Proceedings of the
International Conference on Mechanical Engineering 2011
(ICME2011) 18-20 December 2011, Dhaka, Bangladesh

ICME11-TH-002

EFFECT OF FILLER SIZE AND CONTENT ON THERMAL CONDUCTIVITY OF SGM FILLED EPOXY COMPOSITES

D. Mishra and A. Satapathy

Department of Mechanical Engineering, National Institute of Technology, Rourkela ,India

ABSTRACT

This paper describes the preparation and thermal conductivity characterization of solid glass micro-sphere (SGMs) filled polymer composites. Composite samples are prepared by embedding SGMs of different sizes in the epoxy resin. Three-dimensional spheres-in-cube lattice array models are constructed using finite element method (FEM) to simulate the microstructure of composite materials for various SGM content ranging from 0 to about 35 vol %. Finally, guarded heat flow meter test method is used to measure the conductivity of these composites. The simulations are compared with K_{eff} values obtained from experiments as well as other theoretical models and it is found that the FEM simulations and K_{eff} values of the theoretical model are fairly close to the measured K_{eff}. The incorporation of SGMs thereby results in reduction of conductivity of epoxy resin and thereby improves its thermal insulation capability. Further, the size and content of SGMs influence the extent of reduction of K_{eff}.

Keywords: Composites, Glass Microspheres, Thermal Conductivity.

1. INTRODUCTION

It is quite important, to understand the mechanisms of heat transfer in polymer composites. Many theoretical and empirical models have been proposed to predict the effective thermal conductivity of two-phase mixtures. Lewis and Nielsen, 2003 [1] derived a semi-theoretical model by modification of the Halpin-Tsai equation for a two-phase system. For an infinitely dilute composite of spherical filler particles, the exact expression for the effective thermal conductivity is given by Maxwell, 1873 [2]. The objectives of this work include fabrication of a new class of composites to further improve the insulating properties of epoxy by the incorporation of solid glass micro-spheres.

The heat transfer process of porous materials is very complicated, especially for polymer composites. It is quite important, therefore, to understand the mechanisms of heat transfer in polymer composites, which are potential insulating materials. Foamed plastic is a polymeric material commonly used as thermal insulation. But its application is limited considerably due to its poor mechanical properties. There is, therefore, a focus on fabricating a kind of reinforced polymeric system which is light but has better mechanical strength and good thermal insulation properties. In this context, rigid glass micro-spheres (glass beads) have some advantages as fillers in polymers such as low thermal conductivity, coefficient and density. In addition, these micro-particles do not generate stress concentration in the interface between the fillers and the matrix owing to their smooth spherical surface. This type of composites can be applied in building materials, space flight and aviation industry. Glass beads are preferred as fillers especially when composite properties such as isotropy or low melt viscosity are important. Moreover, the orientation effects associated to molding are minimal. There are only a few published papers on evaluation of effective thermal conductivity of polymer composites filled with glass beads [3-6]. Liang and Li,2006 [3] reported on measurement of thermal conductivity of hollow glass-bead-filled polypropylene composites. Recently, they in 2007 [4] also made two-dimensional and three-dimensional finite element analysis on the heat transfer and simulated the variation of effective thermal conductivity of hollow glass microsphere filled polymer composites. Liang and Li, 2007 [5] further studied the heat transfer in polymer composites filled with hollow glass micro-spheres and proposed a theoretical model to predict the thermal conductivity of such composite system. Yung et al., 2009 [6] reported on the preparation and properties of hollow glass microsphere-filled epoxy matrix composites. But all these studies are for polymer composites filled with hollow glass spheres and surprisingly, there is no report available on evaluation of effective thermal conductivity of solid glass microsphere filled polymer composites. In view of the above, the present work is undertaken to evaluate theoretically and experimentally the thermal conductivity of epoxy matrix composites filled solid glass micro-spheres. Epoxy is chosen primarily because it happens to be the most commonly used polymer and because of its low value of thermal conductivity (about 0.363 W/m-K).

2. MATHEMATICAL MODEL

The theoritical analysis of the heat transfer in this paper is based on the following suppositions: (a) the distribution or dispersion of the solid micro-spheres in the polymer matrix is uniform; (b) the temperature distribution along the direction of heat flow is linear. An element from the composite is selected for analysis which is a straight cube with side length of H and there is a solid glass micro-sphere of radius (r) in the element. The element is divided into polymer phase and micro-sphere shell phase.The heat quantity Q transfers from bottom to the top. The heat transport in solid glass microsphere filled polymer composites has three mechanisms: (i) solid thermal conduction and (ii) heat radiation on the surface between neighbouring particles. Polymer composite works usually under lower temperature conditions where the proportion of the thermal radiation in the total heat transfer is very small, hence the thermal radiation is neglected.

Part I:

$$k_1 = k_p \tag{1}$$

Part II: Taking a thin piece with thickness of dy, according to Fourier's theorem, k_2 is given by:

$$k_2 = \frac{Q_p + Q_g}{\left(\frac{dT}{dy}\right)S} = k_p \frac{Sp}{S} + k_g \frac{S_g}{S} \tag{2}$$

where, T is the temperature, S is the area of whole cross-section. k_p, k_g are the thermal conductivities of polymer matrix phase and micro-sphere phase. S_p and S_g are the cross-sectional areas of the polymer and micro-sphere. Q_p and Q_g are the heat quantities through the polymer matrix and micro-spheres respectively.

Because of the linear distribution of temperature, the average thermal conductivity of each section may first be obtained. Because of the linear distribution of temperature, the average thermal conductivity of each section may first be obtained:

Part I:

$$k_1 = \int_{h_1} k_1\, dy/h_1) = k_p \tag{3}$$

Part II:

$$k_2 = (1/h_2)\int_{h_2} k_p \quad (S_p/S)\ k_g(\ S_g\ /S)\ dy$$

$$= (1/h_2 S)\,(k_p V_p + k_g V_g + k_a V_a) \tag{4}$$

Where V_p and V_g are the volumes of polymer matrix, micro-sphere shell and gas, respectively.

According to the series theorem of heat resistance, the effective thermal conductivity of composites, k_{eff} is given by

$$k_{eff} = \left(\frac{H}{RS}\right) = ((H/(R_1 + R_2)S) \tag{5}$$

Finally, the expression for effective thermal conductivity of the composite is deduced as:

$$k_{eff} = \lfloor 1/k_p(1 - \frac{6vf}{\pi})^{1/3} + 2(k_p(\frac{4\pi}{3vf})^{1/3} + \pi\left(\frac{2vf}{9\pi}\right)^{1/3} \times (k_g \frac{\rho_s}{\rho_g} - k_p))^{-1}\rfloor^{-1} \tag{6}$$

Here, k_p and k_g are the respective heat conductivities of the polymer and the micro-sphere phase, ρ_p and ρ_g are the effective densities of the polymer and the microsphere phase respectively, and v_f is the volume fraction of the filler i.e. the SGM in the composite.

3. EXPERIMENTAL DETAILS

Low temperature curing Epoxy LY 556 resin, used as the matrix material and the hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. Epoxy is chosen primarily of its low density (1.1 gm/cc) and low value of thermal conductivity (0.363 W/m K). Solid glass micro-spheres (SGM) of three different sizes (100, 200 and 300 micron diameter) are reinforced in the resin to prepare the composites. The dough (epoxy filled with SGM) is then slowly decanted into the glass molds, coated beforehand with wax and a uniform thin film of silicone-releasing agent. The composites are cast in these molds so as to get disc type cylindrical specimens (dia 20 mm, thickness 5 mm). Composites of 8 different compositions (0, 5, 10, 15, 20, 25, 30 and 35 vol % of SGM respectively) using SGMs of each size are made. The castings are left to cure at room temperature for about 24 hours after which the glass molds are broken and samples are released.

Unitherm™ Model 2022 is used to measure thermal conductivity of the composites fabricated for this investigation. This is an instrument based on guarded heat flow method and is used for a variety of materials. The test is carried out in accordance with ASTM E-1530 standard.

4. RESULTS AND DISCUSSION

4.1. Numerical analysis

Using the finite-element program ANSYS, thermal analysis is carried out for the conductive heat transfer through the composite body. In order to make this analysis, three-dimensional physical models with spheres-in-cube lattice arrays have been used to simulate the microstructure of composite materials for five different filler concentrations. Furthermore, the effective thermal conductivities of these epoxy composites filled with SGM up to about 35% by volume are numerically determined using ANSYS.

4.2. Description of the problem

The determination of effective properties of composites is of paramount importance for functional design and application of composite materials. One of the important factors that influence the effective properties and can be controlled to an appreciable extent is the microstructure of the composite. Here, microstructure means the shape, size distribution, spatial distribution and orientation distribution of the reinforcing inclusion in the matrix. Although most composite possess inclusion of random distributions, great insight of the effect of microstructure on the effective properties can be gained from the investigation of composites with periodic structure. System with periodic structures can be more easily analyzed because of the high degree of symmetry embedded in the system.

A typical periodic arrangement of solid glass micro-spheres within the epoxy body is schematically shown in Fig. 1. Fig. 2 clearly illustrates the heat flow direction and the boundary conditions for the particulate-polymer composite body considered for the analysis of this conduction problem. The temperature at the nodes along the surfaces ABCD is prescribed as T_1 ($=100^0$C) and the ambient convective heat transfer coefficient is assumed to be 25 W/m^2-K at room temperature of 27°C. The other surfaces parallel to the direction of the heat flow are all assumed adiabatic. The temperatures at the nodes in the interior region and on the other boundaries are unknown. These temperatures are obtained with the help of the finite-element program package ANSYS. In this analysis it is assumed that the composites are macroscopically homogeneous, locally both the matrix and filler are homogeneous and isotropic, the thermal contact resistance between the filler and the matrix is negligible and the composite lamina is free from voids. The problem is based on 3D physical model and the filler arranged in a square periodic array are assumed to be uniformly distributed in the matrix.

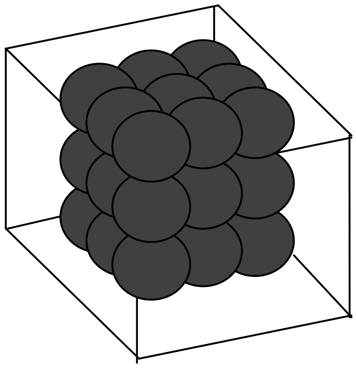

Fig 1. Schematic diagram showing a typical arrangement of SGM within the epoxy body

Thermal conductivities of these SGM-epoxy composites are numerically estimated by using the spheres-in-cube model. A typical 3-D model showing arrangement of spherical fillers with a particle concentration of 1.4 vol% within the cube shaped matrix body is illustrated in Fig.3. The temperature profiles obtained from FEA analysis for the composites with particulate concentrations of 1.4, 3.4, 6.5, 11.3 and 17.9 vol % are presented in Figs 3a, 3b, 3c, 3d and 3e respectively.

The simulated values of effective thermal conductivity of the composites obtained from FEA are presented in Table 1 along with the corresponding measured values.

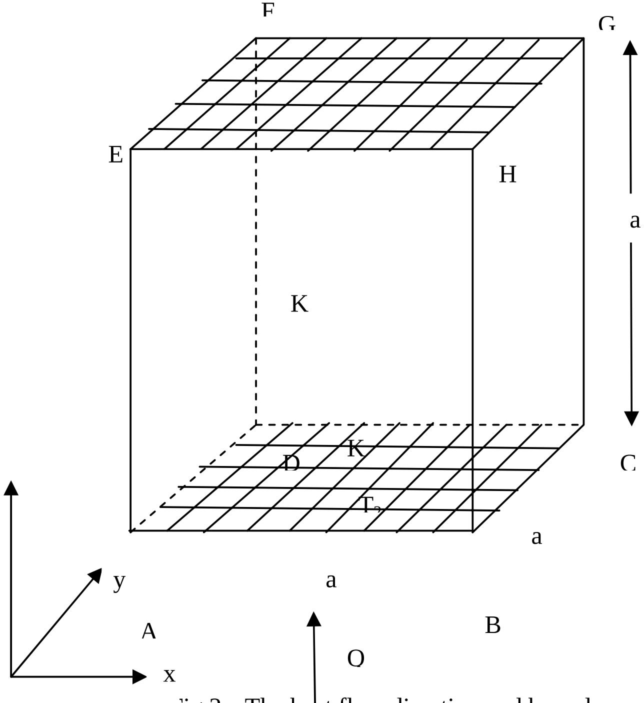

Fig 2a. The heat flow direction and boundary conditions for the particulate-polymer composite

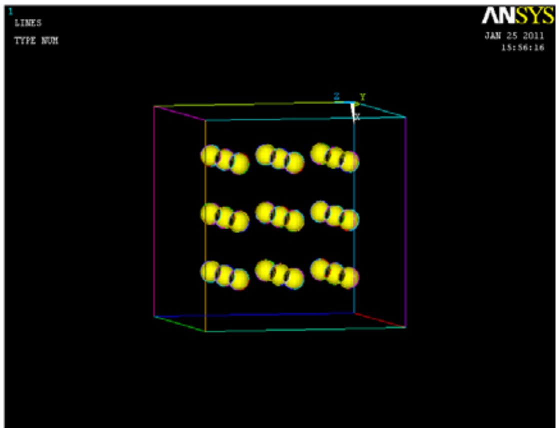

Fig 2. A typical 3-D spheres-in-cube model with filler concentration of 1.4 vol %

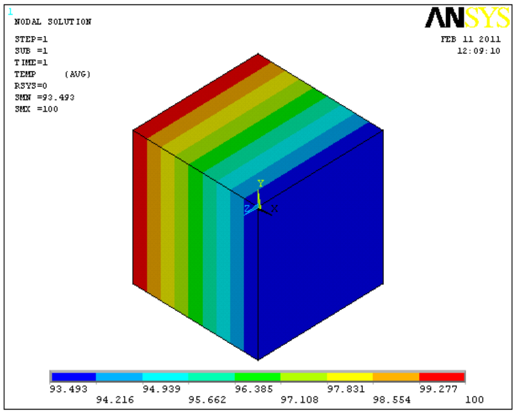

Fig 3a. Temperature profile for epoxy-SGM composite with filler concentration of 1.4 vol %

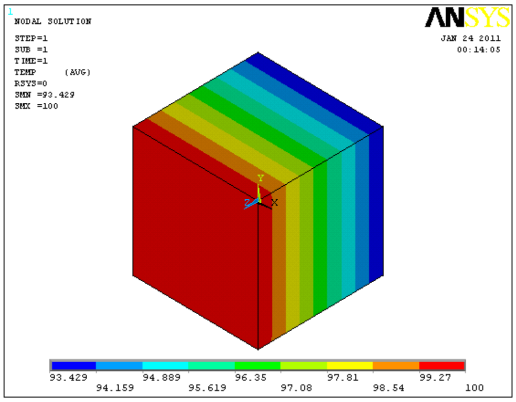

Fig 3b. Temperature profile for epoxy-SGM composite with filler concentration of 3.4 vol %

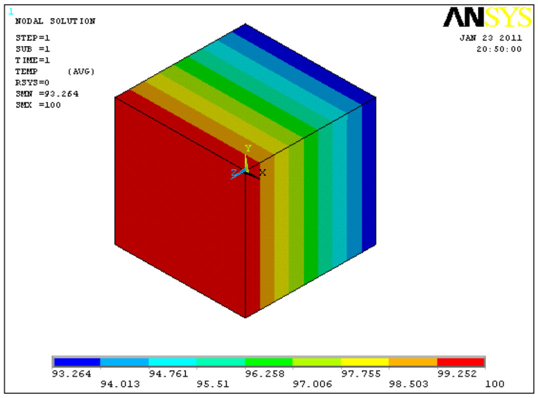

Fig 3c. Temperature profile for epoxy-SGM composite with filler concentration of 6.5 vol %

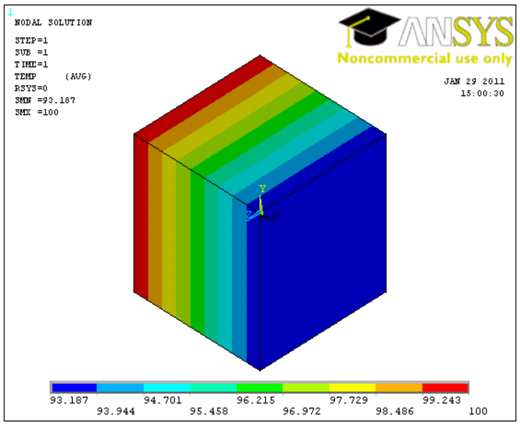

Fig 3d. Temperature profile for epoxy-SGM composite with filler concentration of 11.3 vol %

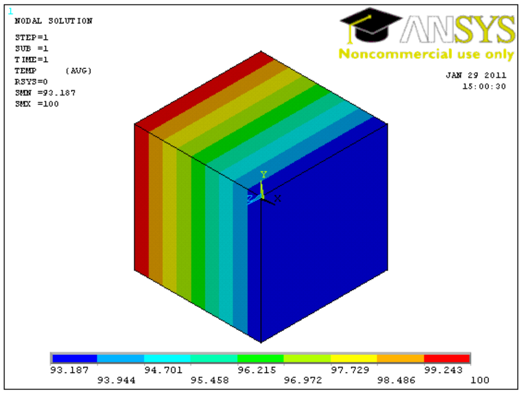

Fig 3e. Temperature profile for epoxy-SGM composite with filler concentration of 17.9 vol %

Sample	SGB Content (vol %)	Effective thermal conductivity of composites K_{eff} (W/m K)		
		FEA Results	Experimental value	Percentage errors
1	0	----	0.363	----
2	1.4	0.2862	0.293	0.7
3	3.4	0.2568	0.267	1.1
4	6.5	0.2283	0.237	0.9
5	11.3	0.2195	0.232	1.3
6	17.9	0.2145	0.229	1.5

Table 1. Thermal conductivity values for composites obtained from FEA, Experiment and the associated percentage errors

It is noticed that the results obtained from the finite-element analysis taking sphere-in-cube composite model are reasonably closer to the measured values of effective thermal conductivity for composites of different filler content. The percentage errors associated with the FEA values with respect to the experimental values is also given in Table 1. It is seen from this table that the errors associated with sphere-in-cube model simulations lie in the range 0-2 %. On comparing, it is further noticed that FEA underestimates the value of thermal conductivity, with respect to the experimental ones. However, it leads to a conclusion that for a particulate filled composite of this kind the finite element analysis can very well be used for predictive purpose in determining the effective thermal conductivity for a wide range of particle concentration.

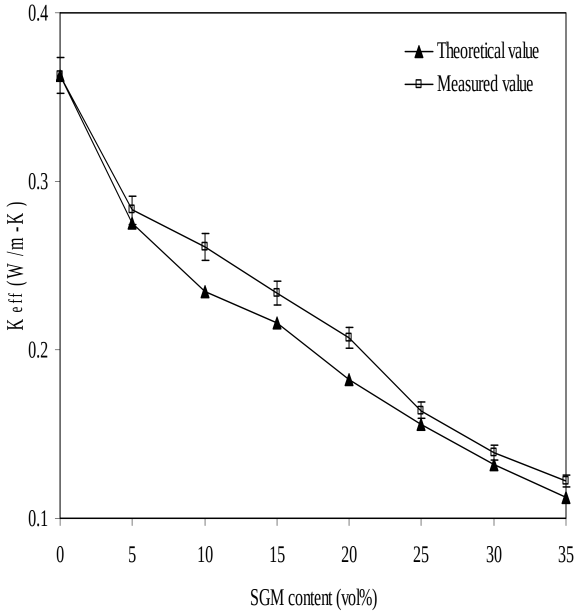

Fig 4. Variation of effective thermal conductivity with SGM content: Comparison of theoretical and measured values

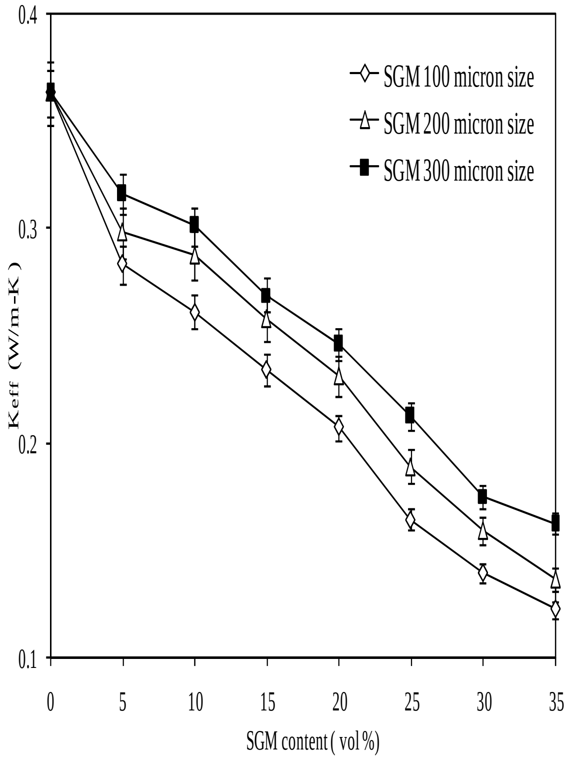

Fig 5. Variation of effective thermal conductivity with SGM content: Effect of SGM size

Figs. 4 and 5 present the variation of effective thermal conductivity (both simulated as well as measured) as a function of the SGM content and size respectively.

The difference between the simulated values and the measured value of conductivity may be attributed to the fact that some of the assumptions taken for the numerical analysis are not real. The distribution of SGM in the matrix body in the numerical analysis is assumed to be in an arranged manner, whereas in the fabricated composite sample, the glass beads are actually dispersed in the resin almost randomly. However, it is encouraging to note that the incorporation of SGM results in significant drop in thermal conductivity of epoxy resin. With addition of 1.4 vol. % of SGM, the thermal conductivity decreases by about 19.283 % and with addition of 17.9 vol.% of SGM the thermal conductivity decreases by about 36.914% when compared with neat epoxy resin.

5. CONCLUSIONS

Successful fabrication of epoxy based composites filled with solid glass micro-spheres by hand-lay-up technique is possible. Incorporation of SGMs results in reduction of thermal conductivity of epoxy resin and there by improves its thermal insulation capability. With addition of 35 vol% of SGM (100 micron size), the thermal conductivity decreases by about 66 % as compared to neat epoxy resin. For same volume fraction of SGM in the composite, the improvement in composite insulation capability is found to be more for smaller SGMs. With light weight and improved insulation capability SGM filled epoxy composite can be used for applications such as electronic packages, insulation board, food container, thermo flasks, building materials, space flight and aviation industry etc.

6. REFERENCES

1. Kumlutas, D. et.al. “Thermal Conductivity of particle filled polyethylene composite materials”, J. Compos.Sci. Technol. 63(1) (2003) 113-117.
2. Maxwell, J., “Electricity and magnetism”, Oxford, Clarendon, 1873.
3. Liang, J.Z., Li, F.H., “Measurement of thermal conductivity of hollow glass bead filled polypropylene composites”, Polym. Test. 25(4) (2006) 527-531
4. Liang, J.Z., Li, F.H., “Simulation of heat transfer in hollow glass-bead- filled polypropylene composites by finite element method”, Polym.Test.26(3)(2007) 419-424.
5. Liang, J.Z., Li, F.H., “Heat transfer in polymer composites filled with inorganic hollow micro-spheres: A theoretical model”, Polym. Test. 26 (2007) 1025-1030
6. Yung, K.C., Zhu, B.L., Yue, T.M., Xie, C.S., “ Preparation and properties of hollow glass microsphere- filled epoxy- matrix composites” Comp. Sci. Technol.(69) (2009) 260 -264

7. NOMENCLATURE

Symbol	Meaning	Unit
R	Radius of the spherical glass bead	Micro-metre
K_p	Thermal conductivities of polymer matrix	Watt/m K
K_g	Thermal conductivity of micro-sphere shell phase	Watt/m K
Q	Heat quantity	Joule
Q_p	Heat quantity through the polymer matrix	Joule
Q_g	Heat quantity through micro-sphere shell	Joule
A	Total area of cross-section (heat transfer area)	Square metre
A_g	Cross-sectional area of the micro-sphere shell	Square metre
A_p	Cross-sectional area of the polymer.	
T :	Temperature	Kelvin