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Study of Thermal Conductivity Enhancement of Epoxy With Copper Oxide(CuO)

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Abstract: To enhance the thermal conductivity of the epoxy material, micro-sized copper oxide (CuO) is taken as the filler material in the epoxy Bisphenol-A-Diglycidyl-Ether (commonly abbreviated to DGEBA or BADGE) in the present investigation. Copper oxide(CuO)has the moderate thermal conductivity and low CTE, hence is a potential filler material to be used in polymeric matrices. The thermal conductivity enhancement of epoxy with copper oxide (CuO) has been investigated and the effective thermal conductivities of these composites have been calculated by numerical as well as experimental methods.

Keywords: Composites, Density, CuO, Epoxy, Effective Thermal Conductivity, Filler

I. INTRODUCTION

Material science plays a vital role in determining and improving economic performance and the quality of life, particularly in the fields : health, living environment, communication, consumer goods transport, etc. The concept of fiber composites is very ancient as it dates back to the times of Israelites, 800 B.C. and also Egyptian pharaohs in third B.C. who used straw in bricks manufacturing as reinforcement. In 1968, composites were used to build a dome structure in Benghazi and hence recorded their first use in construction. Now their application in construction has enhanced manifolds [1]. Li et al.[2] found thermal management has become a crucial issue that strongly affects the performance, reliability, and lifetime of electronic devices in the integration and miniaturization . Dilek Kumlutas et al. [3] established numerically and experimentally the thermal conductivity of particle filled polymer composites. Numerically, the finite-element program ANSYS has been used to evaluate the thermal conductivity of the composite and for it the results of the thermal analysis are used. The composites and nano composites of epoxy resin as base matrix and AlN (Alumi-num Nitride) as micro and nanofiller at various loading of AlN has been fabricated by Mousam Choudhury[4] and dielectric properties nave been investigated. Ye.P. Mamunya et al.[5] studied the systems based on epoxy resin (ER) and poly(vinyl chloride) (PVC) filled with metal powders (Cu and Ni) and calculated the electrical and thermal conductivity. M. Karkri [6] numerically and experimentally investigated thermal properties of composites. Deissler's [7] works were extended by Wakao and Kato [8] for a cubic or orthorhombic array and these with Shonnard and Whitaker [9] have developed a global equation with an integral method for heat transfer in the medium for 2-D and 3-D models. Auriault and Ene [10] have investigated the influence of the interfacial thermal barrier on the effective conductivity and on the structure of the macroscopic heat transfer equations. Veyret at al. [11] used the finite elements method for the study of heat conductive transfer in the periodic distribution of the filler in the composite materials. The finite element method was used by Ramani and Vaidyanathan [12] that have incorporated the effect of microstructural characteristics such as interfacial thermal resistance, filler aspect ratio, volume fraction, and filler dispersion to determine the effective thermal conductivity of a composite with spherical and parallel epipedic fillers. A numerical approach to calculate the ETC of granular reinforced composite was proposed by Cruz [13]. Many other contributing works were attributed to Yin et al. [14], Kumlutas et al. [15] and Jiang et al.[16].

II. EXPERIMENTAL PROCEDURE

Micro-sized copper oxide (CuO) is taken as the filler material in the epoxy Bisphenol-A-Diglycidyl-Ether (commonly abbreviated to DGEBA or BADGE). Hand Lay-Up technique has been employed for the preparation of composite samples. Uncured epoxy (LY556) and its corresponding hardener (HY 951) were mixed in a ratio of 10:1 by weight. Nano-sized CuO powder were mixed with the epoxy in different proportions(5% to 25%). The uniformly mixed dough (epoxy filled with CuO) was then slowly decanted into the glass molds so as to get disc type specimens (diameter mm and thickness 7 mm) , coated beforehand with wax and a uniform thin film of silicone-releasing agent. The castings has been placed at room temperature for about 24 hours and then the glass molds were broken and the samples were released. From this slab

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circular shaped specimens (diameter 65 mm, thickness 7mm) were cut for different characterization tests.

III. CHARACTERIZATION

Macroscopic homogeneity of the composites, locally homogeneous and isotropic characteristics of both the matrix and filler and negligible thermal contact resistance between the filler and the matrix have been assumed for the characterization.

3.1. Density and Volume Fraction of Voids

The theoretical density (ρ_{ct}) of composite materials can be evaluated by using the following formula,

$$\rho_{ct} = \frac{1}{\frac{\omega_f}{\rho_f}+\frac{\omega_m}{\rho_m}} \qquad (1)$$

Here, ω and ρ represent the weight fraction and density respectively.

The actual density (ρ_{ce}) of the composite can be determined experimentally by using the water displacement technique i.e, Archimedes principle. The volume fraction of voids (v_v) in the composites can be evaluated by using the following formula,

$$v_v = \frac{\rho_{ct}-\rho_{ce}}{\rho_{ct}} \qquad (2)$$

3.2. Thermal CharacterizatioN

Thermal conductivity tester is used to the room temperature effective thermal conductivity of the composite specimens. Disc type specimens (diameter = 65 mm, thickness = 7 mm) are used for this purpose. This test is conducted in accordance with ASTM E-1530standards.

For one-dimension heat flow, the equation can be deduced as:

$$Q=\kappa A\,\frac{T_1-T_2}{x} \qquad (3)$$

Here, Q is the heat flux (W), A is the cross-sectional area (m^2), k is thermal conductivity (W/m-K), x is the sample thickness (m), $T_1 - T_2$ is the temperature difference between surfaces (°C or K).

The thermal resistance of the sample can be calculated as:

$$R=\frac{T_1-T_2}{QA} \qquad (4)$$

where, R is sample resistance between hot and cold surfaces. (m^2-K / W).

From the above equations, we can deduce that,

$$\kappa = \frac{x}{R} \qquad (5)$$

Transducers measure the value of heat flux Q and the temperature difference between upper and lower plate and thermal resistance between surfaces can be evaluated. The sample thermal conductivity can be calculated by using different input parameters viz. thickness and known cross-sections.

IV. THEORETICAL CALCULATIONS BASED ON VARIOUS MODELS

4.1. Maxwell Model

Maxwell deduced the analytical expressions for effective conductivity of heterogenic medium in his famous work on electricity and magnetism [17]. He considered the problem of dilute dispersion of spherical particles of conductivity k_1 embedded in a continuous matrix of conductivity k_m, by ignoring thermal interactions between filler particles.

Maxwell's expression is written as follows:

$$\frac{k_{eff}}{k_m} = 1\,\frac{3\Phi}{\frac{(k_1+2k_m)}{k_1-k_m}-\Phi} \qquad (6)$$

Here Φ is the volume fraction of the filler. Maxwell's formula can be used and valid only for low Φ (under about 25%).

4.2 The Lewis-Nielsen Model

Lewis –Nielsen empirical model found place in literature for providing results without including ITR in equations. It was created for moderate filler volume fractions (up to 40%), however, it becomes unstable for higher values.[18].

Simplicity and coverage of a wide range of particle shapes and patterns are the main characteristics of this model. The effective thermal conductivity of a composite according to the Lewis-Nielsen model is given as:

$$k_{eff} = \frac{1+AB\Phi}{1-B\Phi\Psi} \qquad (7)$$

Here,

$$B = \left(\frac{\frac{k_1}{k_m}-1}{\frac{k_1}{k_m}+A}\right) \qquad (8)$$

$$\Psi = 1+\left(\frac{1-\Phi_m}{\Phi_m^2}\right)\Phi \qquad (9)$$

Here, k_m is the thermal conductivity of the matrix, k_1 the thermal conductivity of the filler, Φ is filler volume fraction, ϕm is maximum filler volume fraction and A is shape coefficient for the filler particles.

4.3. Rules of Mixture-Series and Parallel Model

The simplest model for a two-component composite can be with the materials arranged in either parallel or series with respect to heat flow. It provides the upper or lower bounds of effective thermal conductivity. Rules of Mixtures (ROM) is the basis of following two models:

From the parallel conduction model:

$$k_c =(1-\Phi)\, k_m+\Phi\, k_f \qquad (10)$$

Here k_c, k_m, k_f are the thermal conductivities of the composite, the matrix and the filler respectively and ϕ is the volume fraction of filler.

From the series conduction model:

$$\frac{1}{k_c} = \frac{(1-\Phi)}{k_m} + \frac{\Phi}{k_f} \qquad (11)$$

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V. CALCULATIONS AND RESULTS

Based on the above models, the following results are obtained:

Table 1: Comparison of Experimental and Theoretical Densities of the Composites (Epoxy filled with Cuo)

S.NO.	COMPOSITES	ACTUAL DENSITY	THEORITICAL	VOID FRACTION
1	EPOXY +5% CuO	1.100	1.147	4.113
2	EPOXY +10% CuO	1.142	1.199	4.759
3	EPOXY +15% CuO	1.195	1.255	4.793
4	EPOXY +20% CuO	1.250	1.317	5.059
5	EPOXY +25% CuO	1.311	1.385	5.319

Table 2: Comparison of Experimental and Calculated Values of Effective Thermal Conductivity

S.NO.	COMPOSITES	SERIES		MAXWELL		LEWIS		EXPERIMENTAL	
		k_{eff}	%	k_{eff}	%	k_{eff}	%	k_{eff}	%
1	EPOXY +HARDNER	0.363	100.00	0.363	100.00	0.363	100.00	0.363	100.06
2	EPOXY +5% CuO	0.382	105.20	0.418	115.25	0.412	113.50	0.492	135.45
3	EPOXY +10% CuO	0.403	110.98	0.480	132.14	0.469	129.33	0.622	171.27
4	EPOXY +15% CuO	0.426	117.42	0.548	150.94	0.538	148.26	0.759	208.99
5	EPOXY +20% CuO	0.453	124.66	0.624	172.00	0.622	171.46	1.014	279.24
6	EPOXY +25% CuO	0.482	132.85	0.711	195.74	0.729	200.73	1.291	355.66

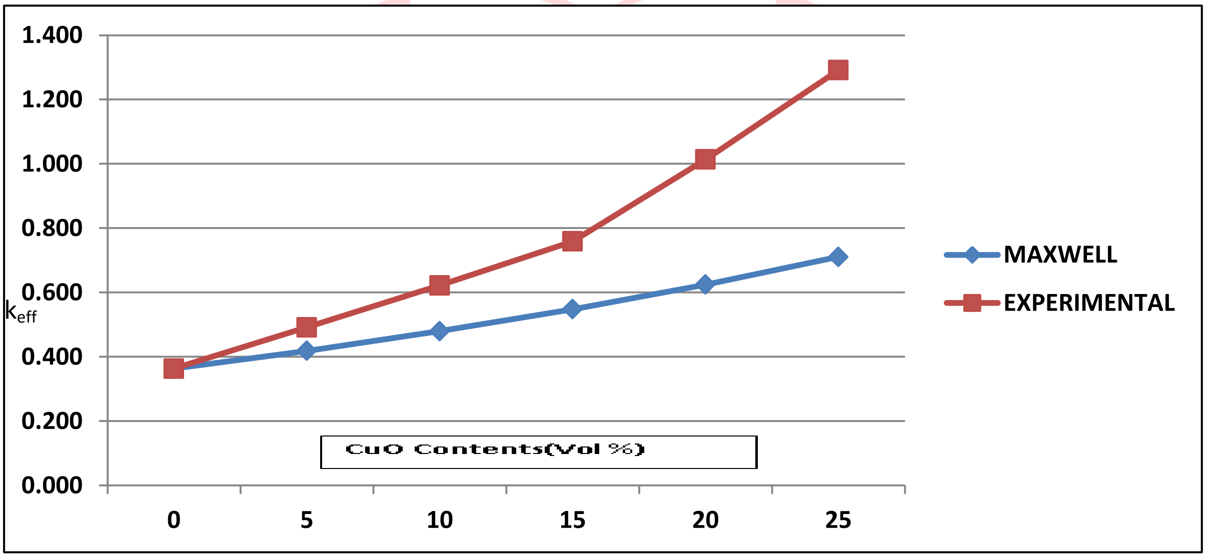

Figure: 1: Comparison of Experimental and Calculated Values of Effective Thermal Conductivity Based on Maxwell Model

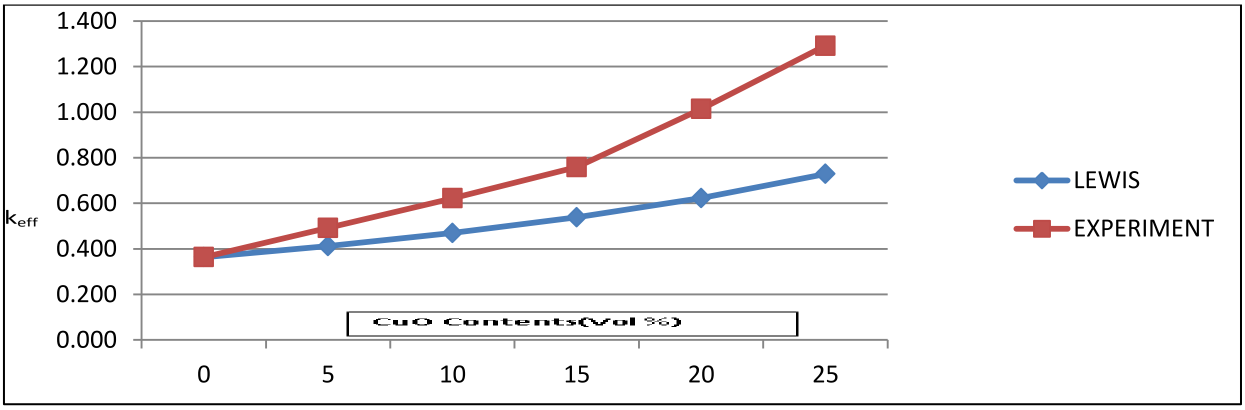

Figure: 2: Comparison of Experimental and Calculated Values of Effective Thermal Conductivity Based on Lewis Model

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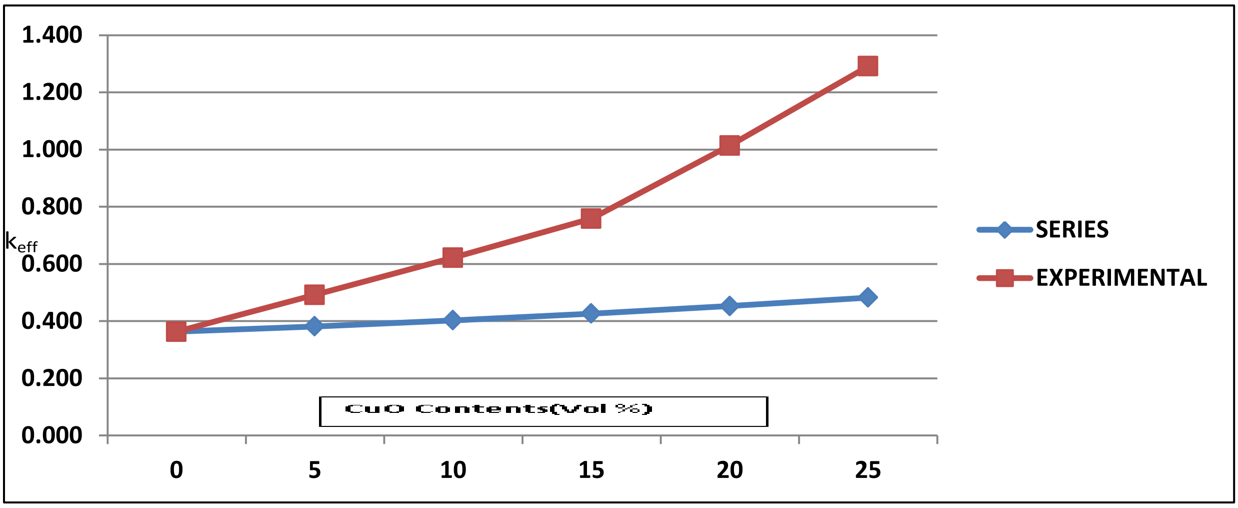

Figure: 3: Comparison of Experimental and Calculated Values of Effective Thermal Conductivity Based on Series Model

Following parameters have been assumed for numerical analysis:

1. Macroscopic homogeneity of the composites.
2. Locally homogeneous and isotropic characteristics of both the matrix and filler.
3. Negligible thermal contact resistance between the filler and the matrix.
4. The composite lamina is free of voids.
5. The problem is based on 3D physical model.
6. The filler are arranged in a square periodic array/uniformly distributed in matrix.

VI. CONCLUSIONS

It has been concluded that successful fabrication of particulate filled epoxy-CuO composites by hand layup technique is possible. Table 1 presents the theoretical and measured densities for the epoxy –CuO composites and also the corresponding volume fraction of voids in the epoxy-CuO composites.. It may be noted that the composite density values calculated theoretically from weight fractions are not equal to the experimentally measured values. This difference is a measure of voids and pores present in the composites. It is found that with the increase in CuO micro-particles content in epoxy resin from 0 to 25vol%, there is a rise in density of the composite by about 20% although there is a simultaneous increase in the void fraction or porosity from 4.113 % to 5.319% .

From Table 2 it has been concluded that the value of k_{eff} keeps on increasing on increasing CuO content in composie. It is deduced that the results obtained from the various models are in good agreement with experimental results up to a filler concentration of about 15 vol % for epoxy . The CuO particles hence show percolation behaviour at these volume fractions (15% for) at which a sudden jump in the thermal conductivity has been noticed. This is the critical concentration, called the percolation threshold, at which CuO particles start contacting with each other within the respective polymer resin.

It is deduced that by incorporating micro-sized CuO particulates into epoxy, the expected effects are achieved in the form of modified physical and thermal properties. Due to the presence of CuO micro-fillers, changes in their heat conduction behavior are seen. When CuO is added to in epoxy, the effective thermal conductivity of the composites is incresed due to conductive nature of CuO. It has been found that there is a sudden increase in the composite thermal conductivity at this point of filler concentration.

VII. ACKNOWLEDMENT

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