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EFFECT OF ADDITION OF MICRO-SIZED BORON NITRIDE ON THE THERMAL PROPERTIES OF THE POLYESTER COMPOSITES

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ABSTRACT

Polyester composites reinforced with micro-sized hexagonal boron nitride (hBN) particles were developed to evaluate their thermal performance for potential thermal management and electronic packaging applications. The composites were fabricated using a hand lay-up technique with filler loading varied from 0 to 40 wt.%. Thermal properties such as thermal conductivity, glass transition temperature (Tg), and coefficient of thermal expansion (CTE) were experimentally investigated under controlled laboratory conditions. The results reveal a significant improvement in thermal conductivity with increasing hBN content due to the formation of thermally conductive pathways within the polyester matrix. The enhancement in thermal conductivity is limited for small filler loading, but once the filler content increases beyond a certain point, due to the formation of a conductive path, there is a sudden increment in the value of thermal conductivity. The glass transition temperature increases steadily with filler loading, indicating restricted polymer chain mobility, while the coefficient of thermal expansion decreases substantially owing to the low CTE of hBN particles. This shows that the prepared composite material can withstand a higher temperature limit with a minimum thermal fatigue. The combined improvement in thermal conductivity, thermal stability, and dimensional stability highlights the suitability of polyester/hBN composites for advanced thermal and electronic applications.

Keywords; Polyester Composite, Hexagonal Boron Nitride, Thermal Conductivity, Glass Transition Temperature, Coefficient Of Thermal Expansion.

1. INTRODUCTION

Nowadays, composite materials are everywhere as they extend their horizons in almost every branch of engineering and science. Polymer matrix composites (PMCs) are the best-established form of advanced composite materials. Of the two classes of polymers used as matrices, thermosets and thermoplastics, thermosets dominate the market for structural applications [1, 2]. Current research is being conducted on composite materials for thermoset as a matrix material. In the present work, epoxy is selected as a matrix material and a ceramic material, i.e. silicon nitride, is used as a filler material. Silicon nitride has been used for the development of composite materials with polymer as a base matrix material in the past.

Gu et al. [3] used micro-sized boron nitride in combination with epoxy resin for the development of polymer composites. The properties investigated by them are mechanical, thermal and dielectric properties. They also compared the two different categories of composites prepared with raw-boron nitride and surface-modified boron nitride on the basis of the investigated properties. They used a silane coupling agent to modify the surface of boron nitride. In their analysis, they found that the inclusion of boron nitride improves the thermal conductivity and thermal stability of the epoxy resin as a function of filler loading. Huang et al. [4] demonstrate that the incorporation of polyhedral oligo silsesquioxane-modified boron nitride nanotubes into epoxy resin yields exceptional dielectric properties, with a dielectric constant as low as 2.7, enhancing electrical insulation in electronic devices. Zhou et al. [5] used flaky hexagonal boron nitride as a filler material and developed epoxy-based composites. They also used a silane coupling agent to modify the surface of the filler material. In their analysis, they found that the incorporation of surface-modified hBN successfully increases the glass transition temperature and improves the thermal stability of the material. While studying the thermal conductivity, they found that the incorporation of filler appreciably increases the thermal conductivity of the epoxy resin. Kim and Kim [6] developed epoxy/boron nitride composites and studied the thermal conductivity of the material as a function of filler loading. They prepared composites for a high filler loading of up to 80 wt. %. In their analysis, they reported that the thermal conductivity of the epoxy resin increases with filler loading till the filler content reaches 70 wt. % and decreases when the content of filler increases beyond it. Hou et al. [7] also investigated the surface-modified boron nitride and epoxy resin composites. The silane coupling agent is used to modify the hexagonal boron nitride. They found noticeable differences between the thermal conductivity of the composites prepared with raw hBN and surface-modified hBN. The composite prepared with surface-modified hBN delivers better thermal conductivity as compared to its counterpart.

Gu et al. [8] achieved outstanding thermal conductivity, with values as high as 16.7 W/mK, making these composites excellent candidates for heat management in high-performance electronics. Jang et al. [9] focus on boron nitride (BN)/epoxy composites and the effects of surface modification using silane coupling agents. Through this surface modification technique, the thermal conductivity of the BN/epoxy composite was significantly enhanced. The untreated composite had a thermal conductivity of 0.56 W/m·K, whereas the modified composite achieved a remarkable value of 3.02 W/m·K. Fang et al. [10] used polydimethylsiloxane (PDMS) as a matrix material with boron nitride foam (BNF) and boron nitride nanosheets (BNNS) as a filler material for the fabrication of a composite material. They investigated the synergistic effect of the addition of the fillers on the mechanical, thermal and dielectric properties of the composites. Jiang et al. [11] focus on enhancing the thermal and dielectric properties of epoxy composites by incorporating polymer-modified hexagonal boron nitride. The thermal conductivity of the pristine epoxy was 0.24 W/m-K, but with the addition of polymer-modified h-BN, it increased to an impressive 2.12 W/m-K, showcasing a nearly nine-fold enhancement.

Pan et al. [12] evaluated the thermal conductivity of the fabricated samples as a function of filler loading and atmospheric temperature. From the investigation, they found that the thermal conductivity of the material improves with the addition of the fillers as a linear function of filler loading. Lee et al. [33] used a combination of epoxy with nano-sized hexagonal boron nitride for the development of a composite that can be used for different microelectronics applications. The thermal conductivity of the nanocomposites was dramatically improved, reaching an impressive value of 13.2 W/m-K, a substantial increase compared to conventional epoxy materials. Agarwal and Chandrakar [14] employed epoxy as the base matrix, enhancing it with two distinct types of hexagonal boron nitride (hBN). Their approach involved integrating raw hBN in one set and silane-modified hBN in another, yielding two distinct categories of composite materials. Thermal conductivity increased with hBN loading and was further improved by silane treatment through better phonon transport across interfaces. Additionally, the dielectric constant and loss were favourably influenced, making the composites suitable for electronic and thermal management applications. Li et al. [15] used boron nitride nanosheets (BNNSs) and sphere BN (S-BN) in a polydimethylsiloxane matrix for the development of a new class of polymeric composites. The prepared BNNSs/PDMS composites exhibited an impressive thermal conductivity of 1.16 W/m-K when containing 35 wt. % fillers, a value approximately five times greater than that of pure PDMS and even surpassing the thermal conductivity of S-BN/PDMS composites. Yu et al. [16] improved the thermal conductivity of carboxylated acrylonitrile-butadiene rubber by adding the boron nitride filler, which was surface-modified with tannic acid. They decorate the surface of BN with noncovalent TA functionalization, while preserving the BN platelets' surface structure, ensuring the retention of high intrinsic thermal conductivity. Furthermore, this treatment enhances the compatibility between the filler and the matrix and promotes the formation of thermally conductive pathways within the composites.

Bashir et al. [17] used hexagonal boron nitride (h-BN) sheets that underwent functionalization using diverse amine compounds. The results demonstrate that both ortho-phenylenediamine (o-PDA)-treated o-PDA-BN and para-phenylenediamine (p-PDA)-treated p-PDA-BN when incorporated into TPU composites at a high filler loading of 45 wt. %, exhibit significantly higher through-plane thermal conductivity values of 2.06 and 1.96 W/m-K, respectively. Wang et al. [18] studied the effect of micro- and nano-sized boron nitride particles on the microstructure, breakdown strength, and thermal and mechanical properties of epoxy resin. The analysis of breakdown strength and thermal conductivity revealed that when the composite consisted of 30 wt% micro-sized BN and 20 wt% nano-sized BN, the resulting BN/epoxy composite exhibited a substantial thermal conductivity of 1.52 W/m·K. Yu et al. [19] developed a highly thermally conductive polymer composite by building a two-level adjustable boron nitride (BN) network with leaf venation structure in an epoxy resin matrix. From the analysis of the thermal conductivity, they found that at a BN loading of 35.9 wt. %, the composite exhibits in-plane and cross-plane thermal conductivities of 10.20 W/m-K and 4.95 W/m-K, respectively. Zhou et al. [20] used a groundbreaking approach to achieving exceptional thermal conductivity in polymer composites, with substantial implications for various industries, especially electronics and thermal management. The method employed for fabricating the composites is particularly noteworthy.

Li et al. [21] present an innovative approach to enhancing the thermal conductivity of polymer composites, a development of significant importance for various applications, particularly in the field of thermal management. At a relatively low BNNS loading of 9.8 wt. %, the composite film achieved a substantial enhancement in thermal conductivity, reaching 1.62 W/m-K. He et al. [22] discussed a significant breakthrough in enhancing the thermal properties of composite materials. The researchers achieved high out-of-plane thermal conductivity by carefully aligning boron nitride nanosheets (BNNS) within an epoxy matrix using a magnetic field during the curing process. At a relatively low BNNS loading of 30 wt. %, the composite films demonstrated remarkable out-of-plane thermal

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conductivity values, reaching up to 7.85 W/m-K. Do et al. [23] explore the influence of different processing techniques on the thermal conductivity and mechanical properties of polymer composites containing hexagonal boron nitride (h-BN). The study is valuable as it provides insights into the most effective fabrication method for achieving the desired material properties. Jang et al. [24] offer a compelling approach to address the critical need for thermally conductive, electrically insulating materials in various applications. The study focuses on an eco-friendly and scalable method for fabricating such composites. He et al. [25] fabricated thermally conductive polymer composites using hexagonal boron nitride (hBN) as the primary filler. The composites achieved an impressive thermal conductivity of 5.2 W/m·K, nearly 10 times higher than pristine polymers. Nie et al. [26] reported the development of advanced polymer composites using POSS-functionalized boron nitride nanosheets (BNNS-POSS) as fillers. By enlarging the nanosheet size and improving surface compatibility, the researchers achieved superior filler dispersion within the polymer matrix. Against this background, in the present work, a class of composite is fabricated in which the continuous phase is a thermoset polyester matrix and the discontinuous phase is micro-sizedboron nitride particles. A simple hand lay-up method is used for the fabrication of composites with a wide range of filler content. The properties evaluated are thermal conductivity, glass transition temperature and coefficient of thermal expansion.

2. MATERIALS AND METHODS

Unsaturated isophthalic polyester resin supplied by Carbon Black Composites, Mumbai, India, was used as the matrix material. Micro-sized hexagonal boron nitride powder served as the reinforcement. Methyl ethyl ketone peroxide (MEKP) was used as a catalyst, and cobalt accelerator was used to promote curing. The composites were fabricated using the hand lay-up technique. Predetermined amounts of hBN particles (10, 20, 30, and 40 wt. %) were mixed with polyester resin to ensure uniform dispersion. The accelerator and catalyst were added sequentially, followed by thorough mixing. The mixture was poured into silicon-coated moulds and allowed to cure at room temperature. After demolding, the specimens were post-cured for 24 hours before testing. Tensile, flexural, and compressive tests were conducted according to relevant ASTM standards using a universal testing machine. Hardness measurements were performed using a Shore-D hardness tester. For each composition, five specimens were tested, and average values were reported.

3. RESULTS AND DISCUSSION

A. Thermal Conductivity

Figure 1 illustrates the variation of thermal conductivity of polyester/hBN composites with filler loading. Neat polyester exhibits a low thermal conductivity of approximately 0.323 W/m·K. With the addition of hBN particles, the thermal conductivity increases progressively and reaches a maximum value of 1.823 W/m·K at 40 wt.% filler content, corresponding to an improvement of about 464%. The enhancement in thermal conductivity is primarily attributed to the high intrinsic thermal conductivity of hBN and the formation of conductive networks within the polymer matrix. At lower filler loadings, hBN particles are isolated and heat transfer occurs mainly through the polymer matrix, resulting in modest improvement. As the filler content increases beyond a critical threshold, particle-to-particle contact becomes significant, leading to the formation of continuous thermally conductive pathways. This percolation phenomenon results in a sharp increase in heat conduction efficiency.

B. Glass Transition Temperature

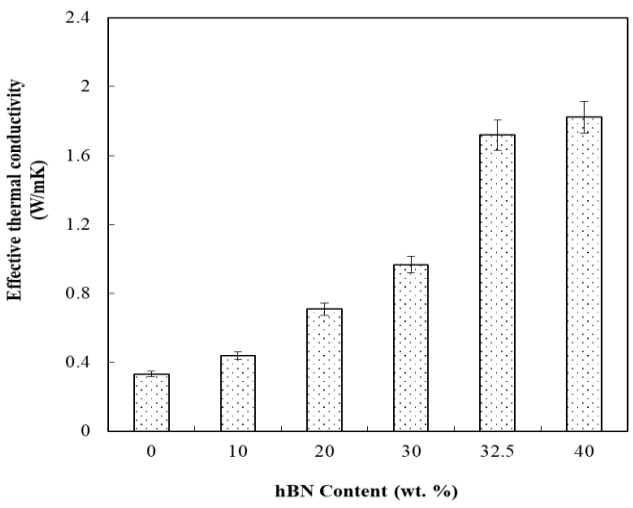

Figure 1: Thermal conductivity of polyester/hBN composites

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The glass transition temperature (Tg) of polyester/hBN composites as a function of filler loading is shown in Figure 2. Neat polyester exhibits a Tg of approximately 81.5 °C. The incorporation of hBN particles leads to a steady increase in Tg, reaching a maximum value of 97.2 °C at 40 wt.% filler content, representing an enhancement of about 19.26%.The increase in Tg is attributed to the reduction in free volume within the polyester matrix caused by the presence of rigid hBN particles. The fillers restrict the mobility of polymer chains and increase the crosslink density, thereby requiring higher thermal energy to initiate segmental motion. Improved interfacial interactions between the matrix and filler further contribute to the observed increase in glass transition temperature.

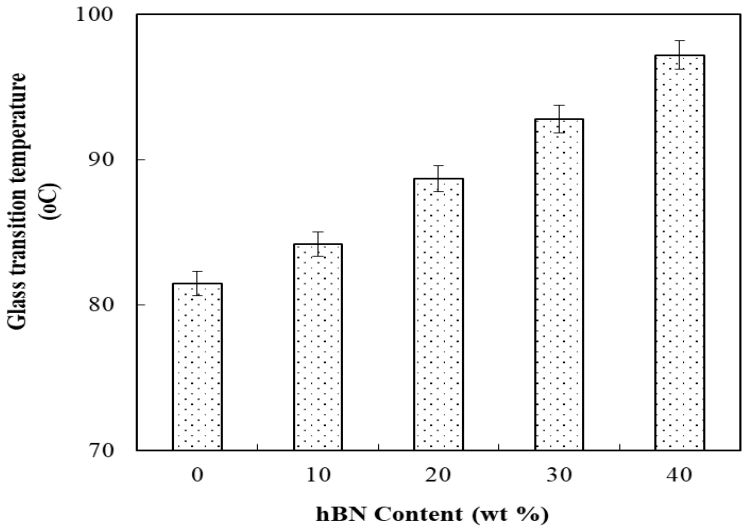

Figure 2: Glass transition temperature of polyester/hBN composites

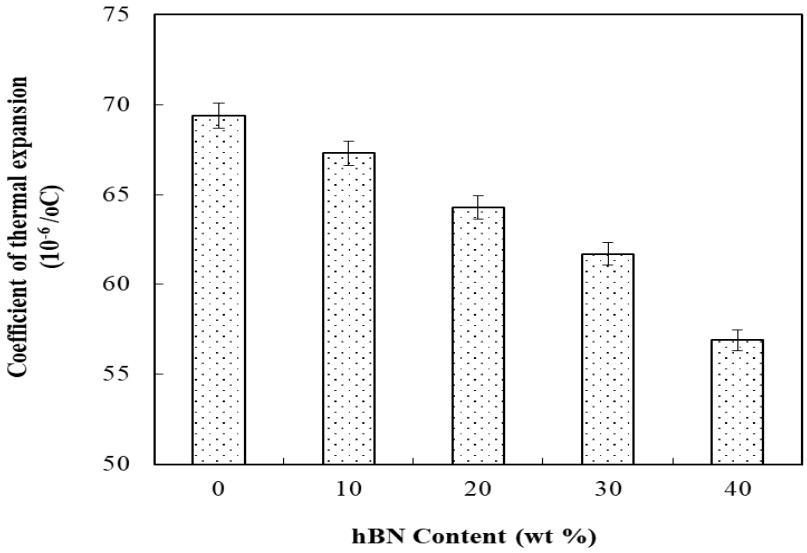

Figure 3: Coefficient of thermal expansion of polyester/hBN composite

C. Coefficient of Thermal Expansion

Figure 3 presents the variation of the coefficient of thermal expansion (CTE) of polyester/hBN composites with filler content. Neat polyester exhibits a relatively high CTE of 69.4 × 10^{-6} /°C. With increasing hBN loading, the CTE decreases consistently and reaches a minimum value of 56.9 × 10^{-6} /°C at 40 wt.% filler content, corresponding to a reduction of approximately 18%.This reduction in CTE is mainly due to the significantly lower CTE of hBN compared to polyester. The incorporation of rigid ceramic particles constrains the thermal expansion of the polymer matrix by limiting molecular mobility. As the filler content increases, the composite structure transitions from a loosely bound polymer network to a more rigid and dimensionally stable system. Enhanced interfacial bonding further improves the resistance to thermal deformation.Overall, the thermal results demonstrate that hBN is an effective filler for improving heat conduction, thermal stability, and dimensional stability of polyester composites. The simultaneous enhancement of thermal conductivity and reduction of CTE is particularly advantageous for applications involving thermal cycling and heat dissipation.

4. CONCLUSION

This experimental investigation on the development of polyester/hBN composites, along with their thermal characterization has led to the following specific conclusions:

1. It is possible to develop a polyester composite reinforced with micro-sized hBN by a simple hand lay-up method.
2. The addition of hBN remarkably enhances the thermal conductivity of the polyester resin. With the inclusion of 40 wt. % hBN, the thermal conductivity reaches a maximum value of 1.823 W/m-K. Further, the percolation took place at 32.5 wt. % hBN loading where the value of thermal conductivity increases sharply.
3. The glass transition temperature of the polyester is 81.5 ^{o}C, and the same increases to 97.2 ^{o}C with the addition of 40 wt. % hBN registering an improvement of 19.26 % over the neat polyester. This shows the application of material at elevated temperatures.
4. The coefficient of thermal expansion of the polyester resin is quite high, i.e. 69.4×10^{-6} /^{o}C. With the inclusion of hBN, it is reduced. For polyester/hBN composite, the minimum CTE obtained is 56.9×10^{-6} /^{o}C for 40 wt. % hBN. This shows that the material will encounter less thermal fatigue at fluctuating temperatures.

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