Effective thermal conductivity of metal filled polymer composites

Ramvir Singh* & P K Sharma
Heat Transfer and Solar Energy Laboratory, Department of Physics, University of Rajasthan, Jaipur 302 055
*E-mail: singhrvs@rediffmail.com

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Theoretical model for predicting effective thermal conductivity (ETC) of metal filled polymer composites has been developed. The concept of averaging the temperature field within different phases has been used. Resistor model has been applied to determine ETC of polymer composites. A parameter estimation technique has been applied to determine the inclination angle $\theta$. An effort has been made to correlate the angle of inclination $\theta$ in terms of the ratio of thermal conductivity of the constituents and their volume fractions. Best-fitted expression so obtained for $\theta$ is presented here. A good agreement has been observed between estimated values and earlier experimental data. Comparison of the proposed relation with Lewis and Nielsen’s models has also been made.

Keywords: Effective thermal conductivity, Series and parallel resistors, Continuous medium, Angle of inclination

1 Introduction

The modeling and estimation of thermo-physical properties of complex materials such as ceramics, metal foams and metal filled polymer composites are becoming increasingly important in the technological developments and in many engineering applications. It has drawn considerable interest from researchers and polymer industry, from manufacturers to industrial end-users. The most important design variables in the development of these materials are production technologies and the choice of the material making up the solid phase of the composite. Thermal behaviour of composite materials is determined by the geometrical arrangement of the fillers and the thermal properties of their constituents. There are many experimental as well as theoretical model studies on thermal conductivity of polymer composites in which the situation has been simplified by assuming that the particles are of specific shape and arranged in a particular geometry within the continuous medium. Dependence of the thermal properties of these materials on pore size, shape and packing of the material is also a matter of concern to engineers, mathematicians and physicists. As it is not often possible to conduct experiments, a theoretical expression is needed to predict its values. Due to the high geometrical complexity of actual polymer composites and the dearth of data available in the whole range make it difficult to predict effective properties of the materials. Designers of industrial components made of metal filled polymer composites are interested in easy to use methods for describing the behaviour of technological materials. A numerical approach for the estimation of the effective thermal conductivity, for metal particles embedded in the matrix of polymer was developed by Kumlutas and Tavman. A finite element technique is used to simulate the microstructure of composite materials for various filler concentrations at various ratios of thermal conductivities of filler to matrix material.

In the present study, efforts are based on the equivalent thermal resistors formed out of the phases in the form of parallel slabs and the resistor model approach has been applied. The slabs are taken to be inclined at an angle $\theta$ parallel to the direction of heat flow. The expression for $\theta$ has been obtained using data fitting technique by simulating the earlier results.

2 Theory

Following Hadley, closure equations for the temperature field can be written as:

$$\nabla <T> = \phi. <\nabla T_f> + (1-\phi). <\nabla T_c>$$  \hspace{1cm} \ldots (1)

$$\frac{\lambda_c}{\lambda_f} \nabla <T> = \phi. <\nabla T_f> + \frac{\lambda_c}{\lambda_f} (1-\phi). <\nabla T_c>$$  \hspace{1cm} \ldots (2)

where $<\nabla T_c>$ and $<\nabla T_f>$ are the average of the gradients in the continuous phase (fluid) and dispersed phase (filler), respectively and $<\nabla T>$ is the gradient of the overall average temperature. $\lambda_c$ and $\lambda_f$ are the thermal conductivities of continuous phase and...
filler phase, respectively and \( \phi \) is the volume fraction of the continuous phase. The Eqs (1) and (2) contain three constants \( \nabla T_f \), \( \nabla T_c \) and cannot be solved unless some relation connecting the constants be assumed. For this we assume

\[
\nabla T_f = \nabla T_c
\]

i.e. average temperature gradients in the two phases are equal. This condition is met in a collection of phase slabs, parallel to the direction of heat flow and gives:

\[
\lambda = \phi \lambda_f + (1 - \phi) \lambda_c \quad \ldots (3)
\]

This is an expression for equivalent thermal conductivity of resistors arranged in parallel. Similarly, the assumption:

\[
\nabla T_f = \frac{\lambda_c}{\lambda_f} \nabla T_c
\]

gives series arrangement

\[
\lambda = \frac{\lambda \lambda_f}{\phi \lambda_c + (1 - \phi) \lambda_f} \quad \ldots (4)
\]

It is an expression for equivalent thermal conductivity of resistors arranged perpendicular to the heat flow. The above condition is equivalent to \( \lambda_c \nabla T_c = \lambda_f \nabla T_f \), i.e. the heat flux passing through different phases is the same. It is a situation met with the slabs perpendicular to the direction of heat flow.

We know that a porous medium is neither composed of slabs parallel to the heat flux nor perpendicular to it, yet the concept of the slabs is capable of predicting the maximum and minimum limits of the ETC. Therefore, it is proposed that the slabs of the continuous and dispersed phases, inclined at an angle \( \theta \) with the direction of heat flux may represent the ETC of the system. As Eqs (3) and (4) do not predict the ETC of real two-phase systems correctly, a different kind of weighted geometric mean is proposed for effective thermal conductivity as follows:

\[
\lambda_c = \lambda \sin^2 \theta + \lambda \sin^{-2} \theta \quad \ldots (5)
\]

Depending on the value of \( \theta \), the forms of the combined thermal conductivity given by Eqs (3)-(5) are easily realizable. Setting \( \theta = 90^\circ \), we get results in parallel arrangement, while using \( \theta = 0 \) produces the series arrangement. Thus by knowing the angle of inclination of the slabs \( \theta \), the ETC of a two-phase system can be obtained. Therefore, Eq. (5) is solved for \( \theta \) in terms of \( \phi, \lambda_c, \lambda_f \) and \( \lambda_e \). The solution is:

\[
\theta = \sin^{-1} \left[ \frac{\ln \left( \frac{\lambda_c}{\lambda_f} + (1 - \phi) \frac{\lambda_e}{\lambda_c} \right)}{\ln \left( 1 + \phi (1 - \phi) \frac{\lambda_c}{\lambda_f} \right) - \frac{\lambda_c}{\lambda_f} - 2} \right] \quad \ldots (6)
\]

The experimental results show that ETC depends upon various characteristics of the system. The most prominent amongst them are non-uniform shape of the particles, random packing of the phases and non-uniform flow of heat flux lines in the phases. Thus for practical utilization of Eq. (5), we have to calculate the value of angle \( \theta \).

3 Results and Discussion

Angle \( \theta \) is calculated for large number of samples reported earlier\(^ {1,2,6-8}\) by putting the values of thermal conductivity of constituent phases and ETC in Eq. (6). A graph has been plotted between \( \sin^2 \theta \) and \( X = \phi \ln \left( \frac{\lambda_c}{\lambda_f} \right) \). It is found that \( \sin^2 \theta \) increases linearly with increasing \( \phi \ln \left( \frac{\lambda_c}{\lambda_f} \right) \). The expression:

\[
\sin^2 \theta = C_1 X + C_2 \quad \ldots (7)
\]

best fitted the curves obtained from the plot. Here \( C_1 \) and \( C_2 \) are constants. The values of these constants are different for different kinds of materials. The variation of these constants with the ratio \( \lambda_c/\lambda_f \) is shown in Fig. 1. It is observed that \( C_1 \) (upper line) and \( C_2 \) (lower line) increase slightly as the ratio \( \lambda_c/\lambda_f \) increases.

![Fig. 1 — Variation of \( C_1 \) and \( C_2 \) with the ratio \( \lambda_c/\lambda_f \)]](image-url)
The value of $\theta$ obtained from Eq. (7) is substituted into Eq. (5) to calculate ETC for a large number of metal filled polymer composite materials reported earlier.

Tables 1-3 present a comparison of experimental results and calculated values of ETC from Eq. (5). The average percentage deviation from experimental results is 2.6% for HDPE/Al oxide systems as shown in the Table 1. Tables 2 and 3 give a comparison of experimental results of thermal conductivity for HDPE/alloy and HDPE/metal systems reported earlier and the values calculated using Eq. (5). It is observed that theoretical values obtained agreed well with the experimental results and the average percentage deviation from experimental results for HDPE/alloy and HDPE/metal systems are 18.7% and 8.1%, respectively. It is seen from the tables that the average percentage deviation from the experimental results is least for HDPE/Al oxide systems. In HDPE/alloy systems, the average percentage deviation is large from the experimental results. It may be due to
the non-homogeneity occurred in the alloy (filler phase).

In Figs 2-7, experimental results of the ETC for the same samples are shown and compared with computed values using Eq. (5) and with the Lewis and Nielsen’s model$^{14}$. It is found that ETC calculated using Eq. (5), gives closer results than Lewis and Nielsen’s model$^{14}$. Comparing our correlation using Eq. (5) with the experimental data, we observed that our model follows the curve of the data points very well as shown in Figs 2-7. This shows that for our model, the average percentage deviation is least as compared to the Lewis and Nielsen’s model$^{14}$.

4 Conclusions

The correlation presented here showed that the ETC strongly depends on the volume fractions and the ratio of thermal conductivity of the constituents. Other factors have small effect on the ETC. It has a single correlation factor, which is valid for metal filled polymer composites. The empirical model proposed here is capable of predicting ETC values closer to the experimental results for metal filled polymer composites. It should be noted that experimental data for the effective thermal conductivity of metal filled polymer composites with well-characterized microstructures having wide range of the volume fractions are still in short supply. It is expected that the experimentally validated model will be helpful in the evaluation of the ETC of composite materials for the whole range of the volume fractions of their constituents.

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Nomenclature
Al Aluminium
ETC Effective thermal conductivity [W m\(^{-1}\)k\(^{-1}\)]
HDPE High density polyethylene
\(<T>\) Average temperature
\(\nabla<T>\) Gradient of the average temperature

Greek symbols
\(\lambda\) Thermal conductivity [W m\(^{-1}\)k\(^{-1}\)]
\(\phi\) Volume fraction of the continuous phase
\(\theta\) Inclination angle

Subscripts and superscripts
\(e\) Effective
\(c\) Continuous phase (fluid)
\(f\) Dispersed phase (filler)
\(\perp\) Perpendicular
\(\parallel\) Parallel

References