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Research paper

Analytical heat conduction model of particle reinforced tertiary composite materials based on complete spatial randomness of fillers in base matrix and its application in the development of cryosorption pump

Aritra Chatterjee[a,b], Ravi Verma[a,b], N.C. Shivaprakash[b], S. Kasthurirengan[a,*], Upendra Behera[a]

[a] Centre for Cryogenic Technology, Indian Institute of Science, Bangalore 560012, India
[b] Department of Instrumentation and Applied Physics, Indian Institute of Science, Bangalore 560012, India

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ABSTRACT

In this article, we propose an analytical heat conduction model within a stochastic frame work which estimates the thermal conductivity (TC) value of particle reinforced composite materials comprising of three parent elements i.e. a base matrix along with two different filler element particles randomly distributed in it. The spatial distribution of the filler particles in a sample of specific dimension has been estimated by applying bivariate Poisson distribution. This distribution is then used to arrive at the TC value of the composite. This concept has been applied to predict the TC of the tertiary composite comprised of epoxy as the base matrix, aluminium and zinc particles as filler elements. The TC values obtained from this model for different volume fractions of fillers were extensively compared with experimental results. The model is found to predict the results fairly well with less aberrations up to the total filler volume fraction of ~20%. The developed model for TC prediction has been used in the design of high efficiency cryosorption pump where the adhesive material used is Epoxy-Aluminium - Zinc composite.

1. Introduction

Thermal conductivity (TC) is an important property of the materials. It is essential to know the precise value of this property, based on which the suitable material can be chosen for a specific application. This is important for the thermal design of individual components as well as the total system. Thermal conductivity of a material depends on various parameters such as temperature, composition, impurities and structure of the material etc. With the advancement of technology, more and more composites with filler materials are being developed whose thermal conductivities values are yet to be known. From engineering point of view, the knowledge of thermal conductivity values of new composite materials is quite important for their applications.

Most of the models for the estimation of TC for particle reinforced composites available in the literature are valid for binary composites i.e. composites comprising of two parent elements, a base matrix and a filler element. In addition, most of them are empirical in nature which prevents one to obtain the actual picture of the heat transfer mechanism through the composite material.

In this article, we propose an analytical model that is suitable for use with the tertiary composites consisting of a base matrix along with two different filler particles randomly distributed in it. The results from this model tends to those of a regular binary composite (the base matrix with a single filler element) when the volume fraction of one of the filler component tends to zero. The developed model has been applied to the Epoxy-Aluminium-Zinc tertiary composite and the TC values predicted by the model are in good agreement with the experimental data of thermal conductivity measured in our laboratory using a cryocooler based TC measurement experimental setup in the temperature range from 50 K to 300 K. The developed model has also been applied to a tertiary composite with graphite and silicon carbide in a common epoxy and the results predicted by the model are in good agreement with the experimental results till the total filler volume fraction of ~ 20%..

1.1. Different models used for predicting TC of composites

Several models have been developed to determine the thermal conductivity of binary composite materials [1–9]. Some of them are briefly described below. Maxwell [1] assumed random dispersion of non-interacting homogeneous spherical filler particles of thermal conductivity k_f within a continuous substrate of thermal conductivity k_m to

* Corresponding author.
E-mail address: ksrinivasan@iisc.ac.in (S. Kasthurirengan).

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Nomenclature

Symbols used

k_{eff}	Effective thermal conductivity, $Wm^{-1}K^{-1}$
K_{th}	Theoretical thermal conductivity, $Wm^{-1}K^{-1}$
K_{exp}	Experimental thermal conductivity, $Wm^{-1}K^{-1}$
Ø	Volume fraction, dimensionless
$Ø_{Al}$	Volume fraction of aluminium in the sample, dimensionless
$Ø_{Zn}$	Volume fraction of zinc in the sample, dimensionless
l	Length of the sample, m
a_p	Average length(width) of the filler particle, m
h_p	Average height of the filler particle, m
V	Volume of the entire sample, m^3
v	Volume of single Epoxy-Aluminium-Zinc rectangular cuboid, m^3
N_T	Total number of rectangular cuboids, dimensionless
N_{Al}	Total number of aluminium rectangular cuboids, dimensionless
N_{Zn}	Total number of zinc rectangular cuboids, dimensionless
N_{Ep}	Total number of epoxy rectangular cuboids, dimensionless
N_{Row}	Number of rows after discretization, dimensionless
N_{Col}	Number of columns after discretization, dimensionless
λ_{Al}	Mean number of aluminium particles in each column of sample, dimensionless
λ_{Zn}	Mean number of zinc particles in each column of sample, dimensionless
X	Random variable that takes value x the range x∈ [0, $2\lambda_{Al}$], dimensionless
Y	Random variable that takes value y in the range y∈ [0, $2\lambda_{Zn}$], dimensionless
k_i	Effective thermal conductivity of i^{th} column, $Wm^{-1}K^{-1}$
$E_{X,Y}$	Expected number of columns with X aluminium particles and Y Zinc particles, dimensionless
$P_{Al,x}$	Probability of a column having x aluminium particles, dimensionless
$P_{Zn,y}$	Probability of a column having y zinc particles, dimensionless
$P_{X,Y}$	Joint probability function, dimensionless

obtain the thermal conductivity of the system. However, the model holds good only for low volume fraction of filler material since it assumes no thermal interaction between the particles.

The effective thermal conductivity k_{eff} is given by:

$$\frac{k_{eff}}{k_m} = \frac{k_f + 2k_m + 2Ø(k_f - k_m)}{k_f + 2k_m - Ø(k_f - k_m)} \quad (1)$$

Bruggeman's model [2] allows predicting many properties of composite materials such as thermal conductivity, thermal diffusivity, magnetic permeability etc. This theory assumes that a composite material may be constructed incrementally by introducing infinitesimal changes to an already existing material. This approach is known as Differential Effective Medium (DEM) theory. In this model, k_{eff} is given as,

$$\frac{k_{eff} - k_f}{(k_{eff})^{1/3}} = \frac{(1-Ø)(k_m - k_f)}{(k_m)^{1/3}} \quad (2)$$

Lewis-Nielsen [3] proposed another empirical model which is applicable up to the volume fraction Ø ~ 40% of filler in base matrix. The effective thermal conductivity k_{eff} is given as,

$$k_{eff} = k_m\left(\frac{1 + PQØ}{1 - P\Phi\Psi}\right) \quad (3)$$

where P, Q and Ψ are constants depending on the shape of the filler particles. Russell's theoretical model [4] calculates the thermal conductivity according to the formula:

$$k_{eff} = k_m \frac{Ø^{\frac{2}{3}} + \frac{k_f}{k_m}\left(1 - Ø^{\frac{2}{3}}\right)}{Ø^{\frac{2}{3}} - Ø + \frac{k_f}{k_m}\left(1 + Ø - Ø^{\frac{2}{3}}\right)} \quad (4)$$

The well-known rule of mixtures [5] is another model used by several researchers for determination of thermal conductivity k_{eff} in composites of binary components.

Presently available statistical and mathematical models [10–16] for estimation of TC of composites and polymers provided crucial understanding in the development of the proposed model. Results from available numerical heat transfer models [17–20] of composites and experimental works [21–28] especially at low temperatures helped in foreseeing the trend of TC curve as a function of volume fraction and temperature. Also, to our knowledge no models have been reported in the literature for the determination of TC for a tertiary composite material and ours may be the first one in this direction.

2. Development of heat conduction model for tertiary composites

The procedure of arriving at the TC value of a tertiary composite can be divided into a sequence of steps. First a specific tertiary composite is chosen. In the present case, we have selected the Epoxy-Aluminium-Zinc composite where the epoxy forms the base matrix and the aluminium and zinc are two filler components. SEM analysis is a standard method to find the average dimension of microscopic particles and this has been used to obtain the same for aluminium and zinc particles. Based on the dimensions of the particles measured, a suitable cuboidal shaped sample of the composite is chosen and the latter is further discretized into rows and columns. Then the *bivariate Poisson distribution* has been applied on the discretized sample to obtain the spatial distributions of the two different filler particles in the grid-like discretized sample. These spatial distributions are then used to arrive at the TC of the chosen sample by applying the well-known series-parallel approach. The detailed steps are elaborated in the later sections. It is observed that increasing the volume fractions of the filler particles has a significant effect on the covariance of the distribution of filler particles in the base matrix.

2.1. Implementation of the model to a specific tertiary composite

The model is valid for any tertiary composite material, but for the sake of elaboration, we will consider the specific case of an Epoxy-Aluminium-Zinc. The reason for choosing this particular composite is that it is used as a conducting adhesive material in the fabrication of adsorption panels of cryosorption Pumps. The evaluation of the thermal conductivity of the composite with different volume fractions of fillers is important towards improving the performances of such pumps. The thermal conductivity of the epoxy used in this study is much higher (~1.052 $Wm^{-1}K^{-1}$) than that of the commonly available epoxy resin at room temperature (typically ~ 0.2 $Wm^{-1}K^{-1}$). In view of the strategic nature of patent and other angles, the exact molecular formula for this epoxy cannot be disclosed at the present time. However, the composition of the epoxy for its contents such as Carbon and Oxygen has been determined using an EDAX Energy Dispersive spectroscopy. These details are presented in Table 1. For comparison purposes, we have also included the same analysis for Stycast 2850FT, a commercial epoxy adhesive. From the table, it can be seen that the carbon content for the developed epoxy is significantly higher (~86%) when compared to that of Stycast 2850FT (~55%). Therefore the developed epoxy has a higher

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