An experimental investigation on the effect of particle size on the thermal properties and void content of Solid Glass Microsphere filled epoxy Composites

Debasmita Mishra1, Alok Satapathy2
1Department of Mechanical Engineering, VSSUT, Burla, Odisha, 768018
2Department of Mechanical Engineering, NIT, Rourkela, Odisha, 769008
E-mail - debasmita.mishra01@gmail.com

Abstract This paper investigates about the thermal characterization of Solid glass micro-sphere (SGM) filled epoxy composites. SGMs of different sizes are embedded in epoxy resin to fabricate composites by hand-layup technique. The composites for various SGM content ranging from 0 to about 35 vol % are thus fabricated and the effective thermal conductivities ($k_{eff}$) of the composites are estimated. The theoretical values are then compared with $k_{eff}$ values obtained from the experiment. This study shows that the incorporation of SGM results in an improvement in thermal insulation capability of the polymer. Further, the influence of size and content of SGMs in the extent of reduction of $k_{eff}$ was studied. Also, the effect of void content on improving insulation capability of the composites was analysed.

Keywords: Composites; Solid Glass Micro-spheres; Thermal Conductivity; Insulation

1. Introduction

Many parts of the country experience large changes in temperature from season to season. The demand for structurally stable, cost effective and light-weight insulation materials is therefore increasing day-by-day but the conventional engineering materials are unable to meet the requirements like high strength, low density and low conductivity. Foamed plastic is commonly used for thermal insulation. However, due to its poor mechanical properties, its application is considerably limited [1]. Therefore, there has been a focus to fabricate a kind of light, porous material with better mechanical strength and good thermal insulation properties. Against this backdrop, polymer composites are emerging as a class of promising engineering insulation materials.

Epoxy is chosen as the polymer to be used as the matrix material for this work [2]. Today, nearly 80% of the world die attach market for plastic encapsulated devices is solvent-less conductive epoxies. A few reasons why epoxies are widespread and common in electronic assemblies include: low temperature cure profiles which allow for greater availability of substrates and temperature sensitive
components; compliant nature of the polymer; low out-gassing and good thermal stability at elevated
temperatures; very good chemical and solvent resistance; and also because epoxies readily accept
fillers.

The heat transfer process of porous materials is very complicated, especially for polymer composites.
Considerable work has been reported on the subject of heat conductivity in polymers by Hansen and
Ho, 1965[3], Choy and Young, 1977 [4], Tavman, 1991 [5] and 1997[6]. Progelhof et al., 1976 [7]
was the first to present an exhaustive overview on models and methods for predicting the thermal
conductivity of composite systems. It is quite important, therefore, to understand the mechanisms of
heat transfer in polymer composites, which are potential insulating materials.

In this context, Solid glass micro-spheres (glass beads) have some advantages as fillers in polymers
such as low thermal conductivity, coefficient and density. Also, they have strong filling ability,
smooth spherical surface, small and well distributed internal stress in the products and good
processibility of the filled materials. So, SGM filled epoxy would be suitable mostly for applications
related to insulation [8-11]. 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. The
orientation effects associated with moulding are also minimal. They are preferred as fillers especially
when composite properties such as isotropy or low melt viscosity are important. Compared to the
investigations carried out on polymer composites filled with metal and ceramic powders, behaviour of
glass micro-sphere filled composites has remained a relatively less studied area. This type of
composites can be applied in building materials, space flight and aviation industry.

There are only a few published papers on evaluation of effective thermal conductivity of polymer
composites filled with glass beads. Liang and Li, 2006 [8] reported on measurement of thermal
conductivity of hollow glass-bead-filled polypropylene composites. Recently, they [9] in 2007also
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 [10] 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 [11] 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. 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. 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).

The objective of this work is to fabricate different sets of epoxy composites filled with micro-
sized SGM and measurement of their physical, mechanical and thermal characteristics, estimation of
the effects of fillers and filler size on properties like void content of these composites and exploring
the possibility of their use in applications for thermal insulation.
2. Materials and Methods

2.1 Processing of the composites
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 for 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. The test is carried out in accordance with ASTM E-1530 standard.

2.2 Epoxy Composites filled with solid glass micro-spheres
Epoxy composites of 8 different compositions (Table 1) were made for each of the three microsphere sizes (100, 200 and 300 μm). For each composition, the composites were cast in glass molds so as to get both disc type specimens (diameter 10, 20 mm and thickness 5, 10 mm) and rectangular specimens (length 150 mm, width 20 mm, thickness 4 mm).

| Composition | Epoxy + 0 vol % SGM | Epoxy + 5 vol % SGM | Epoxy + 10 vol % SGM | Epoxy + 15 vol % SGM | Epoxy + 20 vol % SGM | Epoxy + 25 vol % SGM | Epoxy + 30 vol % SGM | Epoxy + 35 vol % SGM |
|-------------|---------------------|---------------------|---------------------|---------------------|---------------------|---------------------|---------------------|---------------------|

Table 1 Epoxy composites filled with Solid glass micro-spheres

| SGM content (vol %) | Effective Thermal Conductivity (W/m-K) |
|---------------------|----------------------------------------|
|                     | Maxwell's equation | Lewis Nielsen's equation | Russell's model | Measured Values |
| 0                   | 0.363 | 0.363 | 0.363 | 0.363 |
| 5                   | 0.2663 | 0.3390 | 0.345 | 0.298 |
| 10                  | 0.3189 | 0.3163 | 0.327 | 0.287 |
| 15                  | 0.2982 | 0.2947 | 0.237 | 0.258 |
| 20                  | 0.2783 | 0.2741 | 0.221 | 0.231 |
| 25                  | 0.2591 | 0.2545 | 0.201 | 0.189 |
| 30                  | 0.2407 | 0.2358 | 0.187 | 0.159 |
| 35                  | 0.2229 | 0.2179 | 0.172 | 0.136 |

Table 2 Comparison of $k_{eff}$ of composites obtained from different models.
Effective thermal conductivity ($k_{eff}$) of epoxy based composites with SGM content ranging from 0 to 35 vol% are estimated and compared by using some of the existing theoretical models. These values are compared with the corresponding measured values of conductivity (Table 2). The $k_{eff}$ values obtained from the Russell’s theoretical model that is Rule of mixture Series Model are in good agreement with the experimentally measured values. It is thus observed that the analytical model serves as a very good empirical model for the estimation of effective thermal conductivity for spherical inclusions and that the correlation can very well be used to estimate $k_{eff}$ of composites for a broad range of SGM concentrations.

![Variation of effective thermal conductivity with SGM content: Effect of SGM size](image)

Fig.1:- Variation of effective thermal conductivity with SGM content: Effect of SGM size

Using the theoretical model proposed in this work, the effective thermal conductivity $k_{eff}$ of the composites are calculated. The variation of $k_{eff}$ with the SGM (100 micron) content is shown in Figure 3, which also presents a comparison of the calculated values with the experimentally measured ones. It is seen that the results obtained from the proposed correlation are in good agreement with experimental results. Figure 4 presents the variation of $k_{eff}$ with filler content for different glass micro-sphere sizes and it is evident that the reduction in heat conduction capability is maximum for the SGMs with 100 micron size. It is encouraging to note that the incorporation of SGM results in significant drop in thermal conductivity of epoxy resin. With addition of 35 vol. % of SGM (100 micron), the thermal conductivity decreases by about 66% as compared with neat epoxy resin. It is also noted that, smaller the size of solid glass micro-spheres, higher is the drop in conductivity value of the composites.

2.3. Effect of micro-sphere size on effective thermal conductivity

Thermal conductivities of composites filled with SGMs of three different sizes (100, 200 and 300 μm) are measured to study the effect of micro-sphere size on $k_{eff}$. The variation of $k_{eff}$ with micro-sphere size is presented in Figure 1. It is evident from the figure that with addition of SGMs, irrespective of the micro-sphere size, there is always a reduction in the heat conduction capability of the composites.
However, it is found that this reduction is maximum in case of SGMs of smallest size i.e. 100 μm. In other words, smaller the size of solid glass micro-spheres, higher is the drop in conductivity of the composites. With addition of 35 vol% of SGM (100 μm), the thermal conductivity decreases by about 66% as compared to that of neat epoxy. At the same time, with the incorporation of same 35 vol% of SGM (300 μm), the drop in thermal conductivity is only about 53%.

3. Physical Characterization

3.1 Density and volume fraction of voids

Density of a composite depends on the relative proportion of matrix and reinforcing materials and it is one of the most important factors for determining the properties of the composites. The theoretical density ($\rho_{ct}$) of composite materials in terms of weight fractions of different constituents can easily be obtained using the following equation given by Agarwal and Broutman [11].

$$\rho_{ct} = \frac{1}{\\left\{\\left(\\frac{w_f}{\rho_f}\\right) + \left(\\frac{w_m}{\rho_m}\\right)\\right\}}$$ (1)

where, $w$ and $\rho$ represent the weight fraction and density respectively. The suffixes $f$ and $m$ stand for the filler and matrix respectively in a composite with single filler. In the present work, since some of the composites are hybrid type i.e. filled with double fillers (SGM and BN), the above expression for density has been modified for them as:

$$\rho_{ct} = \frac{1}{\\left\{\\left(\\frac{w_{f1}}{\rho_{f1}}\right) + \left(\\frac{w_{f2}}{\rho_{f2}}\right) + \left(\\frac{w_m}{\rho_m}\\right)\\right\}}$$ (2)

where, the suffix $f_1$ and $f_2$ stand for the first and second filler and $m$ stands for the matrix respectively.

The actual density ($\rho_{ce}$) of the composite can be determined experimentally by the Archimedes principle or water displacement technique (ASTM D 792-91).

The volume fraction of voids ($v_v$) in the composites is calculated by using the following equation:

$$v_v = \left(\frac{\rho_{ct} - \rho_{ce}}{\rho_{ct}}\right)$$ (3)

The theoretical and measured densities along with the corresponding volume fraction of voids in the epoxy-SGM composites are presented in Table 3. It may be noted that the composite density values calculated theoretically from weight fractions using Eqn.1 are not equal to the experimentally measured values. This difference is a measure of voids and pores present in the composites. It was found that with the increase in Solid glass micro-sphere content in epoxy resin from 0 to 35 vol%, there was a rise in density of the composite by about 11% although there was a simultaneous increase in the void fraction or porosity from 0.727 % to 2.258% (Table 3).

However, unlike other composites, the void-content in the SGM epoxy composites is desirable as the voids enhance the insulation properties of the composites due to presence of air pockets. It was also observed that smaller sized SGM filled epoxy composites showed less amount of voids than larger particles.
3.2 Scanning Electron Microscopy

The micro-structural features of the various particulate filled composite specimens were examined by Scanning Electron Microscope JEOL JSM-6480 LV (Figure 2(a) & 2(b)). The specimens were mounted on stubs with silver paste. To improve the penetration of light and for better surface micrographs, a thin film of platinum is vacuum-evaporated onto the samples before the pictures were taken.

From the SEM micrographs, it is evident that the shape of SGM particles is spherical and hence they have no stress concentration effects inside the matrix and the distribution of SGMs in the epoxy matrix is uniform. It was also observed that the smaller sized SGMs are more uniformly distributed in the matrix than the larger particles.

Table 3 Measured and Theoretical densities of the composites (Epoxy filled with SGM)

| Composites             | Measured density (gm/cc) | Theoretical density (gm/cc) | Void fraction (%) |
|------------------------|---------------------------|-----------------------------|------------------|
| Neat Epoxy (hardened)  | 1.092                     | 1.100                       | 0.727            |
| Epoxy + SGM (5 vol%)   | 1.107                     | 1.119                       | 1.072            |
| Epoxy + SGM (10 vol%)  | 1.126                     | 1.140                       | 1.228            |
| Epoxy + SGM (15 vol%)  | 1.142                     | 1.160                       | 1.551            |
| Epoxy + SGM (20 vol%)  | 1.158                     | 1.179                       | 1.781            |
| Epoxy + SGM (25 vol%)  | 1.174                     | 1.200                       | 2.167            |
| Epoxy + SGM (30 vol%)  | 1.193                     | 1.220                       | 2.213            |
| Epoxy + SGM (35 vol%)  | 1.212                     | 1.240                       | 2.258            |
4. Conclusion

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 thereby 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 lightweight 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.

References

[1] D.Kumlutas et.al., Thermal Conductivity of particle filled polyethylene composite materials, J. Compos. Sci. Technol. 63(1) (2003) 113-117.
[2] D.Hansen, C.Ho, J Polym Sci. Part A 3(2) (1965) 659-670.
[3] C.L.Choy, K.Young, J Polym Sci. 18(8) (1977) 769-776.
[4] Ismail H.Tavman, Thermal Anisotropy of polymers as a Function of their Molecular orientation, Experimental Heat Transfer, Fluid Mechanics, and Thermodynamics, Elsevier 27(1991) 1562-1568.
[5] Ismail.H.Tavman, J.Powder Technol. 91(1997) 63-67.
[6] R.C.Progelhof, J.L.Throne, R.R. Ruetsch , Methods of Predicting the Thermal Conductivity of composite Systems, Polyem Eng Sci 16(9) (1976) 615-625.
[7] J.Z. Liang, F.H. Li, Measurement of thermal conductivity of hollow glass bead filled polypropylene composites, Polym. Test. 25(4) (2006) 527-531.
[8] J.Z. Liang, F.H. Li, Simulation of heat transfer in hollow glass-bead- filled polypropylene composites by finite element method, Polym.Test. 26(3) (2007) 419-424.
[9] J.Z. Liang, F.H. Li, Heat transfer in polymer composites filled with inorganic hollow micro-spheres: A theoretical model, Polym. Test. 26 (2007) 1025-1030.
[10] K.C. Yung ,et al., Preparation and properties of hollow glass microsphere- filled epoxy- matrix composites, Comp. Sci. Technol. (69) (2009) 260-264.
[11] B. D. Agarwal and L. J. Broutman, Analysis and Performance of Fiber Composites, John Wiley and Sons, New York, NY, USA, 2nd edition, 1990.
[9] J. Z. Liang , F. H. Li Heat transfer in polymer composites filled with inorganic hollow micro-spheres: A theoretical model Polym. Test. 26 (2007)1025-1030.
[10] K. C. Yung et al. Preparation and properties of hollow glass microsphere- filled epoxy- matrix composites Comp. Sci. Technol. 69 (2009) 260 -264.
[11] B. D. Agarwal and L. J. Broutman, Analysis and Performance of Fiber Composites, John Wiley and Sons, New York , NY, USA, 2nd edition, 1990.