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A NUMERICAL ON HEAT CONDUCTIVITY BEHAVIOR OF ZIRCONIA FILLED EPOXY COMPOSITES

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ABSTRACT

A numerical simulation of the heat-transfer process within epoxy matrix composite filled with micro-sized zirconia particles using finite element method is proposed in this paper. Three-dimensional spheres-in-cube lattice array models are constructed to simulate the microstructure of composite materials with zirconia content ranging from about 1.41 to 11.31 volume fraction and the effective thermal conductivities of the composites are estimated. A commercially available finite-element package ANSYS is used for this numerical analysis. The result shows that the effective thermal conductivity (k_{eff}) increases with increase in the volume fraction of the zirconia in the epoxy matrix. The simulated values are compared with calculated k_{eff} values obtained from other established correlations such as Rule-of-Mixture (ROM), Maxwell's model and Lewis Neilson model.

KEYWORDS: Polymer matrix composites, epoxy, zirconia, thermal conductivity, numerical simulation.

I. INTRODUCTION

Modern day electronic devices are packaged with many polymeric materials, such as electronic molding compound (EMC) and glob top encapsulation [1]. Usually thermosets like epoxy or polyester resin and thermoplastics like polypropylene (PP) or polyamide systems are used for encapsulating a variety of electronic components because of their high thermal stability, moisture resistance and low cost. But unfortunately, these resins have poor thermal properties like high coefficient of thermal expansion and low thermal conductivity [2]. The high thermal expansion coefficient of plastic resin is lowered with the addition of ceramic powder filler, like fused silica or quartz. But silica-filled epoxy resins are less desirable for the encapsulation of silicon integrated circuit (IC) chips because of silica's low thermal conductivity. Since recent applications of polymers as heat sinks in electronic packaging require new composites with relatively high thermal conductivity, it is important to enhance the conductivity of the polymers [3]. Improved thermal conductivity in polymers may be achieved either by molecular orientation or by the addition of conductive fillers. Thermally conducting polymer-matrix composites are becoming increasingly important for electronic packaging because the heat dissipation ability limits the reliability, performance and miniaturization of electronics. By the addition of fillers to plastics the thermal behaviour of polymers can be increased significantly. Such filled polymers with higher thermal conductivities than unfilled ones become more and more an important area of study because of the wide range of applications, e.g. in electronic packaging in applications with decreasing geometric dimensions and increasing output of power, like in computer chips or in electronic packaging [4].

Reports are available in the existing literature on experimental as well as numerical and analytical studies on thermal conductivity of some filled polymer composites. Most of the work was conducted taking metal powder as filler material. In this series, Rusua et al. [5] first used zinc powder as filler material and found appreciable increase in the thermal conductivity. Later Mamunya et al. [6] also reported the improvement in electrical and thermal conductivity of polymers filled with metal powders. Further Boudenne et al. [7] used aluminium as filler material in polypropylene matrix for improvement in thermal conductivity of the matrix body. Later they further went with similar study and this time they had taken copper as filler material for the improvement of thermal conductivity of polypropylene composites [8].

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Carbon-based fillers with high thermal conductivity and low density appear to be the most promising fillers. Graphite, carbon fiber and carbon black are well-known carbon-based fillers. Graphite is considered as the best conductive filler because of its good thermal conductivity and low cost [9]. Carbon fiber, typically vapor grown carbon fiber (VGCF), is important carbon-based filler [10]. Studies conducted on modified thermal conductivity of polymer composites filled with carbon nanotubes have recently been reviewed by Han and Fina [11].

Metallic and carbon-based fillers are highly conductive thermally, but they are highly electrically conductive as well. There are certain areas where high thermal conductivity is required but at the same time electrical resistivity is of prime importance, like in electronic devices. Ceramic powder reinforced polymer materials have been used extensively for such applications because of their high thermal and low electrical conductivity. Some promising ceramic fillers such as SiC, AlN, Al2O3 and ZnO [12, 13] are in use to improve thermal conductivity of various polymers. In a more recent work Agrawal et al. [14] found increase in the value of thermal conductivity and glass transition temperature whereas decrease in the value of coefficient of thermal expansion for epoxy/AlN composites and polypropylene/AlN composites. They also reported that for wide range of filler content, dielectric constant of the composite remains constant for both sets of composites. Incorporation of multiple fillers into the polymer matrix for the improvement of thermal conductivity has also been reported.

A numerical approach to evaluate the effective thermal conductivity of granular or fibrous reinforced composite materials was proposed by Veyret et al. [15], whereas Kumlutas and Tavman [16] have developed a numerical model for particulate filled polymers which shows good agreement with the experimental values. Nayak et.al [17] has reported on the modified thermal conductivity of pine wood dust filled epoxy based composites using computational method.

Numerous theoretical and empirical models have been proposed in the past to estimate and predict the effective thermal conductivities of particulate filled composites. Comprehensive review articles have discussed the pertinent applicability of many of these analytical models. The simplest alternative for a two-component composite system would be with the arrangement of materials in either parallel or series with respect to heat flow which gives the upper or lower bounds of effective thermal conductivity.

For series conduction model [18]

$$\frac{1}{k_c} = \frac{1-\phi_f}{k_m} + \frac{\phi_f}{k_f} \tag{1}$$

where, k_f, k_m, k_c are thermal conductivities of filler, composite matrix and conductivity of the composite as a whole and Φ_f is volume fractions of filler.

The correlation represented by Equations (1) is derived on the basis of the rules-of-mixture.

Maxwell [19] has obtained an exact expression for thermal conductivity, using potential theory for an infinitely dilute composite of spherical particulates dispersed randomly and devoid of mutual interaction in a homogeneous medium, which is given by

$$k_c = k_m \left[\frac{k_f + 2k_m + 2\phi_f (k_f - k_m)}{k_f + 2k_m - 2\phi_f (k_f - k_m)} \right] \tag{2}$$

Eq. (2) is well known for dilute composites which is the earliest flux law in which a cube of suspension for a single particle was considered.

Lewis and Nielsen [20] derived a semi-theoretical model for a two phase system which assumes an isotropic particulate reinforcement and also takes into consideration the shape of particle as well as its orientation

$$k_c = k_m \left[\frac{1 + AB\phi}{1 - B\phi\psi} \right] \tag{3}$$

Where,

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

Zirconia powder with a moderate thermal conductivity and low cost therefore emerges as a suitable filler material to be used in polymeric materials. In view of this, in the present work, zirconia is chosen as the ceramic filler to be dispersed within epoxy resin. The objective of this work is to analyze the heat transfer through the ZrO2-epoxy composites and to evaluate the equivalent thermal conductivity of these composites by numerical methods. It reports the estimation of the equivalent thermal conductivity of this particulate-polymer composite system using finite element method. Later, the numerically obtained values are compared with the values obtained from theoretical model.

II. MATERIALS AND METHODS

Material considered

Matrix materials are the base of composite fabrication. The presently used matrix is a thermoset polymer epoxy. The epoxy resin Lapox-12 is considered in the present work which belongs to the epoxide family. Bisphenol-A-Diglycidyl-Ether (commonly abbreviated to DGEBA or BADGE) is the common name of the presently used epoxy resin and its molecular chain structure. It provides a solvent free room temperature curing system when it is combined with the hardener tri-ethylene-tetramine (TETA) which is an aliphatic primary amine with commercial designation HY 951. Epoxy is chosen from present investigation because it possesses good thermal conductivity of 0.363 W/m-K.

Zirconium dioxide (ZrO2), sometimes known as zirconia (not to be confused with zircon), is a white crystalline oxide of zirconium. Its most naturally occurring form, with a monoclinic crystalline structure, is the mineral baddeleyite. A dopant stabilized cubic structured zirconia, cubic zirconia, is synthesized in various colours for use as a gemstone and a diamond simulant. Zirconia is chemically unreactive. Zirconium dioxide is one of the most studied ceramic materials. Zirconia adopts a monoclinic crystal-structure at room temperature and transitions to tetragonal and cubic at higher temperatures. The main reason for the selection of zirconia in present study is because of its good thermal conductivity (16.7 W/m-K) and low cost. Table 1 shows the list of composites considered in present investigation for numerical analysis.

Table 1 List of composites filled with micro-sized powder

S. No.	Composition
1	Neat epoxy
2	Epoxy + 1.41 vol % ZrO_2
3	Epoxy + 3.35 vol % ZrO_2
4	Epoxy + 5.236 vol % ZrO_2
5	Epoxy + 7.85 vol % ZrO_2
6	Epoxy + 9.42 vol % ZrO_2
7	Epoxy + 11.31 vol % ZrO_2

III. RESULTS AND DISCUSSION

Numerical method

The Finite Element Method (FEM), originally introduced by Turner in 1956, is a powerful computational technique for approximate solutions to a variety of "real-world" engineering problems having complex domains subjected to general boundary conditions. FEM has become an essential step in the design or modeling of a physical phenomenon in various engineering disciplines.

Description of the problem

The determination of equivalent properties of composite materials is of paramount importance for functional

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design and application of composite materials. One of the important factors that influence the equivalent 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 equivalent 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.

Using the finite-element program ANSYS, thermal analysis is carried out for the conductive heat transfer through the composite body. In order to make a thermal analysis, three-dimensional physical models with spheres-in-a-cube lattice array have been used to simulate the microstructure of composite materials for different filler concentrations. Furthermore, the equivalent thermal conductivities of these epoxy composites filled with micro size zirconia particle up to about 11.3% by volume is numerically determined using ANSYS.

Assumptions

In the analysis of the ideal case it will be assumed that
1. The composites are macroscopically homogeneous.
2. Locally both the matrix and filler are homogeneous and isotropic.
3. The thermal contact resistance between the filler and the matrix is negligible.
4. The composite lamina is free of voids.
5. The problem is based on 3D physical model.
6. The filler particles are in a square periodic array/uniformly dispersed in matrix.

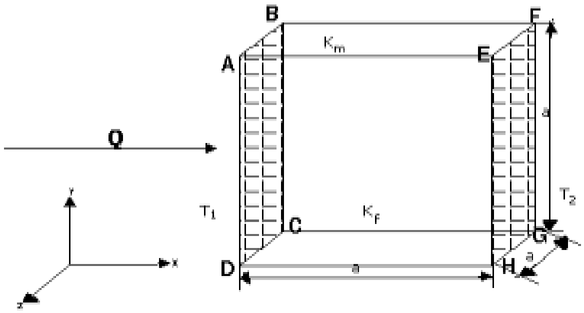

Fig. 1 Boundary conditions

Numerical Analysis

In the numerical analysis of the heat conduction problem, the temperatures at the nodes along the surfaces ABCD is prescribed as T_1 (=100^{0}C) and the convective heat transfer coefficient is assumed to be 2.5 W/m^2-K at ambient temperature of 27°C. The heat flow direction and the boundary conditions are shown in Fig. 1. 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 adiabatic boundaries are unknown. These temperatures are obtained with the help of finite-element program package ANSYS.

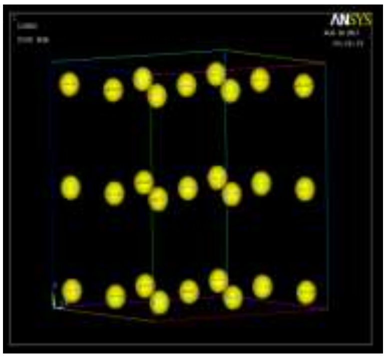

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Fig. 2 A typical 3-D spheres-in-cube model

Thermal conductivities of epoxy composites filled with micro size zirconia particles up to 11.3 % by volume 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 3.35 vol% within the cube shaped matrix body is illustrated in Fig. 2. The temperature profiles obtained from FEM analysis for the composites (spheres-in-cube arrangement) with particulate concentrations of 1.4, 3.35, 5.236, 7.85, 9.42 and 11.31 vol. % are presented in Figs. 3.

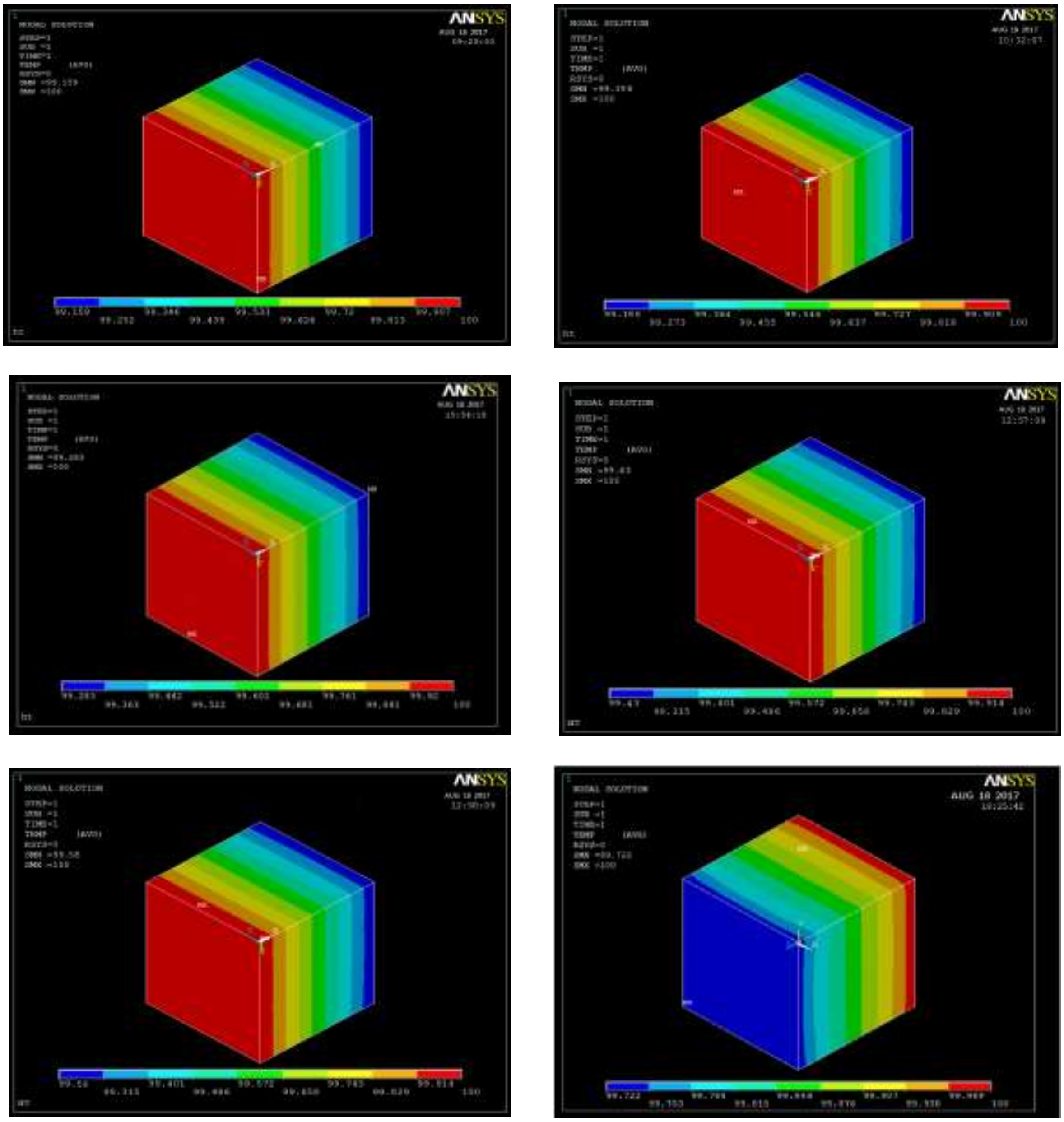

Fig. 3 Temperature profile for composite with particle concentration ranging from 1.41 to 11.31 vol%

Comparison of values obtained from different methods

The numerical results are compared with the values calculated using some of the existing theoretical and empirical models. Rule of mixture model, Maxwell's equation and Lewis and Nielsen's equation are presented in Table 2. It presents a comparison among the results obtained using these models with regard to the corresponding values of equivalent conductivity obtained by numerical simulation.

It is evident from the table that there is appreciable increase in thermal conductivity as the concentration of zirconia particle is increasing. It is also clear that the FEM results are in better agreement with the theoretical model for low filler content but as the amount of filler content increasing there is divergence in the value obtained from numerical simulation and theoretical model.

Table 2 Equivalent thermal conductivities obtained from different methods

Sr. No	**Zirconia particles (vol. %)**	**Equivalent thermal conductivity of the composite [W/mK]**			
		Rule of mixture model	**Maxwell's model**	**Lewis and Neilson**	**FEM Simulation**
1	1.41	0.368	0.377	0.375	0.385
2	3.35	0.375	0.398	0.393	0.410
3	5.236	0.382	0.418	0.412	0.442
4	7.85	0.393	0.449	0.441	0.494
5	9.42	0.399	0.468	0.459	0.584
6	11.31	0.408	0.492	0.483	0.654

Though the results obtained from Lewis and Neilson model are giving better results as compare to the other theoretical models. It is noticed that while the FEM analysis can very well be used for predictive purpose in determining the equivalent thermal conductivity for a wide range of particle concentrations. Moreover, although the distribution of zirconia particulates in the matrix body is assumed to be in an arranged manner, it is actually dispersed in the resin almost randomly. However, it is encouraging to note that the incorporation of zirconia particle results in enhancement of thermal conductivity of epoxy resin. With addition of 1.4 vol. % of zirconia particle, the thermal conductivity improves by about 5.23 % and with addition of 11.31% of zirconia particle the thermal conductivity improves by about 73 % when compared with neat epoxy resin. It can be seen from the graph that for less filler concentration, the slope of the curve is less and as the filler volume fraction increases, the curves representing FEM become steeper. It might be due to the fact that with increase in filler concentration, the inter-particle distance reduces and the conductive chains begin to form which increase the thermal conductivity quite reasonably.

IV. CONCLUSION

This numerical investigation has led to the following specific conclusions:
1. Finite element method (FEM) can be gainfully employed for determination of effective thermal conductivity of these composites with different amount of zirconia content.
2. A good agreement of FEM results with those obtained from theoretical model for low filler content is obtained validates the usefulness of this numerical method.
3. Incorporation of zirconia particles results in increase of thermal conductivity of epoxy and thereby improves its conduction capability. With addition of 1.41 vol% of $ZrO2$ particle, the thermal conductivity increases by about 5 % as compared to neat epoxy and it increases by as high as 73 % by adding 11.31 % of same particle.
4. This new class of zirconia particle filled epoxy composites can be used for applications such as thermal fuses, electronic packaging etc

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