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Processing and Thermal Conductivity Characterization of Solid Glass Micro-Spheres Filled Polymer Composites

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Abstract
This paper describes the preparation and thermal conductivity characterization of solid glass micro-spheres (SGMs) filled polymer composites. SGMs of different sizes are embedded in epoxy resin to develop composites by hand layup technique. A numerical simulation of the heat-transfer within the composites is made by using finite element method (FEM). Three-dimensional spheres-in-cube lattice array models are constructed to simulate the microstructure of composite materials for various SGM content ranging from 0 to about 27 vol % and the effective thermal conductivities (K_{eff}) of the composites are estimated. K_{eff} values are also calculated using some of the existing theoretical models. 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 and it is found that the FEM simulations are fairly close to the measured K_{eff}. This study shows that the incorporation of SGMs 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; FEM; Thermal Conductivity; Simulation*

Introduction
The heat transfer process in porous materials is very complicated, especially for polymer composites filled with micro-spheres. There are only a few published papers on evaluation of effective thermal conductivity of polymer composites filled with micro-sized glass beads. Liang and Li [1] reported on measurement of thermal conductivity of hollow glass-bead-filled polypropylene composites. Recently, they [2] 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 [3] 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 systems. Yung et al [4] have also reported 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 the thermal conductivity of epoxy filled with solid glass micro-spheres both experimentally as well as numerically using FEM.

Thermal Conductivity Models

Many theoretical and empirical models have also been proposed to predict the effective thermal conductivity of two-phase mixtures. Lewis and Nielsen [5] derived a semi-theoretical model by a modification of the Halpin-Tsai equation for a two-phase system:

$$k_c = k_m[(1 + AB\phi)/(1 - B\phi\psi)]$$

Where, $B = \frac{\left\{\left(\frac{k_f}{k_m}\right) - 1\right\}}{\left\{\frac{k_f}{k_m} + A\right\}}$ $\qquad \psi = 1 + \{(1 - \phi_m)/\phi_m^2\}\phi$

For an infinitely dilute composite of spherical filler particles, the exact expression for the effective thermal conductivity is given by Maxwell [6] as:

$$\frac{k}{k_c} = 1 + 3\left(\frac{k_d - k_c}{k_d + 2k_c}\right)$$

where k, k_c and k_d are thermal conductivities of composite, continuous-phase (matrix), and dispersed-phase (filler), respectively and Ø is the volume fraction of the dispersed-phase. Russell obtained the conductivity using a series parallel network as

$$k_c = k_m[\Phi^{\frac{2}{3}} + \frac{k_m}{k_f(1-\Phi^{2/3})}]/[\Phi^{\frac{2}{3}} - \Phi + \frac{k_m}{k_f}(1 + \Phi - \Phi^{2/3}]$$

Experimental Details and Numerical Methodology

Spherical glass microspheres of 100 and 200 micron size supplied by Glass Bead Industries India Ltd. are reinforced in epoxy resin to prepare the composites. This low temperature curing LY 556 epoxy resin and the corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. The dough (epoxy filled with SGM) is then slowly decanted into the glass molds, coated beforehand with wax. The composites are cast in these molds so as to get disc type cylindrical specimens (dia 25 mm, thickness 5 mm). Composites of different compositions (0, 1.4, 3.4, 6.5, 11.3 and 17.9 vol % of SGM for 100 micron size and (0, 6.72, 11.3 and 26.8 vol % of SGM for 200 micron size respectively) are made. The castings are left to cure at room temperature for 24 hours after which the molds are broken and samples are released.

Unitherm™ Model 2022 is used to measure thermal conductivity of a variety of materials. The tests are conducted in accordance with ASTM E-1530 standard. 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 different filler concentrations.

Results and discussion

Fig. 1 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 surface ABCD is prescribed as 100^0C and the ambient convective heat transfer coefficient is assumed to be 25 W/m^2-K at a room temperature of 27°C. The other surfaces parallel to the heat flow direction are all assumed adiabatic. The unknown temperatures at the interior nodes and on the other boundaries are obtained with the help of ANSYS.

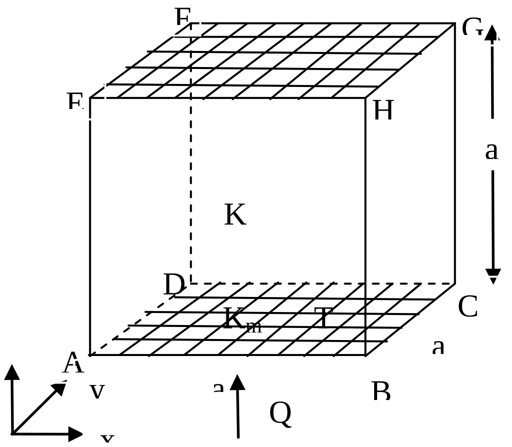

Fig .1 . The heat flow direction and boundary conditions

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 spheres-in-cube 3D physical model. The solid glass microspheres are assumed to be uniformly distributed in the matrix. K_{eff} of these SGM-epoxy composites are then numerically estimated.

It is noticed that the results obtained from the finite-element analysis are reasonably closer to the measured values, rather it underestimates the value of effective thermal conductivity for composites of different filler content. 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.

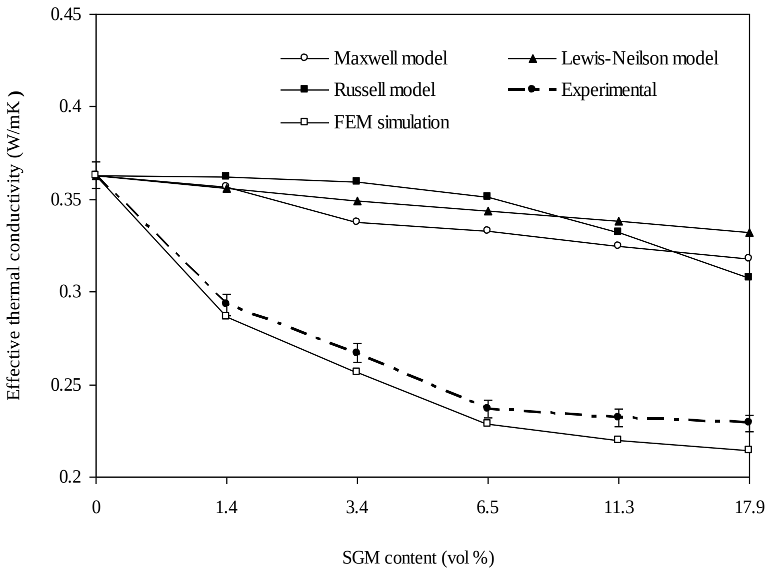

Fig. 2 Variation of thermal conductivity with SGM content

Fig. 2 presents the variation of effective thermal conductivity (both simulated as well as measured) as a function of the SGM content (100 micron) in the composites. The difference between the simulated values and the measured values of thermal conductivity may be attributed to the fact that some of the assumptions taken for the numerical analysis are not real. 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 (100 micron), the thermal conductivity of epoxy decreases by about 19.283 % and with addition of 17.9 vol.% of SGM the conductivity decreases by about 36.9 %. Similarly, with the addition of 11.3 vol % of SGM (200 micron), the thermal conductivity drops by 21.2 % while for SGM content of 26.8 vol% it decreases by 30.85%.

Conclusions

Successful fabrication of epoxy based composites filled with SGM by hand-lay-up technique is possible. The value of effective thermal conductivity obtained for various composite models using FEM are in reasonable agreement with the experimental values for a wide range of filler content. Incorporation of SGM results in reduction of thermal conductivity of epoxy resin and thereby improves its thermal 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.

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