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Thermal Properties of Polyester Composites Filled with Micro-sized Aluminium Oxide Particulates

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***Abstract*— This investigation consists of the development of polyester composites filled with micro-sized aluminium oxide particulates at varied content and the investigation of the thermal properties of the fabricated samples. The present work consists is an experimental investigation where different thermal properties are evaluated. Here, a set of composites is developed with a filler loading ranging from 0 wt. % to 50 wt. % using a simple hand lay-up method. The properties investigated are thermal conductivity, glass transition temperature and coefficient of thermal expansion of the developed material. From the experimental results, it is found that the inclusion of aluminium oxide particulates in the polyester resin improves the different thermal properties of the composites. The properties like glass transition temperature and thermal conductivity of the composites increase as the content of the filler in the resin increases. Against that, the coefficient of thermal expansion of the material gainfully decreases as the content of filler in the matrix decreases.**

***Keywords*—Aluminium oxide, formatting, polyester, polymer composite, thermal properties**

I. Introduction

Heat management is one of the most crucial challenges in recent microelectronic technology as the power density of electronic devices is rapidly increasing due to the miniaturization and integration of such devices [1]. Also, it operates at high frequency and high-power conditions where large heat flux is generated. For better performance of the devices, the efficient removal of accumulated heat is essential to maintain device efficiency and longer lifespan. If not, power leakage, malfunctioning or even failure of the device may take place [2]. Materials used for microelectronic packaging applications need to simultaneously fulfil diverse requirements such as high thermal conductivity to dissipate the generated heat effectively and quickly, low coefficient of thermal expansion to avoid thermal fatigue, high glass transition temperature for good dimensional stability, moderate dielectric constant to reduce signal propagation delay, low dielectric loss for better device performance, good moisture absorption resistance and good mechanical flexibility [3]. Polymeric materials always play an important role in microelectronic applications because of their ease of processing, low cost, low dielectric constant, hydrophobic and very good adhesive properties [4]. However, common polymers available cannot effectively dissipate the generated heat because they have low thermal conductivity. Also, the high coefficient of the thermal expansion of the polymer results in thermal failure and the low glass transition temperature does not provide thermal stability [5].

The past work concerning thermal conductive polymer composites are mainly focused on the use of various kinds of fillers such as aluminium nitride (AlN) [5, 6], aluminium oxide (Al_2O_3) [7], boron nitride (BN), $Sm_2Si_2O_7$ [3], magnesium silicate (Mg_2SiO_4) [4], Silicon nitride (Si_3N_4) [8], silicon carbide (SiC) [9]. The key properties of the composite material are dependent on various parameters such as the content of the filler, the number of phases, intrinsic properties of phases, the interaction between the filler and matrix and the method of preparation of composites [3]. Among all, the content of filler is mostly considered to be the dominant factor in improving the properties of the filled polymer. Keeping that in mind, little work has been reported. Kushwaha et al. [10] contribute to the field of microelectronic applications by fabricating and characterizing hexagonal boron nitride (hBN)/polyester composites. Published in Polymer Composites, the study investigates the influence of filler loading and surface modification on composite properties. The results reveal a systematic enhancement in thermal conductivity with increasing hBN loading, reaching 6.85 W/m-K at 15 wt. % loading. Surface modification further improves the interfacial adhesion, enhancing mechanical properties. Notably, tensile strength and modulus show improvements of 38% and 26%, respectively, with surface modification at 10 wt. % loading. This work not only advances the understanding of hBN/polyester composites for microelectronic applications but also provides valuable insights into tailoring their properties through filler loading and surface modifications. Wei et al. [11] present a significant contribution by enhancing the thermal conductivity of epoxy composites through the incorporation of aluminium nitride honeycomb reinforcements. The study demonstrates impressive results, revealing a substantial increase in thermal conductivity. The constructed honeycomb lead to an enhanced thermal conductivity of 5.86 W/m-K. Balaji et al. [12] present a comprehensive study focusing on the thermal characteristics of polymer resin reinforced with rice husk and aluminium nitride filler composites. The research combines experimental and computational analyses to provide valuable insights. The results highlight a significant enhancement in thermal conductivity, with the composite incorporating rice husk and aluminium nitride filler exhibiting an improved thermal conductivity of 0.94 W/m-K.

Micro-sized aluminium oxide is being manufactured for a wide variety of uses in various industries. However, its potential as a reinforcing element in polymers has not been adequately explored. Micro-sized aluminium oxide has several advantages and can be preferred over other particles in many engineering applications due to their low surface area high strength, high thermal conductivity, low coefficient of thermal expansion etc. Given this, the present work concentrates on the development of polyester-based polymeric composites with micro-sized aluminium oxide as a filler material. The effect of the addition of the aluminium oxide micro-particulates on the thermal properties is investigated in the present work. The properties investigated are thermal conductivity, glass transition temperature and coefficient of thermal expansion.

II. MATERIALS AND METHOD

Unsaturated isophthalic polyester supplied by Carbon black composites, Mumbai India, is the matrix material in the present investigation. Polyester resin is used with its corresponding accelerator i.e. cobalt accelerator and catalyst i.e. MEKP catalyst. The advantage of polyester resin composites is that they can be cured in a variety of ways without altering the physical properties of the finished part. Their advantages include low viscosity, low cost, and fast cure time. Aluminium oxide (Al2O3) is an aluminium-based ceramic material that has been used as a filler in the present work. It is an inorganic material. It also possesses high strength and stiffness. It is therefore chosen as the primary filler material with an average size of 20 microns which is procured from Rankem Corporation Limited located in New Delhi, India. The preparation of the different categories of composite with the simple hand lay-up method. Multiple sets of samples are prepared by varying the content of the aluminium oxide filler in the polyester resin. The different sets of composites prepared are shown in Table 1.

TABLE I. LIST OF SAMPLES PREPARED

S. No.	Composition
1	Neat Polyester
2	Polyester + 10 wt. % Al_2O_3
3	Polyester + 20 wt. % Al_2O_3
4	Polyester + 30 wt. % Al_2O_3
5	Polyester + 40 wt. % Al_2O_3
6	Polyester + 50 wt. % Al_2O_3

The thermal conductivity of the composite materials is measured by the Unitherm Model 2022. The tests are conducted as per ASTM E-1530 Standard. The device works on the principle of guarded heat flow meter method. The coefficient of thermal expansion (CTE) is measured according to ASTM D 696 with the help of a Perkin–Elmer thermal mechanical analyser. The glass transition temperature is evaluated using the same thermal mechanical analyser per ASTM D 3418 standard.

III. RESULTS AND DISCUSSION

The effective thermal conductivities of polyester composites filled with micro-sized aluminium oxide with filler weight fraction ranging from 0 to 50 wt. % as a function of temperature is studied in the present work. The thermal conductivity is measured in the present work for four different temperatures i.e. 30 ºC, 40 ºC, 50 ºC and 60 ºC. Figure 1 shows the variation in thermal conductivity for different filler loading and at different working temperatures.

It can be seen from the graph that with an increase in the content of Al2O3, the conductivity of the composite body increases irrespective of the working temperature. This is obvious as the intrinsic thermal conductivity of aluminium oxide is much greater as compared to the intrinsic thermal conductivity of polyester matrix. This results in an increase in the conduction capability of the fabricated composite. The maximum value of effective thermal conductivity among the various sets of fabricated composites is obtained when 50 wt. % of aluminium oxide is incorporated in the polyester matrix i.e. 0.883 W/m-K at an ambient temperature of 60 oC which is maintained inside the testing chamber. This increment is around 256 % against neat polyester.

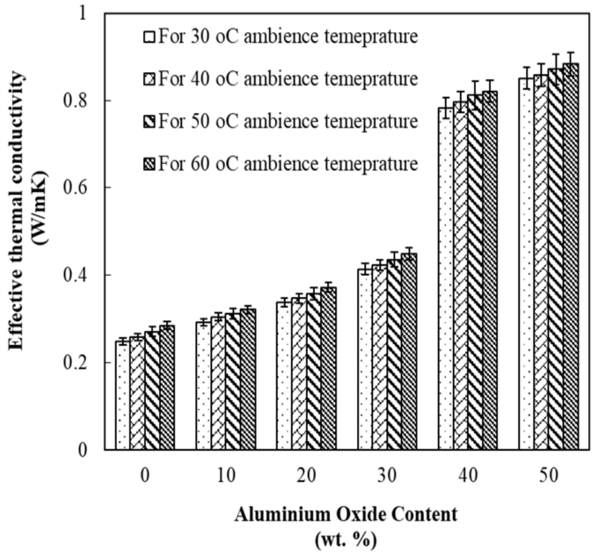

Figure 1: Thermal conductivity of polyester/aluminium oxide composite

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Further, with low filler content, the value of thermal conductivity increases very marginally which is mainly due to the random dispersion of filler material in the polyester resin. As the weight fraction of filler increases, the distance between the particles reduces and after reaching a particular filler content the particles begin to touch each other. This physical interaction among the particles gives rise to the formation of a path which is conductive and once when this conductive path is established, a sudden increment in the value of thermal conductivity is observed. In the present case, the percolation threshold reaches when filler content increases beyond 30 wt. %.

Further study of thermal conductivity includes the dependence of thermal conductivity on ambient temperature. From the figures, it can be observed that with an increase in ambient temperature, the thermal conductivity of the sample increases for the same filler content. This is mainly because, with an increase in temperature, the prime factor for conduction i.e. vibration of particle increases and hence improves the overall conductivity of the samples. This behaviour is universally proven for all kinds of solid bodies.

Figure 2 presents the variation of glass transition temperature with aluminium oxide content. It is observed that the glass transition temperature of neat polyester is about 71.2 °C and it gradually increases to 89.6 °C for a filler loading of 50 wt. %. A maximum increase of 18.4 ºC in glass transition temperature is obtained as the aluminium oxide content increases from 0 wt. % to 50 wt. %. A maximum increase of about 25.84 % in glass transition temperature is obtained. Usually, the addition of such filler increases the glass transition temperature of the composites, which results from the interaction between a filler and a polymer by forming a network structure between them. Because of this network structure, the movement of molecular segments is limited and hence the glass transition temperature increases.

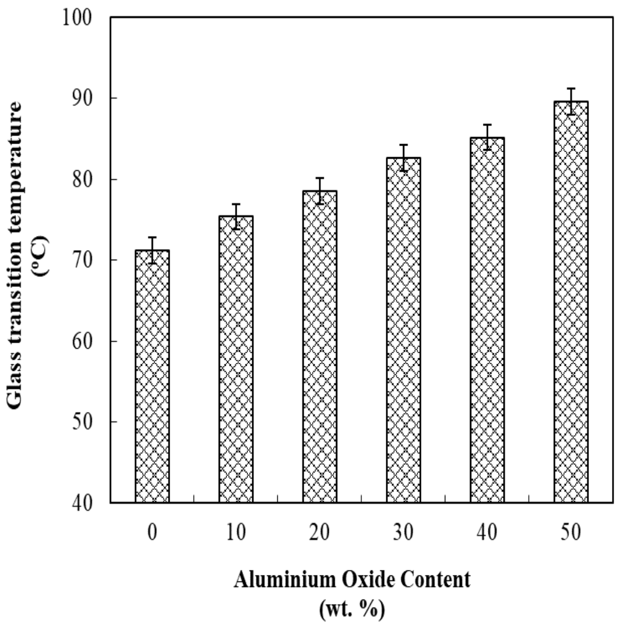

Figure 2 Glass transition temperatures of polyester/ aluminium oxide composites

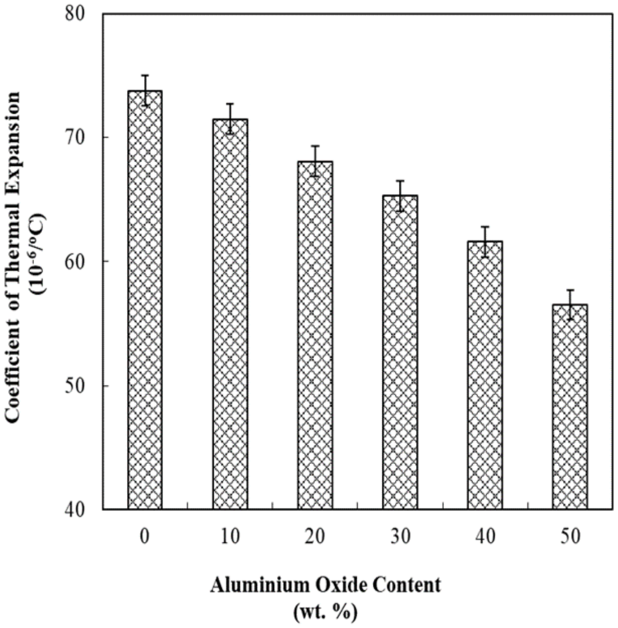

Figure 3 Coefficient of thermal expansion of polyester/ aluminium oxide composites

Figure 3 shows the coefficient of thermal expansion of the entire fabricated composite specimen under investigation. It is clear from the graph that the coefficient of thermal expansion of the composite decreases with an increase in the content of aluminium oxide. The coefficient of thermal expansion of neat polyester was measured to be 73.8×10^{-6}/°C. This value of the coefficient of thermal expansion reduces to only 71.5×10^{-6}/°C when 10 wt. % of aluminium oxide is added to the polyester. The decrement in the value of the coefficient of thermal expansion progresses as the filler content increases and reaches 56.5×10^{-6}/°C for a maximum aluminium oxide content of 50 wt. %. A maximum decrease of about 23.44 % in the coefficient of thermal expansion is obtained.

IV. CONCLUSION

This present investigation on particulate Aluminium oxide/polyester composites has led to the following conclusions:

1. Aluminium oxide possesses ample reinforcing potential to be used as a filler material in polyester matrix composites and successful fabrication of polyester matrix composites reinforced with aluminium oxide particles is possible by simple hand-lay-up technique.

2. Effective thermal conductivity of polyester increases with an increase in aluminium oxide content. Thermal conductivity also increases with an increase in ambient temperature. The maximum value of thermal conductivity is obtained for a maximum filler content of 50 wt. % at an ambient temperature of 60 ºC. The maximum value of thermal conductivity obtained is 0.883 W/m-K.

3. A significant improvement in the glass transition temperature is noticed with the incorporation of aluminium oxide in the polyester matrix. Improvement of around 25.84 % is registered for a maximum filler content of 50 wt. %. The maximum value obtained is 89.6 ºC.

4. It is also observed that the coefficient of thermal expansion of polyester is reduced by the addition of aluminium oxide filler. It is observed that a drop of 23.44 % in the coefficient of thermal expansion of polyester is obtained with 50 wt. % inclusion of micro-size aluminium oxide. The minimum value obtained is 56.5 10^{-6}/ºC.

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