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TRANSVERSE THERMAL CONDUCTIVITY OF FIBER REINFORCED POLYMER COMPOSITES

I.H.Tavman, H. Akıncı
Mechanical Engineering Department
Dokuz Eylül University, 35100 Izmir - Turkey

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ABSTRACT

Transverse thermal conductivity of high density polyethylene reinforced with chopped strand glass fiber mat is investigated experimentally for temperatures ranging from 10°C to 85°C. Models predicting the transverse thermal conductivity of composites filled with long fibers are stated and are compared with each other and with experimental results. Samples are prepared by compression molding process, sandwiching layers of glass fibers between layers of polyethylene. A modified hot wire technique is used to measure thermal conductivity. © 2000 Elsevier Science Ltd

Introduction

A polymer composite is a material formed by dispersing reinforcing or filling particles or fibers in a polymeric matrix. Unlike the case of polymer blends and alloys, the components of a composite material maintain the physical properties that they display in isolation from each other. However the physical properties of the composite may be significantly different from those of its components and in the case of fiber reinforced composites the physical properties may be anisotropic depending on fiber orientation. The determination of the effective properties of composite materials is of great importance in effective design and application of composite materials. There are numerous theoretical, empirical, as well as numerical methods to predict effective thermal conductivity of composites, each of these methods have certain assumptions, therefore they may be applicable for certain specific cases and ranges. It is therefore necessary to have experimental data on each type of composite materials.

Reinforced and filled polymer composites are being widely used in electronic systems due to

their ease of manufacturability, light weight, tailorable and convenient physical properties. During operation electronic systems produce a lot of heat that must be dissipated in order to keep the elements at a temperature suitable for their reliable operation. An increase in operating temperature of about 10°C reduces the mean lifetime of the element by a factor of two [1]. Therefore, the thermal performance of molded polymer packages is very important for the right operation of electronic systems. Due to the increasing use of composite materials in the electronic industry, there is a renewed interest in theoretical and experimental determination of the effective thermal conductivity of fiber and particle filled polymer composites. The main disadvantage of pure polymer materials is their low thermal conductivity, ranging from 0.15 W/m.K for PVC to 0.44 W/m.K for HDPE, generally the reinforcing or filling materials have thermal conductivities several order of magnitude higher. Thus, apart from improving mechanical properties, the role of fillers is to enhance heat transfer properties. Although the thermal conductivity of particle filled polymers has been investigated theoretically and experimentally by many researchers [2-14], there are few studies on thermal conductivity of fiber filled polymers [15-18].

Thermal Conductivity Models For Fiber Filled Polymers

The simplest models are the series and parallel models where different components of the composite are arranged in layers series or parallel to heat flow. These two models give the lower and the upper bounds of the effective thermal conductivity.

For the series model:

$$k_c = \frac{k_p k_f}{k_p (\phi) + k_f (1 - \phi)} \quad (1)$$

For the parallel model:

$$k_c = \phi \cdot k_f + (1 - \phi) \cdot k_p \quad (2)$$

Where, k_c , k_p and k_f are respectively the thermal conductivities of the composite, polymer matrix and filler materials, and ϕ are the volume proportion of filler.

For the geometric mean model:

$$k_c = k_f^\phi \cdot k_p^{(1-\phi)} \quad (3)$$

The semi-theoretical model developed by Springer and Tsai [19] assumes a square distribution of cylindrical fibers in the matrix material. The estimation of the thermal conductivity in the direction normal to the fibers is based on the analogy between the response of the composite to shear loading and heat transfer.

$$k_c = k_p \left[1 - 2\sqrt{\frac{\phi}{\pi}} + \frac{1}{B} \left(\pi - \frac{4}{\sqrt{1 - (B^2\phi/\pi)}} \tan^{-1} \left(\frac{\sqrt{1 - (B^2\phi/\pi)}}{1 + B\sqrt{\phi/\pi}} \right) \right) \right] \quad (4)$$

Where, $B = 2 \left(\frac{k_p}{k_f} - 1 \right)$

Rayleigh [20] analyzed the influence of obstacles arranged in rectangular order upon the properties of the medium.

$$k_c = k_p \left[1 - \frac{2\phi}{\gamma + \phi - \frac{C_1}{\gamma} \phi^4 - \frac{C_2}{\gamma} \phi^8} \right] \quad (5)$$

With $C_1 = 0.3058$, $C_2 = 0.0134$, and $\gamma = \frac{(k_p/k_f) + 1}{(k_p/k_f) - 1}$

Assuming a parabolic distribution of the discontinuous phase Cheng and Vachon [21] developed a model applicable for spherical particles as well as for fiber filled composites. For the case $k_f > k_p$, the thermal conductivity of the composite is given by:

$$\frac{1}{k_c} = \frac{1}{\sqrt{C.(k_p - k_f)(k_p + B.(k_f - k_p))}} \ln \frac{\sqrt{k_p + B.(k_f - k_p)} + B/2\sqrt{C.(k_p - k_f)}}{\sqrt{k_p + B.(k_f - k_p)} - B/2\sqrt{C.(k_p - k_f)}} + \frac{1 - B}{k_p} \quad (6)$$

Where, $B = \sqrt{\frac{3.\phi}{2}}$, $C = -4.\sqrt{\frac{2}{3.\phi}}$

If the thermal conductivity of the polymer matrix is much smaller than the thermal conductivity of the filler material, $k_p \ll k_f$ or $k_f/k_p > 100$, as long as $\phi < 0.667$, effective thermal conductivity of the composite may be approximated by the second term of equation (6):

$$k_c \approx \frac{k_p}{1 - B} \quad (7)$$

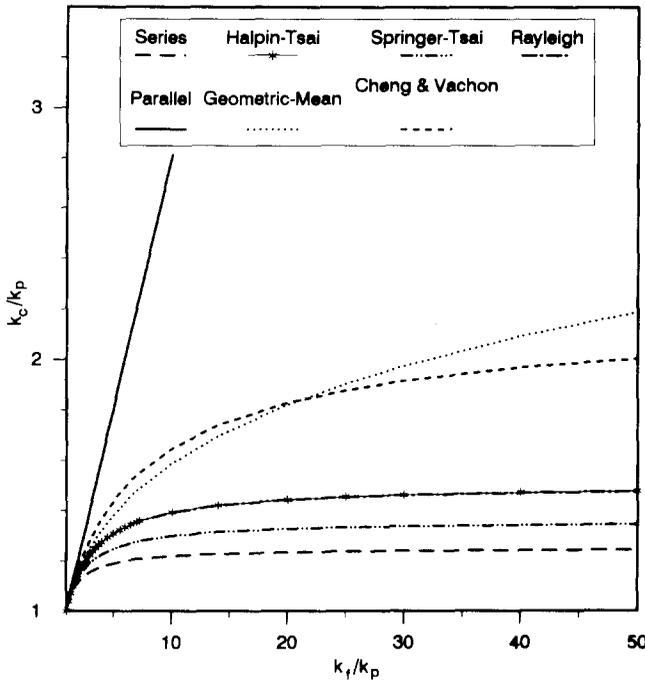


FIG. 1

Comparison of the models predicting effective thermal conductivity of fiber reinforced composite materials, for constant fiber filler content of 20% by volume and $1 \leq k_f/k_p \leq 50$.

Halpin-Tsai [22] derived a theoretical model for the transverse thermal conductivity using the analogy between in-plane field equation and boundary conditions to the transverse transport coefficients.

$$k_c = k_p \left[\frac{1 + \xi \phi \eta}{1 - \eta \phi} \right] \tag{8}$$

Where,

$$\eta = \frac{k_f/k_p - 1}{k_f/k_p + \xi} \tag{9}$$

For platelets of width “a” and thickness “b”: $\xi = \sqrt{3} \log(a/b)$, and for circular or square fibers $\xi = 1$.

In figure 1, thermal conductivity values predicted by the models are compared between each other for a constant fiber filler content of 20% by volume. It may be noticed that the Halpin-Tsai

with $\xi = 1$ and the Rayleigh models estimate very near thermal conductivity values. It may also be seen from this figure that for high values of k_f/k_p , all the models, except for the parallel and the geometric mean models, show an asymptotic behavior. This means that for sufficiently high values of k_f/k_p , the effective thermal conductivity of the composite is only a function of filler content, but not a function of the thermal conductivity of the filler material.

Sample Preparation and Measurement Technique

The matrix material is a blow molding high density polyethylene in granular form, with a density of 0.964 g/cm^3 , melt index 0.35 g/10 min. and measured thermal conductivity 0.50 W/m.K at 27°C . Glass fibers are in the form of mat laminates, distributed randomly in the direction perpendicular to heat flow. The solid density of glass fibers is 2.5 g/cm^3 and its thermal conductivity is 1.03 W/m.K in the temperature range under consideration. Thus, the thermal conductivity of glass fiber reinforcement is about two times that of the matrix material.

Samples, which are rectangular in shape, with 105 mm in length, 58 mm in width and 13.5 mm . thick, are prepared by mold compression. First, high density polyethylene plates of 1 mm . in thickness are prepared by compressing HDPE granules under about 200 kg/cm^2 pressure at 180°C . Pure HDPE sample is prepared by piling up layers of plates prepared and melting it under the same pressure and temperature. Whereas, the two composite samples, containing respectively 14% and 27% glass mat by volume, are prepared by putting layers of glass fibers between layers of HDPE and compressing in a dye under about 200 kg/cm^2 pressure at 180°C and maintaining it for 10 minutes at this pressure and temperature. As the HDPE melts at 130°C , the matrix material infiltrates in the glass mat to constitute a solid composite material. Both the HDPE and glass mat layers are weight with a precision of 0.01 gr. and the respective volume percents are calculated using their respective densities.

Thermal conductivity measurements of pure high-density polyethylene as well as glass fiber filled HDPE composites are carried out with the Shotherm QTM thermal conductivity meter. The instrument uses a modified hot wire technique to measure thermal conductivity. A thin straight wire through which a constant electric current is passed generating constant heat (Q) per unit length of wire, per unit time, is placed between two rectangular shaped materials, the first

one is an insulating material of known thermal properties which is a part of the measuring probe and the second one is the sample for which the thermal conductivity has to be measured. A constant power is supplied to the heater element and the temperature rise ΔT of the heating wire is measured by a thermocouple and recorded with respect to time during a short heating interval, the thermal conductivity (k) of the sample is calculated from the temperature-time (ΔT - Δt) record and power input (Q). The details about the working principal are explained in detail in an earlier publication by Tavman [23]. By this method, the thermal conductivity is measured with an accuracy of $\pm 5\%$ and reproducibility of $\pm 2\%$. For each specimen the thermal conductivity is measured five times and the mean values are reported.

Results and Discussion

The thermal conductivity of the matrix material was measured within the temperature range 14°C to 84°C , but the solid thermal conductivity of glass fiber was taken from the literature [18], and assumed to be constant in the narrow temperature range under consideration.

The experimental results for pure high density polyethylene as well as composites with 14% and 27% reinforced with glass mat were given in figures 2 and 3, in the temperature range from about 10° to 85°C . As the measurement range studied was well above the glass transition temperature which is -110°C for high density polyethylene, thermal conductivity decreased with increasing temperature, in accordance with other literature values.

The measured thermal conductivity values for glass fiber mat reinforced composites were compared with those calculated from Rayleigh, parallel, Springer & Tsai, Halpin-Tsai with $\xi = 1$, series and Cheng & Vachon models, in figures 2 and 3. As the values estimated by the geometric mean model were very close to those calculated by the Rayleigh model, this model was not plotted in these figures. From these figures, it may be seen that the parallel model was not adequate for estimating transverse thermal conductivity of polymer composites with fibers dispersed in the direction of heat flow and gave values higher than 10% in most cases. All other models predicted transverse thermal conductivity within $\pm 5\%$, which was typically the expected experimental error range. However, Rayleigh, geometric mean, Springer & Tsai, Halpin-Tsai models predictions were within $\pm 2\%$, they may be considered as best suited for predicting

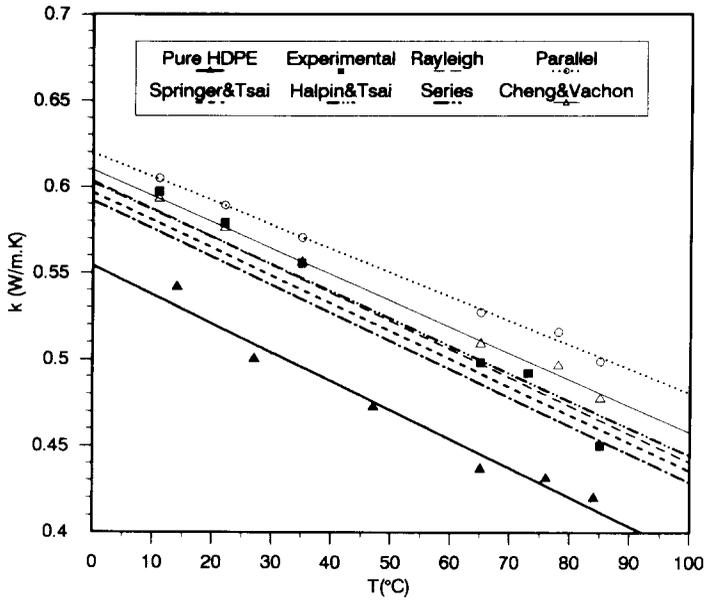


FIG. 2

Measured and calculated thermal conductivity of HDPE + 14% glass mat as a function of temperature.

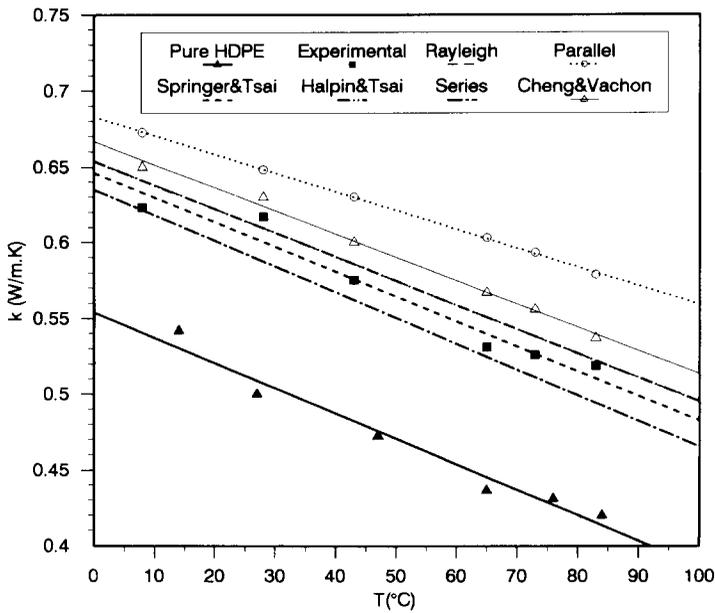


FIG. 3

Measured and calculated thermal conductivity of HDPE + 27% glass mat as a function of temperature.

transverse thermal conductivity. More measurements with different percentages of reinforcing material and with different materials, especially with materials with higher k_f/k_p ratios, have to be performed in order to be able to generalize with model is best suited for predicting transverse thermal conductivity.

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Nomenclature

- k_c thermal conductivity of the composite, W/m.K
 k_f thermal conductivity of the filler materials, W/m.K
 k_p thermal conductivity of the polymer matrix, W/m.K
 ϕ volume proportion of fiber filler.
 ξ shape factor for Halpin-Tsai model

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