fillers. A series of new three-dimensional generalized model are proposed by numerical method. The filler contacts are considered in this model, the deviation with the experiment tested is in an acceptable range at wide filler contents.

1. Introduction

Heat exchangers are the key equipments in various industries [1,2]. The conventional heat exchanger manufactured in metal (such as aluminum, copper and stainless steel) has the disadvantages in terms of weight and cost. In addition, specially treated metal heat exchangers are needed if the working fluids are corrosive [3,4]. Given these considerations, polymers instead of metals have been used as the materials to make heat exchangers. With the advantages of greater fouling and corrosion resistance, greater geometric flexibility and ease of manufacturing, reduced energy of formation and fabrication, and the ability to handle liquids and gases, polymer heat exchangers have been widely studied and applied in the field of micro-electronic cooling devices, water desalination systems, solar water heating systems, liquid

desiccant cooling system, etc [5–7]. However, the thermal conductivity of most polymers is lower than $0.2 \text{ W m}^{-1} \text{ K}^{-1}$, which are around 200 times lower than those of most metals. Because of this, it limits the application of polymers in heat transfer [8,9].

To increase the thermal conductivity of polymers, adding fillers of high conductivity to the base materials has been practiced as an efficient method to make new materials for heat exchangers. The major fillers used in heat conductive composites are particles, fibers, flakes, and laminas [10–12]. It has always been the focus to predict the thermal conductivity of filled composite materials by models. Several classic models are Maxwell mode [13], Russell model [14], Nielsen model [15] et al. In addition, many new models are being proposed continuously [16–19]. These models consider the volume fraction, filler shape, filler size and thermal conductivity as the factors that influence

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Nomenclature

c_p	specific heat ($\text{kJ kg}^{-1} \text{K}^{-1}$)
h	convective heat transfer coefficient ($\text{W m}^{-2} \text{K}^{-1}$)
K	thermal conductivity ($\text{W m}^{-1} \text{K}^{-1}$)
L_x	length along the x axis (m)
L_y	length along the y axis (m)
L_z	length along the z axis (m)
n	normal direction
q	heat flux (W m^{-2})
T	temperature (K)
V	volume content

x, y, z	coordinates (m)
x_0, y_0, z_0	coordinates of filler center (m)

Greek letters

ρ	density (kg/m^3)
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Subscripts

c	composite
f	filler
m	polymer matrix

the final thermal conductivity. Regretfully, none of these models is a general model that can predict the final thermal conductivity accurately for various filled composite materials. Most predicted values are smaller than the experiment data, the difference increasing with the filler content increasing. The reason is that these models have not considered the filler contacts [20–22]. The larger fillers and heat conductive paths are formed by filler contacts in a random filler position arrangement. There are less filler contacts at lower contents. However, the number of filler contacts rapid increase with contents. These will greatly affect the final thermal conductivity of filled composite materials. No research has provided a generalized model which considers filler point contact, line contact and surface contact to predict the thermal conductivity of filled composite materials.

The goal of this study is to build the generalized model to predict the thermal conductivity of filled composite materials. The isolated model without filler contact is also built to investigate the effects of particle shape and three filler contacts. Furthermore, the accuracy of three-dimensional and two-dimensional models are validated by experiments.

2. Numerical work

2.1. Generation of cell model

Two-dimensional models and its effect on the final thermal conductivity resulting are investigated in Section 3. This section will demonstrate the numerical work using three-dimensional model as an example. The effects of point contact, line contact and surface contact are researched by contrasting generalized model and isolated model. The shapes of particle filler are sphere, or cube. The sphere filler can contact with other fillers by point contact. They only have one-point contact between each two sphere fillers. However, a sphere filler can in contact with multiple sphere filler. The contact between each two cube fillers can be one of point contact, line contact, or surface contact. Meanwhile, a cube filler can in contact with multiple other fillers. The volume of filler studied is 0.001 cm^3 . The cell selected here is a $1 \times 1 \times 1 \text{ cm}^3$ and has a dozen of fillers, represents a periodic section in the filled composite materials.

The fillers are randomly assigned in the base materials. A computer program is developed to automatically generate the positions of fillers [23]. Let (x_0, y_0, z_0) represent the positions of filler centers. Lets L_x, L_y, L_z denote the length, width and height of the cell model respectively, as shown in Fig. 1. Then a random number r_x ($0 < r_x < 1$) is generated by the computer. The coordinate x_0 is selected as the product r_x of and L_x . In a similar way, the coordinate y_0 and z_0 are obtained. In the process of the filler generation for isolated models, if a new filler is found to overlap with any other old fillers, filler surfaces, or unit cell boundaries, it is canceled and a new one is generated. When generating a generalized model, the point contact, line contact and surface contact are retained. They have no the volume overlap between fillers in composites. In the process of filler generation, if a new filler is found to

overlap with any other old fillers, it is cancelled and a new one is generated. The filler generation process ends when a desired filler content is reached. The generalized model and isolated model generated by the computer are shown in Fig. 2(a) and (b). The filler shape is cube and filler content is 10%. As seen, the filler distribution in the isolated model is quite random without filler contact. But in the generalized model, when a filler has contact with one or more fillers by its points, lines and surfaces, a new larger filler is formed and the heat conductive path is extended. The number and size of such heat conductive paths increases with filler contents. There are beneficial for thermal conductivity increasing. The two-dimension models are also built by the similar way. To reduce uncertainty of random model, there are five models are generated for each filler content. The effective thermal conductivity of these models is stable and the differences are within 3.5%.

Fig. 3 presents the effects of computational mode sizes on the thermal conductivity of composite. As seen, when the model size becomes larger than $0.5 \times 0.5 \times 0.5 \text{ cm}^3$, the thermal conductivity is stable and the fluctuations from generation of cell is lower than 2%, regardless of the filler content. The differences of thermal conductivity in x, y and z directions are within 2%. The cell selected here is a $1 \times 1 \times 1 \text{ cm}^3$ cube. It is large enough for this three-dimensional thermal conductivity calculation.

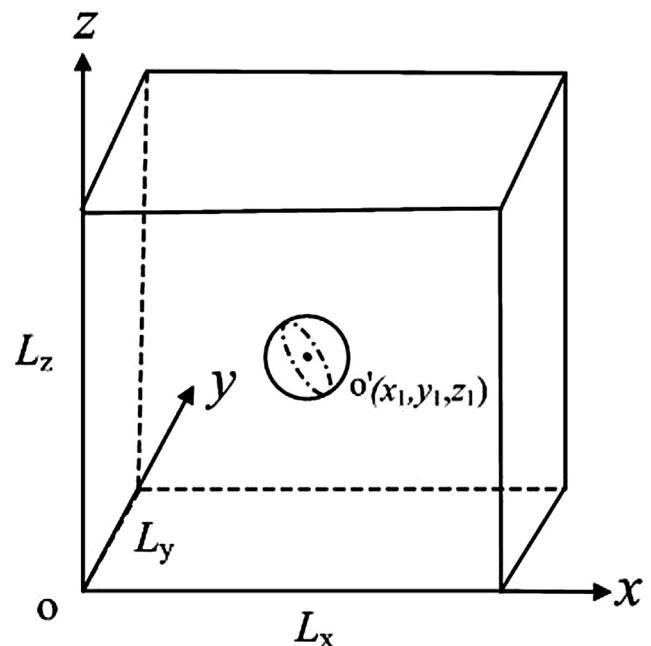


Fig. 1. Schematic of a filler arranged in base materials.

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