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A Numerical Study of Interface Effect on The Effective Thermal Conductivity of Glass Microsphere Filled Polymer Composites

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Abstract

Reinforced polymeric materials have wide spread applications and are especially used in electronic industries as potting compound because of its attractive features such as weight reduction, high modulus, ease of manufacture, and flexibility in designing the properties. These electronic compounds are subjected to thermal variations which can adversely affect the reliability of the system and electrical performance of the product. Therefore thermal characterization of such compounds is highly essential. In this study the effective thermal conductivity of glass spheres embedded in epoxy polymer matrix is investigated numerically. The numerical tool used to model the heat transfer mechanism is Finite element software ANSYS and the effective thermal conductivity is calculated. The main focus of this paper is to study how the particle size and interface affect the overall thermal properties of the composites. A finite element model created with a periodical arrangement of filler is used to measure the thermal conductivity of the composite consisting of an epoxy resin matrix filled with glass spheres of different sizes. The values predicted numerically are compared with that of the thermal conductivity models and the results are compared. The significance of the interface on thermal conductivity for various loading concentration is reported in the paper.

Keywords: Polymer composites, interface, thermal conductivity, finite element and analytical model.

Introduction

Filled polymeric composites offer less weight, high strength, better thermal and electrical conductivity and play an important role in aerospace and aviation electronics where mass is a critical design factor. These are the applications where there is a great demand for filled polymeric composites since the electronic devices are often encapsulated with a highly filled polymeric material, known as a potting compound, that primarily serves to protect the sensitive and conductive components from outside environmental effects[1,2].The matrix polymer is typically composed of a thermosetting epoxy resin, which has a long curing characteristic allows flow ability of resin for continuous encapsulation of the electronic component before curing happens. For the composite system to be used as a potting compounds the desirable qualities are low density, CTE and able to conduct heat to the surrounding so that the heat sinks can be eliminated. Epoxies alone cannot serve the purpose. Inclusion of particulate fillers have been used extensively to reinforce thermosetting epoxy resins to reduce cost, degree of shrinkage, CTE, and to raise the thermalconductivity and glass transition temperature [3]. Above all particulate fillers increases the stiffness relative to the bulk epoxy and there by extending its application.Thus particulate filled polymer composites i.e. polymers filled with thermally conductive particulate matters are coming up as a cost effective way to cope with such thermal management issues [4]. These types of encapsulating compounds are inexpensive and commonly used in a wide range of commercial electronics products, from personal computers to cell phones [5].

The overall CTE of the potting compound can be reduced by Silica-based E-glass, having a CTE of $\sim 5 \times 10^{-6}/°C$, and it is used commonly as a particulate filler in epoxy-based resins which is more suitable for electronics applications[2, 6].The addition of glass microsphere has a large reduction in CTE with desirable mechanical properties. Therefore glass bead filled epoxies are preferably used as electronic potting compound because of its low thermal coefficient of expansion and it act as an electrical insulator. With the increased use and demand for electronic products, the critical issue of heat dissipation which adversely affects the performance of the product has to be addressed carefully. Due to the wide spread use of potting compounds in electronic industries which are subjected to thermal variation during its applications, there is a need to study the thermal characteristics of potting compound.

The determination of effective thermal conductivity is thus considered as an important design parameter for the effective design of filled composite materials. There are only limited literatures available on evaluation of effective thermal conductivity of polymer composites filled with glass beads. The thermal characteristics of Glass bead filled composites investigated by the researchers are reported here. The authors [7] have experimentally measured thermal conductivity of hollow glass bead filled thermoplastic resin, polypropylene. Also the authors [8] have made a numerical investigation for studying the heat transfer mechanism in glass bead filled epoxies by using finite element method, for both two-dimensional and three-dimensional models. They have simulated the effective thermal conductivity of hollow glass microsphere filled polymer composites. The authors [9] further have studied the heat transfer mechanism in polymer composites filled with hollow glass micro-spheres and proposed a theoretical model to predict the thermal conductivity of such composite systems. The authors [10] have reported about the fabrication and estimation of properties of hollow glass microsphere-filled epoxy- matrix composites. Recently Debasmita etal. in the year 2011 [11] have reported the estimation of effective thermal conductivity of a solid glass bead filled epoxy both analytically and numerically using ANSYS. Research is going on in this field to exactly evaluate the effective thermal conductivity of the particle filled composite systems. The presence of the interphase between particle and matrix can change the final properties of the composite. The thickness of the interphase is usually determined indirectly from a composite property, but the results depend very much on the method of determination.

The use of interphase concept as shown in Fig. 1to accurately model the experimental results is found in several studies for evaluating the stiffness of nano composites [12, 13]. Among several researchers Vo, et al. [14] was the first to investigate the effects of the interphase on the modeling of particle-reinforced composites by accurately fitting various closed form models with the experimental data for composites composed of micron-sized Al_2O_3 particles in a silver matrix. The author Brown, et al. [15] have established that interphase thickness in polymer nano composites is independent of particle size by using molecular dynamics. Therefore, reduced particle size can lead to an increasing influence of the interphase on overall composite behaviour for a fixed particle volume fraction. The thickness of the interphase study depends only on the matrix and particle chemical composition and seems to be independent of the size of the particle.

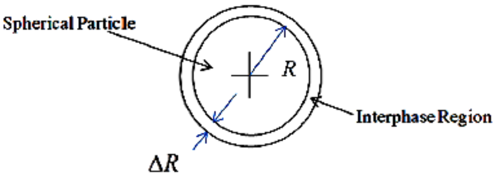


Figure 1: Spherical particle with interface region

In this work the effective thermal conductivity of glass microsphere filled epoxy composite is modelled using finite element software Ansys in 2D. Further the effect of the interface between the matrix and the filler is studied by modelling the interface region.

Analytical Model For Thermal Conductivity Prediction

Increased use of particle filled composites in various applications such as building materials, space flight and aviation industry emphasizes its importance in the evaluation of thermal property of the system. Conductivity is one such important thermal property that needs to be evaluated for any new composite system. For a binary or ternary composite system the effective thermal conductivity of a composite material is a complex function and rely on their geometry of the particle, the thermal conductivity of the matrix and reinforcement phases, distribution of particle within the matrix medium and interface between the particles. Although it can be measured by experimental methods, analytical methods and equations are often essential to predict thermal conductivities of composite materials in order to have a better understanding of the heat transfer process and mechanisms in composite materials. Numerous models have been developed over the last century for determining the effective thermal conductivity of two-phase composites. The models which were developed have assumed particle geometry, particle distribution and complete interfacial contact between the two phases. This section reviews some of the important theoretical / empirical models reported in the literature for calculating the effective thermal conductivity of particle filled composites.

For a binary or ternary composite system, the simplest model is based on the materials arranged in either parallel or series with respect to the heat flow. These models give the upper or lower bounds of effective thermal conductivity as in the relation (1) and (2) respectively.

Parallel Conduction Model:

$K_c = (1-\phi_1-\phi_2)K_m + \phi_1 K_{f1} + \phi_2 K_{f2}$ (1) **Series Conduction Model:**

$$\frac{1}{K_c} = \frac{1-\phi_1-\phi_2}{K_m} + \frac{1}{K_{f1}} + \frac{1}{K_{f2}} \tag{2}$$

The correlations presented in Eqns. (1) and (2) are based on the rule of mixture.

Where, K_c,K_m, K_{f1}, K_{f2} are thermal conductivity of composite material, matrix material, 1st filler, 2nd filler, ϕ_1 , ϕ_2 are volume fraction of 1st and 2nd filler respectively.

Agari and Uno Model:

By taking into account both the parallel and series conduction mechanisms, Agari and Uno have proposed a model for filled and is given by the relation (3).

$$\log K_c = \phi C_2 \log K_f + (1-\phi)\log(C_1.K_m) \tag{3}$$

Where c_1 and c_2 are constants determined experimentally which is of order unity.

Ratcliffe Empirical Model:

The conduction mechanism in a particle filled composites is assumed to have a geometric mean by Ratcliffe. The emprical relation proposed by him is given in equation (4)

$$K_c = K_m^{(1-\phi_1-\phi_2)} K_{f1}^{\phi_1} K_{f2}^{\phi_2} \tag{4}$$

Russell Model:

Russell model predicts the effective thermal conductivity model using series parallel network, assuming similar sizes of cubes and pores and is given in equation (5).

$$K_c = K_m \left[\frac{\phi^{\frac{2}{3}} + \frac{K_m}{K_f}\left(1-\phi^{\frac{2}{3}}\right)}{\phi^{\frac{2}{3}} - \phi + \frac{K_m}{K_f}\left(1+\phi+\phi^{\frac{2}{3}}\right)} \right] \tag{5}$$

Lewis Model:

Lewis and Nielsen have proposed a model for two phase system including shape of particle and orientation for calculating the effective thermal conductivity and is given in equation (6).

$$K_c = K_m\left[\frac{1+AB\phi}{1-B\phi\psi}\right] \tag{6}$$

Where

$$B=\left[\frac{\left(K_f/K_m\right)-1}{\left(K_f/K_m\right)+A}\right] \text{and, } \psi = 1+\left[\frac{1-\phi_m}{\phi_m^2}\right]$$

Where, K_f is thermal conductivity of filler material, Ø is the volume fraction of filler material. The value of A and Ø_m for different shapes are provided [16].

Maxwell Model:

Maxwell has assumed the dispersion of the particle in the matrix medium as the spherical particle suspended in cube. He has proposed the expression mentioned in equation (7) for predicting the effective thermal conductivity based on the flux model where the temperature field is solved for the given geometry.

$$K_c = K_m\left[\frac{K_f+2K_m+2\phi\left(K_f-K_m\right)}{K_f+2K_m-2\phi\left(K_f-K_m\right)}\right] \tag{7}$$

DebasmitaMishra Model:

The expression for effective thermal conductivity of particle filled composites is derived by DebasmitaMishra andAlokSatapathy in the year 2013 [11] and is given in equation (8).They derived the expression by assuming the heat transfer mechanism as pure conduction. The expression for thermal conductivity of a two phase composite is deduced by averaging the thermal conductivity in two different regions.The regions are pure polymeric region and the other is a combination of filler and polymer region. (8)

$$K_c = \left[\frac{1}{\left(K_m(A)^{\frac{1}{3}}\right)+2[B+CD]^{-1}}\right]^{-1} \tag{8}$$

Where,

$$A=1-6\left(\frac{v_f}{\pi}\right); B=K_m\left(\frac{4\pi}{3v_f}\right)^{\frac{1}{3}}; C=\pi\left(\frac{2v_f}{9\pi}\right)^{\frac{1}{3}}; D=\left(\frac{K_f\rho_m}{\rho_f}-K_m\right)$$

Numerical Analysis

In the applications of aerospace and aviation industries the electronic potting compounds are subjected to large thermal variations. The estimation of effective properties of filled composites is of vital importance for proper design and application of filled composite materials. The effective properties of the composite are dependent on the micro-structural characteristics. Micro-structure defines the shape, size, spatial distribution and orientation of the embedment in the matrix. Thus the factors affecting the effective thermal conductivity of dispersion composite are structural features of the composite and thermal conductivity of each component material. Hence one need to exactly model the microstructure of the Particle filled composites. Finite element method is one of the numerical tool to solve complex engineering problems which provides an approximate solution rather than an exact solution in a simpler way. The structure of dispersion composite is considered to be composed of some very simplified basic models. In this work the shape of matrix is assumed to be cubical and the dispersion as sphere. The heat transfer mechanism within the composite system is assumed to be heat conduction.

Concept and Basic steps

In FEM, the actual continuum is discretized and represented as an assemblage of subdivisions called finite elements. The field variable is approximated by an approximating functions called interpolation models within the element. By solving the field equations the nodal values of the field variable is computed. Finite element modeling package ANSYS is used for solving a wide variety of mechanical problems which entails to static/dynamic, structural analysis (both linear and non-linear), heat transfer and fluid flow problems as well as acoustic and electromagnetic problems.

In the present work, using the finite-element software ANSYS thermal analysis is carried out for epoxy-solid glass microsphere to determine the thermal conductivity. In order to perform this analysis, two cases of 2D physical model with spheres-in-cube (SGM in epoxy) have been considered. In the first case the dispersed phase is considered as a periodic arrangement as shown in Fig. 2 and in the second case a single representative volume element (RVE) as shown in Fig. 3 is considered for analysis. The finite element model for the two different cases is used to simulate the microstructure of composite materials for five different particle loading concentrations. Furthermore, the effect of interface between the particle and matrix on the effective thermal conductivities is numerically determined for various loading concentrations.

The particle filled polymer composite considering the interface is modelled as three phase continuum represented by a matrix and homogenous spherical particle. The interface region of the filled composites is modelled using the linear correlation between the matrix and filler material.

Problem Description

The matrix material considered for Analysis is Epoxy resin and the filler is Solid Glass microsphere (SGM). The filler particle size considered in this analysis is 100 µm diameter. Table 1 shows the material properties of the filler and matrix material considered for the analysis.

Table 1: Material parameters

Material	Property	
	Thermal conductivity W/(m*K)	Density g/cm^3
Epoxy	K_m= 0.363	1.19
Solid Glass Microsphere	K_f=0.00363	2.54

Initially for the first case the 2D model of the solid glass micro-spheres embedded in an epoxy body having a periodic arrangement as shown in Fig. 2 at volume fraction of 0.4 % of filler is created. The prescribed boundary conditions for the thermal analysis of particle filled composite and the direction of heat flow for the conduction problem is as shown in Fig. 4.The temperature at the nodes along the surface ABCD is T_{high}=100°C and the analysis of the problem is done at a room temperature of T_{low} = 27°C. The remaining surfaces parallel to the direction of the heat flow are assumed as insulated.

Then the thermal analysis is carried out by modeling as single RVE for various particle volume fractions.

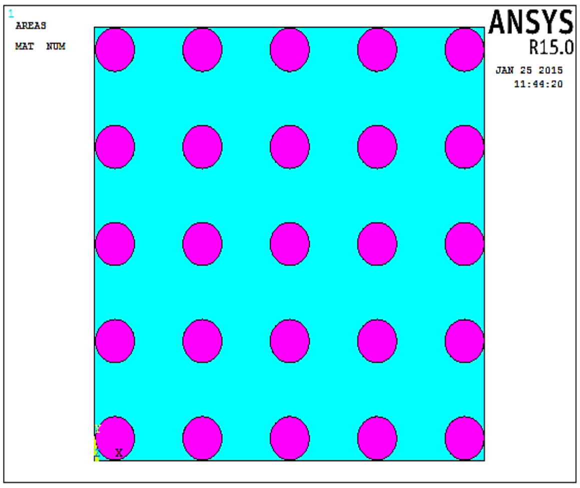


Figure 2: 2D Geometric Model of SGM filled epoxies at V_f =6.5% (Periodic arrangement)

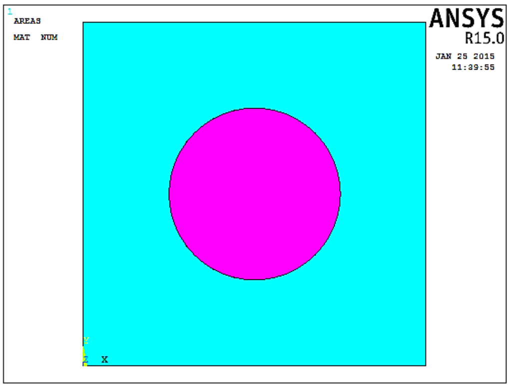


Figure 3: 2D Geometric Model of SGM filled epoxies at $V_f = 6.5\%$ (Single RVE element)

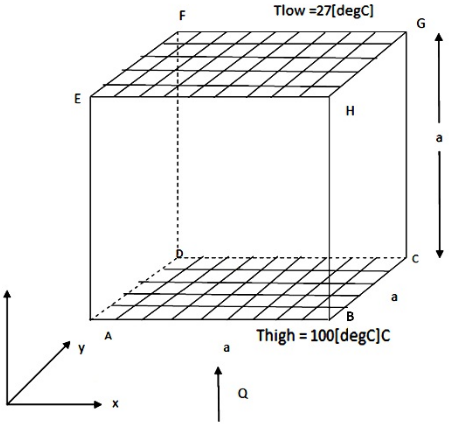


Figure 4: Heat flow and boundary Conditions for SGM filled Polymer composites

Further the concept of interface region between the matrix and filler is numerically modelled. The thermal analysis of SGM filled epoxies considering the interface region is performed. The interface region is created around the filler for a small increment in the particle radius as 8%, 10% and 12% of the particle radius in a single RVE model. The finite element model considering the interface region is as shown in the Fig 5. The interface region is modelled as a third material and the material behavior is assumed as a linear correlation between the matrix and the filler. The values of effective thermal conductivity are predicted for different particle loading concentrations and are compared with the analytical and literature experimental data.

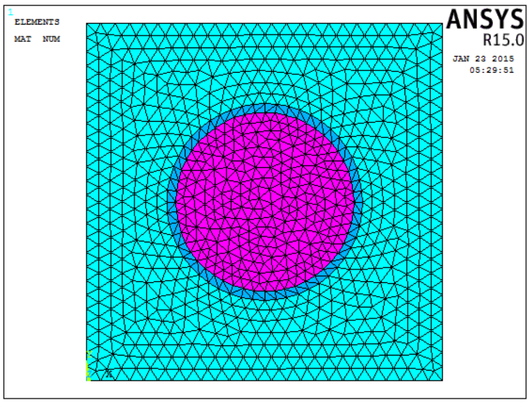


Figure 5: Finite element model of SGM filled composite with interface

Result and Discussions

The thermal analysis of solid glass microsphere filled epoxies is carried out by considering the particle arrangement as periodic arrangement and single RVE 2D model at different filler proportions. The different volume fractions ranging from 0.1 to 0.17 of the glass microsphere of 100 micrometer is considered in this study. Fig. 6 and Fig.7 shows the temperature distribution and the heat flux distribution for the given boundary conditions of a 2D model. From the average heat flux value obtained the thermal conductivities are computed and listed in Table 2. The Table 2 shows the effect of filler content in the epoxies on thermal conductivity.

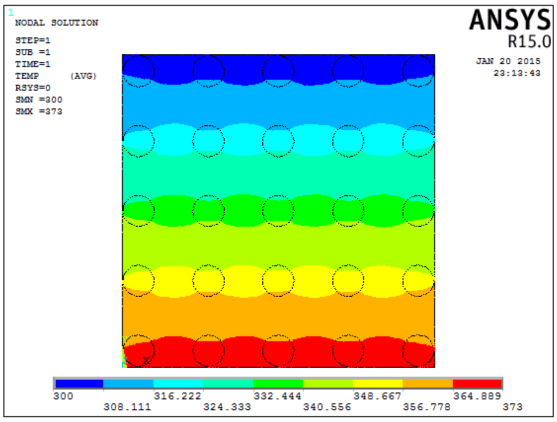


Figure 6: Temperature distribution for 2D model of SGM filled epoxies at V_f = 6.5% (Periodic arrangement)

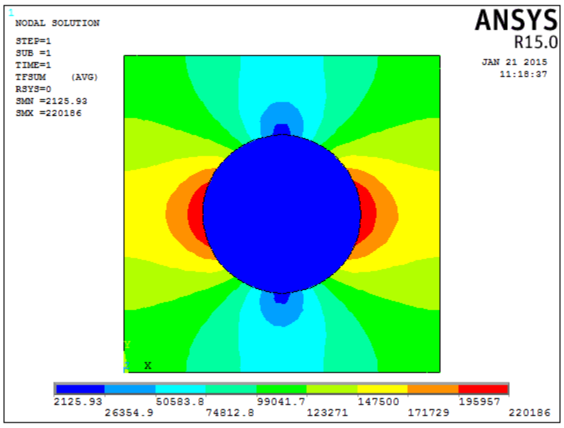


Figure 7: Heat flux distribution for 2D model of SGM filled epoxies at $V_f = 6.5\%$ (Single RVE)

From the Fig 8, it is clear that the numerical results for the 2D model of the both cases are in close agreement. Hence the numerical analysis emphasize that the single RVE model can be used for thermal analysis, which reduces the computation time. Further the numerical results obtained are compared analytically by using various models described in the section 2 are listed in Table 3.

Table 2: Effect of filler content in the epoxies on thermal conductivity

SGM content (Vol. %)	Effective Thermal Conductivity$K_{eff,}$ W/mK			
	2D FEA Periodic	2D FEA RVE	2D FEA Interface	Experimental [11]
0	0.363			
1.4	0.32	0.32	0.30	0.293
3.4	0.29	0.29	0.27	0.267
6.5	0.25	0.25	0.24	0.237
11.3	0.2	0.21	0.23	0.232
17.9	0.17	0.17	0.21	0.229

Table 3: Prediction of Thermal conductivity by Analytical and Numerical Methods for SGM filled epoxies

SGM content (Vol. %)	Effective Thermal Conductivity$K_{eff,}$ W/mK					
	K-ROM	K - RAT	K-MAX	K-LEWIS	2D FEA Interface (10%)	Experimental [11]
1.4	0.358	0.340	0.353	0.349	0.3	0.293
3.4	0.351	0.310	0.339	0.331	0.27	0.267

6.5	0.340	0.269	0.319	0.304	0.24	0.237
11.3	0.322	0.216	0.290	0.268	0.23	0.232
17.9	0.299	0.159	0.254	0.226	0.21	0.229

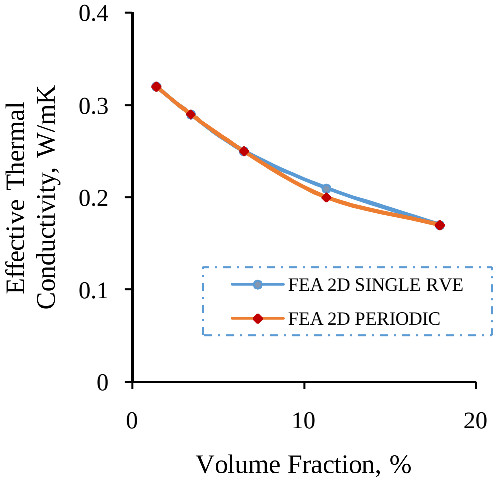


Figure 8: Effective thermal conductivity of SGM filled epoxies using FEA

The interface effect on the effective thermal conductivity of SGM filled epoxies predicted using numerical analysis is listed in Table 2 and Table 3.The effect of interphase region has a significant role and is as shown in the Fig. 9. The Fig. 9 shows that the numerical analysis carried out considering the interface is closer to the literature experimental values [11]. Further the effect of interface thickness as a fraction of particle diameter is also studied and is shown in Fig. 10.

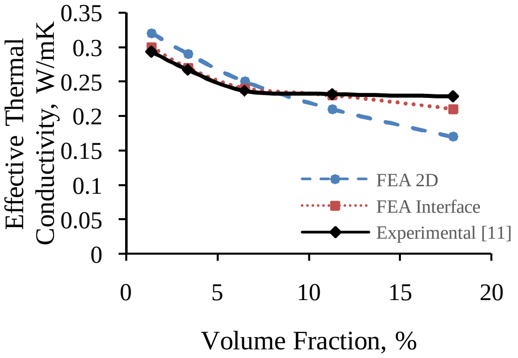


Figure 9: Effect of Interface on effective thermal Conductivityof SGM filled epoxies

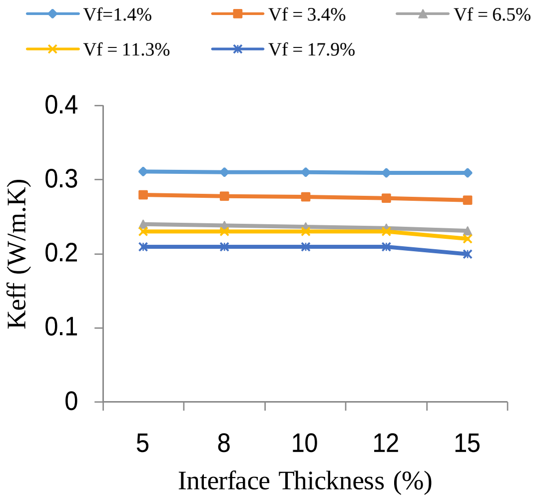
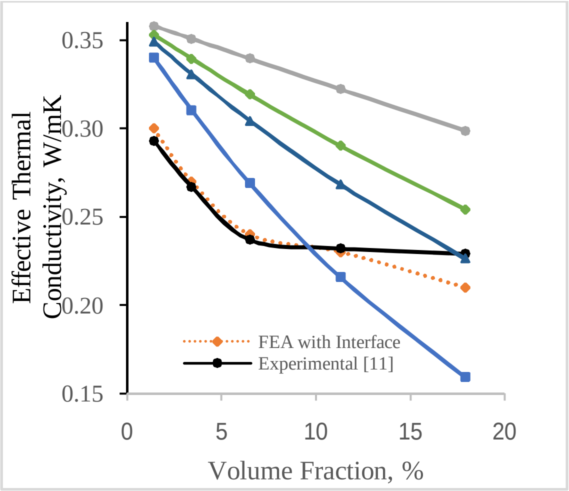


Figure 10: Effect of Interface thickness on Thermal Conductivityof SGM filled epoxies

From the Fig. 10, it is clear that the role of interface thickness as a function of particle radius is almost constant upto 10%. Thus concept of interface around the particle as a function of particle radius can be taken around 5 – 10 % and it relies only on the characterization of the interface material model. Thus the thermal conductivity study on SGM filled epoxies is summarized in the Figure 11. Figure 11 shows the effective thermal conductivity computed analytically and numerically and are compared with literature experimental values. The numerical results by considering the interface are closer to the experimental values. Further the analytical models referred are predicting the values with an error percentage more than 20%. The percentage error in the prediction is lowered with the increase in particle loading concentration. Lewis Neilson model tends to exactly evaluate only at higher particle loading concentration.



Conclusions

In this work the solid glass microsphere filled epoxy resin modeled as 2D is analyzed thermally using the ANSYS software. The variation in the temperature and heat flux distribution at different filler proportions is obtained for both single RVE and periodic arrangement. The effective thermal conductivity values of the SGM filled epoxy composite system is found at different volume fractions for both cases. From the results of the thermal analysis the numerical modelling as single RVE is sufficient for effective computation. The numerically obtained values are compared with the analytical model. The results show that the analytical model prediction is with an error of more than 20%. This may be due to the fact that the empirical model does not consider the heat transfer mechanism at the interface of the particle which is suspended in a matrix. The effect of interphase region around the spherical particle is studied in this work. From the numerical study made, it is clear that the exact modelling of the interface region exactly predicts the effective thermal conductivity of two phase composite system which agrees with the literature experimental data.

References

[1] Wong, C. P., and R. S. Bollampally, 1999, “Thermal Conductivity, Elastic Modulus, and Coefficient of Thermal Expansion of Polymer Composites Filled With Ceramic Particles for Electronic Packaging”, *Journal of Applied Polymer Sci.*, **vol. 74**, pp. 3396–3403.

[2] Harper,C.A., 2000, “Electronic Packaging and Interconnection Handbook,3rd Ed. NewYork, McGraw-Hill.

[3] Garg, A. C., and Y. W. Mai, 1988,“Failure Mechanisms in Toughened Epoxy Resins – A Review” Composites Science and Technology, **vol. 31**, pp. 179–223.

[4] Bujard P., Kuhnlein G., Ino S., Shiobara T., 1994, Thermal conductivity of molding compounds for plastic packaging, *IEEE Transactions on Components Hybrids and Manufacturing Technology Part A*,**vol.17(4**), pp527–32.

[5] Zweben, C., 1992 “Metal-Matrix Composites for Electronic Packaging.” *Journal of ElectronicsMaterials*, **vol. 44,** pp. 15–23.

[6] Smith, J. C., 1976 “Experimental Values for the Elastic Constants of a Particulate-Filled GlassyPolymer.” *Journal of Applied Physics and Chemistry*, **vol. 80A,** pp. 45–49.

[7] J.Z. Liang, F.H. Li., 2006, Measurement of thermal conductivity of hollow glass bead filled polypropylene composites”, *Polym. Test*, **vol.25(4**), 527-531.

[8] J.Z.Liang, F. H. Li, 2007, “Simulation of heat transfer in hollow glass-bead- filled polypropylene composites by finite element method”, Polym.Test, **vol.26(3),**pp. 419-424.

[9] J.Z.Liang, F.H.Li, 2007,"Heat transfer in polymer composites filled with inorganic hollow micro-spheres:A theoretical model", ,Polym. Test. **Vol.26,**pp. 1025-1030.

[10] K.C.Yung,B.L. Zhu., T.M. Yue,C.S.Xie, 2009, "Preparation and properties of hollow glass microsphere- filled epoxy- matrix composites", Comp. Sci. Technol., **Vol.69, pp.**260-4.

[11] Debasmita Mishra, Lucy Mohapatra, AlokSatapathy and Amar patnaik, 2011, "Determination of Thermal Conductivity of Polymer composites filled with Solid Glass Beads", Proc. Ínt. Conference on Advancement in Polymeric Materials.

[12] V. Cannillo, F. Bondioli, L. Lusvarghi, M. Montorsi, M. Avella, M. Errico, and M. Malinconico, 2006, "Modeling of ceramic particles filled polymer–matrix nanocomposites", Compos SciTechnol**vol. 66**, pp.1030-1037.

[13] H. Liu, and L. Brinson, 2008, "Mimicking mussel adhesion to improve interfacial properties in composites", Compos Sci Technol., **vol. 68**, pp.1502.

[14] H. Vo, M. Todd, G. Shi, A. Shapiro, M. Edwards, 2001, "Towards model-based engineering of underfill materials: CTE modeling", Microelectron J. **vol.32** pp. 331

[15] D. Brown, V. Marcadon, P. Mele, N. Alberola, 2008, "Effect of particle size on the properties of model nanocomposites", Macromolecules, **vol. 41**, pp.1499

[16] Serhat Durmaz, 2004,M.S. Thesis, Dokus Eyulil University