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Thermal Properties of Polymer Composites Filled with Ceramic Particles

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Abstract - The present work aims at developing a class of polymer composites consisting of epoxy as a matrix material with micro-size aluminium nitride as a filler material. A set of composites with varying loading of filler content ranging upto 50 wt. % has been fabricated. The effect of filler content on thermal properties of such fabricated samples is investigated. From the experimental results, it is found that inclusion of aluminium nitride filled epoxy composites exhibit improved thermal conduction property, increased glass transition temperature and reduced coefficient of thermal expansion. With modified thermal property, the presently fabricated composites have the potential to be used in applications where thermal conductivity is of prime importance. Also with improved glass transition temperature and coefficient of thermal expansion material can find its application where resistance to thermal fatigue is required.

Keywords: Polymer composites, aluminium nitride, thermal conductivity, glass transition temperature, coefficient of thermal expansion.

I. INTRODUCTION

In microelectronics application, the requirement is quite different. Here high thermal conductivity is required for proper heat dissipation but at the same time electrical resistivity is mandatory for proper signal distribution and to avoid short circuiting. Because of high electrical conductivity of metallic and carbon based fillers, several ceramic materials gained more attention due to their high thermal conductivity and electrical resistivity. On that note, Zhou et al. [1] reported that for 55 wt % of SiC content in linear low density polyethylene

(LLDPE) composites, its thermal conductivity reached as high as 1.48 W/m-K. Another silicon based ceramic i.e. Si_3N_4 with still higher intrinsic conductivity value than SiC is proved to be an excellent filler, which steadily improves the conductivity of polystyrene [2]. In a more recent work Similarly, An et al. [3] also agreed reinforcing potential of Si3N4 when they found thermal conductivity increased up to 1.42 W/m K at 30 vol% of filler. Ramdini et al. [4] modified the surface of silicon to obtain much better results. They used silane coupling agent for surface treatment of silicon nitride particles and found that the thermal conductivity of polybenzoxazine matrix improved from 0.18 W/m-K to 5.78 W/m-K when 70 % of nano silicon nitride is added to it. This shows that the surface modification have great impact in the improvement of thermal conductivity of the material. Anjana et al. [5] reported a sharp increase in conductivity from 0.54 to 3.22 W/m-K as cerium oxide (CeO2) loading increased to 50 vol % in HDPE. Recently, Ozmihci and Balkose [6] found that

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incorporation of ZnO increased the thermal conductivity of LLDPE to 1.56 W/m-K with addition of 20 vol % of filler. Jia et al. [7] used montmorillonite as filler material in epoxy matrix and found appreciable increase in the value of thermal conductivity. They also modified the surface of filler using silane coupling agent and found appreciable results. They further reported that, in addition to increase the thermal conductivity of epoxy matrix, the filler pays a major role in reducing the dielectric constant and dielectric loss.

Boron nitride with its hexagonal structure has proved its potential to be used as potential filler material incorporated in polymer matrix for microelectronics applications. With its high thermal conductivity and low dielectric constant property, boron nitride attracts various researcher to work on it and considered it as a potential filler material. Gu et al. [8] reported that thermal conductivity of epoxy matrix increases with increase in boron nitride content. In their study, they found an increment of around 500 % in the value of thermal conductivity with 60 wt. of BN content in epoxy matrix. Hou et al. [9] used boron nitride in its hexagonal for and studied the thermal behaviour of developed material. They also used epoxy as base matrix in their study. They found that incorporation of hexagonal boron nitride significantly enhanced the thermal conductivity of the composites. With little filler content of 30 wt. %, they reported 6 times enhancement in thermal conductivity value. They give the credit of such increase to the modification of surface of boron nitride. In a very recent work, Fang et al. [10] used boron nitride in the form of nano-sheets and reported unbelievable enhancement in the value of thermal conductivity. Pan et al. [11] used surface modification technique over boron nitride particles and introduce them in PTFE matrix. With this, they found enhancement in thermal conductivity of around 2.7 times with 30 vol. % of filler. According to them, enhanced interfacial adhesion and reduced surface hydrophilicity results in improved thermal conductivity. Also, with surface treatment, interfacial polarization is decreased and with this dielectric constant and dielectric loss also reduces.

Aluminium nitride is ceramic filler which is thermally conductive but electrically insulative in nature. Zhou et al. [12] worked on the combination of aluminium nitride and Polymethyl methacrylate (PMMA) matrix. They incorporated AlN over a wide range of filler content varied from 10 vol. % to 70 vol. % and performed detail investigation on thermal conductivity, relative permittivity and dielectric loss. From their analysis, they concluded that the thermal conductivity with maximum filler content reach 1.87 W/(m-K) and relative permittivity for similar filler content restricted to 4.4 (at 1 MHz), respectively. This is considered to be the noticeable improvement in both the properties which was beneficial for certain application. Later Pan et al. [13] also used AlN as filler in PTFE matrix and performed similar study. They reported excellent thermal stability of the developed material. They also found good mechanical properties of the developed material and have reasonably improved dielectric behaviour which were beneficial for the material to be used for microelectronic applications. Against this background, an attempt has been made in this research work to develop aluminium nitride based epoxy composites using simple hand lay-up technique and to study their thermal behavior under controlled laboratory conditions.

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terials and Methods

A. *Material considered*

The presently used matrix is a thermoset polymer epoxy. The epoxy resin Lapox-12 is used 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. 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. The epoxy resin with its corresponding hardener is procured from M/s Atul Limited, Bhopal. It is a liquid, unmodified epoxy resin of medium viscosity which can be used with particular hardener for making composites.

Aluminium nitride is an aluminium based ceramic material is used as one of the filler material in present work. The used aluminium Nitride has been procured from Souvinier Chemical, Mumbai, India. The particle size analysis of the material provides the information that the average sizes of particles are about 60-70 microns. AlN with hexagonal structure is taken as filler material because it proivde best thermal conductivity among other structure of it. It also has low coeffcient of thermal expansion are are reasonably good strength. They are not very reactive substance and also shows good dieelctric properties. Figure 3.4 shows the pictorial microstructural view of aluminium nitride particles.

B. *Composite Fabrication*

Simple hand lay-up technique is used in the present investigation for fabrication of aluminium nitride particles in epoxy matrix. This method is considered as the simplest technique for composite fabrication. The fabrication of composite using hand lay-up method involves following steps:

1. The room temperature curing epoxy resin epoxy resin (Lapox-12) and corresponding hardener (HY 951) are mixed in a ratio 10:1 by weight as recommended.
2. Micro-size aluminium nitride particles were then added to the mixture of epoxy and hardener which is later mixed thoroughly by hand stirring.
3. The uniformly mixed dough is then slowly poured into the mould.
4. The cast is than cured for 8 hours before it was taken from the mould.

Composites were fabricated with different weight fraction of filler ranging from 0 wt. filler i.e. neat epoxy to 50 wt. % filler. The composite fabricated under the investigation is given in table 1.

TABLE I

EPOXY COMPOSITES FILLED WITH ALUMINIUM NITRIDE

S.No.	Composition
1	Neat Epoxy
2	Epoxy + 10 wt % Aluminium Nitride
3	Epoxy + 20 wt % Aluminium Nitride
4	Epoxy + 30 wt % Aluminium Nitride
5	Epoxy + 40 wt % Aluminium Nitride
6	Epoxy + 50 wt % Aluminium Nitride

C. *Characterization*

Thermal conductivity of the fabricated composites are measured by Unitherm Model 2022. The tests are in accordance with ASTM E-1530 Standard. Glass

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tion temperature and Coefficient of thermal expansion of the composites are measured with a Perkin Elmer DSC-7 Thermal Mechanical Analyzer. During the measurement, the specimen is heated from 30 to 150°C at a heating rate of 5°C/min.

III. RESULTS AND DISCUSSION

A. Effective thermal conductivity

Polymers are known for its intrinsic low thermal conductivity value. There thermal conductivity are generally in the range of 0.1 W/m-K to maximum 0.5 W/m-K. With this low thermal conductivity, there applications in microelectronics are questionable.

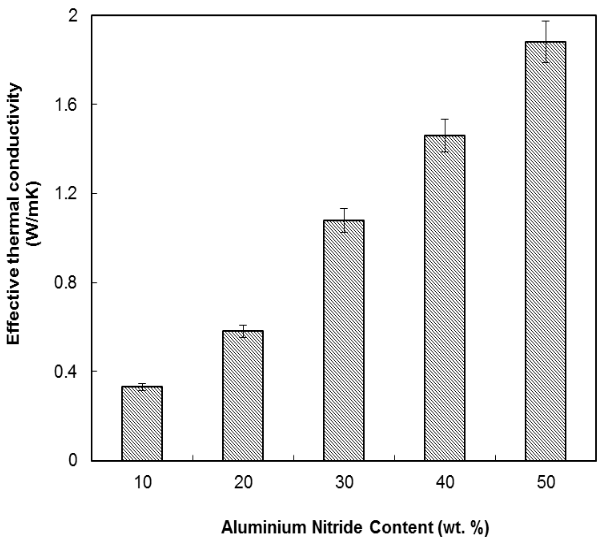

Fig. 1 Effective thermal conductivity of epoxy/aluminium nitride composites

Epoxy also come in the same category and possesses intrinsic thermal conductivity of 0.211 W/m-K. To improve this quantity, aluminium nitride in the form of micro particulates were added. By the addition of AlN, the thermal conductivity of the combination increases. The same can be seen in figure 1. The rate of increase in purely a function of content of aluminium nitride i.e. with increase in content of the aluminum nitride, thermal conductivity increases. For low filler content of 20 wt. %, the rate of increase of thermal conductivity is low, i.e. conductivity of neat epoxy reaches to 0.58 W/m-K with this filler content.

B. Glass Transition Temperature

Glass transition temperature of epoxy/AlN composite as a function of AlN content is shown in figure 2. be of 25.4 % which is an appreciable improvement.

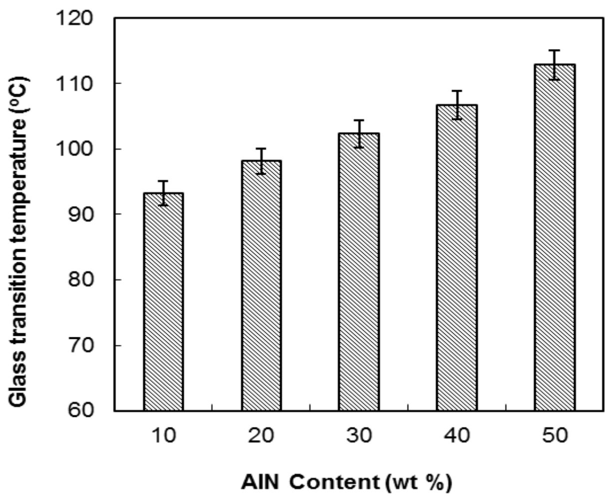

Fig. 2 Glass transition temperature of epoxy/ aluminium nitride composites

It is an important property of polymer, as at this temperature polymer changes its behaviour from glassy state to rubbery state. When this change occurs, material suddenly changes its property and become brittle. With this brittle nature, it will be very difficult to handle the material and little impact may break it as once the material cross the glass transition temperature, it property will not again change from brittle to rubbery even if the temperature falls. From the figure it is clear that the glass transition temperature of epoxy composites was

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r as compared to that of neat epoxy and it increases with increase in AlN content. Glass transition temperature increases from 90 °C for neat epoxy to 112.9 °C for 50 wt. % AlN filled epoxy.

C. Coefficient of thermal expansion

Coefficient of thermal expansion is an important property where material undergoes continuous thermal loading variation.

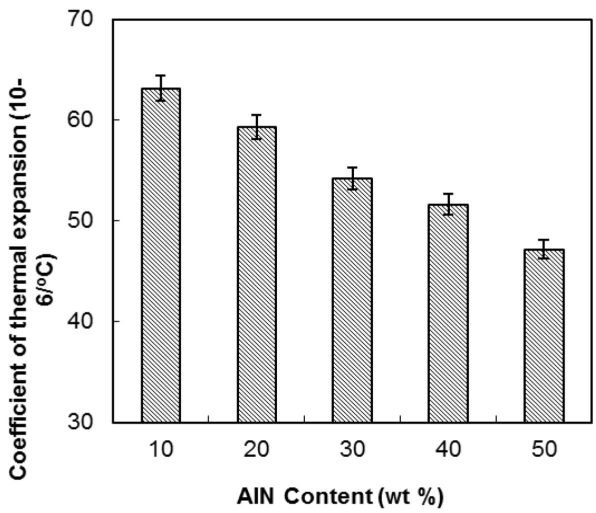

Fig. 3 Coefficient of thermal expansion epoxy/ aluminium nitride composites

Keeping this in mind, inclusion of AlN provided is beneficial as with increase in content of AlN, CTE of epoxy matrix decreases. The same can be seen in figure 3. It is expected because intrinsic CTE of AlN is much lower than that of epoxy matrix. Also, the addition of AlN particles into epoxy results in a reduction in the value of CTE of the composites due to the restricted mobility of the polymer molecules arising out of adsorption of AlNsurfaces. The CTE of the composite reduces from 66 × 10^{-6}/°C to 47.2 x 10^{-6}/°C for 50 wt. % of AlN. The decrement observed in this analysis is 28.5 %.

IV. CONCLUSIONS

This experimental investigation has led to the following specific conclusions:

1) The thermal conductivity of epoxy resin increases with increase in AlN content. By addition of 50 wt. % AlN, thermal conductivity of epoxy matrix increases from 0.211 W/m-K to 1.88 W/m-K. This improvement is of around 790 % over neat epoxy.

2) Glass transition temperatures of epoxy resin increases when AlN filler is added into it. Glass transition temperature increases from 90 °C for neat epoxy to 112.9 °C for 50 wt. % AlN filled epoxy.

3) Coefficient of thermal expansion of epoxy matrix decreases with increase in AlN fillers. The CTE of the composite reduces from 66 × 10^{-6}/°C to 47.2 × 10^{-6}/°C for epoxy based composite incorporated with 50 wt. % of AlN.

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