Mechanical and Thermal Behaviour of Epoxy/Hexagonal Boron Nitride/Short Sisal Fiber Hybrid Composites

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Abstract. Hybrid composite i.e. surface modified hexagonal boron nitride (hBN) and short sisal fiber reinforced in epoxy matrix is fabricated using hand lay-up method. The effect of surface modified hBN filler and sisal fiber content on mechanical and thermal properties of epoxy based hybrid composites were investigated in this paper. The main aim of the investigation is to develop a material which can find its application in microelectronic components. As per the requirement of microelectronic industry, the material should possess high thermal conductivity. Hence, thermal conductivity of epoxy increases with increase in hexagonal boron nitride content. Inspite of insulative nature of sisal fiber, the study shows that its inclusion in combination with hBN enhances the thermal conductivity if the content of both the fillers were properly selected. Other thermal property like coefficient of thermal expansion and glass transition temperature appreciably improves when combination of fillers were added in epoxy matrix. Mechanical properties under study i.e. tensile strength and compressive strength also enhances when combination of sisal fiber and hBN were incorporated as compared to when single filler hBN were used. Hence, usage of hybrid filler as reinforcement in epoxy improve overall mechanical and thermal property of the developed material.

1. Introduction
With rapid development in modern microelectronic industries, there is a continuous increase in power density of the electronic devices. The power density increases mainly because of everlasting trend towards miniaturization of such devices. Miniaturization results in overheating of the device and hence will cause thermal problem if the generated heat will not dissipated effectively and efficiently. This further cause many other problems in the performance of the device like power leakage, deterioration of performance and even device failure [1]. Apart from reasonably high thermal conductivity for heat dissipation, other requirement of material to be used in electronic devices are low dielectric constant for proper signal distribution with high speed or low delay time and to diminish probability of short circuiting, low coefficient of thermal expansion to avoid thermal fatigue, high thermal stability to withstand sudden rise of temperature, high glass transition temperature to maintain its crystal structure at elevated temperature and reasonably good mechanical properties to protect them from external damages or impact failure. All the mentioned properties to be achieved in monolithic material are difficult. With this, demand of reliable and high performance packaging material is increasing. Thus,
proper selection of material is necessary to enhance device efficiency and protect them from premature failure [2].

This leads to the demand of developing composite material. Polymers have excellent dielectric properties with added advantage of ease of process ability at low cost. But because of low thermal conductivity of polymers, their applications in electronic components are limited [3]. Ceramic fillers with good thermal conductivity and low dielectric constant are of interest among the researchers to be used as filler material in polymeric matrix. The various ceramic fillers used in different polymeric matrix for similar applications are silicon carbide [4], aluminium oxide [5], aluminium nitride [6], boron nitride [7], montmorillonite [8] etc. In all the above study, filler content govern the thermal conductivity of the developed material. However, increasing the filler content to higher value results in brittle material with low melt flow index, increased porosity, increase in water absorption rate and have adverse effect on dielectric properties [9]. Thus, to design and develop an electrically insulated and high thermal conductive composite without compromising its mechanical properties is important.

In view of this, in present investigation, hybrid filler were used in epoxy matrix to develop such composites. In the process of developing such composites, it has been kept in mind to use material which is environmental friendly. Synthetic and natural fibers reinforcement in polymeric composites is reported to increase the mechanical behavior by various researchers [10–12]. They are electrically insulated as well. Hence, a natural fiber i.e. sisal fiber is utilized as reinforcement in epoxy matrix together with ceramic filler i.e. hexagonal boron nitride (hBN). Sisal fiber is used mainly to provide ductile behavior to material which would have been brittle in nature if only hBN were used in epoxy matrix as seen in previous work. Also, sisal fiber is highly insulative which controls the dielectric property of the material. The main focus of this work is to investigate the improvement in mechanical and thermal behavior provided by the addition of natural fiber to the hBN/epoxy composites.

2. Experimental details
Matrix material used in present investigation is Lapox L12 with its corresponding hardener K6. The matrix material used is supplied by ATUL India Ltd., Gujarat, India. Hexagonal boron nitride of size 5 microns is used as primary filler material. It is procured from Souvenier Chemicals, Mumbai. It has density of 2.3 g/cc. Sisal fiber in short form i.e. around 4 mm length is used as second filler. The sisal fiber possesses low density of 1.41 g/cc and good tensile strength of approx. 350-370 MPa [13].

Hand lay-up technique is implemented for preparation of epoxy based hybrid composites. The resin and hardener used in present investigation is combined in 10:1 ratio by weight. Surface modified hexagonal boron nitride and short sisal fiber were added to the epoxy-hardener solution. The combination is than mixed manually and slowly. Silane coupling agent of 2wt. % concentration in ethanol is used for surface treatment of hBN [9] and 2 mole aqueous solution of sodium hydroxide is used for surface treatment of sisal fiber [14]. Prepared mixture is than poured slowly in to the mould. Before pouring the mixture in to the mould, coating of silicon spray is mandatory for easy removal of composite after it gets cured. Cast take duration of 8 hours before it gets completely cured and ready for removal from the mould. The list of fabricated composite is presented in table 1.

Tensile strength of the samples is measured as per ASTM D638 and compressive strength is measured as per ASTM D695 standard. Both the tests were performed in Tinius Olsen universal testing machine. Thermal conductivity is measured as per ASTM E-1530 with the help of Unitherm™ Model 2022. Perkin-Elmer thermal mechanical analyzer (TMA-7) is used to measure Coefficient of thermal expansion and glass transition temperature as per ASTM D 618 and ASTM E 831 respectively.

3. Results and discussions
3.1. Tensile strength
The variation in tensile strength on incorporation of hexagonal boron nitride and sisal fiber in epoxy matrix is presented in Figure 1. It can be seen from the figure that when 10 wt. % hBN is added in epoxy matrix, the tensile strength increased from 40.5 MPa for neat epoxy to 45.2 MPa.
Table 1. List of fabricated specimens.

| Set | Composition | Set | Composition | Set | Composition | Set | Composition |
|-----|-------------|-----|-------------|-----|-------------|-----|-------------|
| A1  | Epoxy + 10 wt % hBN | B1  | Epoxy + 20 wt % hBN | C1  | Epoxy + 30 wt % hBN | D1  | Epoxy + 40 wt % hBN |
| A2  | Epoxy + 10 wt % hBN + 1 wt % SF | B2  | Epoxy + 20 wt % hBN + 1 wt % SF | C2  | Epoxy + 30 wt % hBN + 1 wt % SF | D2  | Epoxy + 40 wt % hBN + 1 wt % SF |
| A3  | Epoxy + 10 wt % hBN+ 2 wt % SF | B3  | Epoxy + 20 wt % hBN + 2 wt % SF | C3  | Epoxy + 30 wt % hBN + 2 wt % SF | D3  | Epoxy + 40 wt % hBN + 2 wt % SF |
| A4  | Epoxy + 10 wt % hBN+ 3 wt % SF | B4  | Epoxy + 20 wt % hBN + 3 wt % SF | C4  | Epoxy + 30 wt % hBN + 3 wt % SF | D4  | Epoxy + 40 wt % hBN + 3 wt % SF |
| A5  | Epoxy + 10 wt % hBN+ 4 wt % SF | B5  | Epoxy + 20 wt % hBN + 4 wt % SF | C5  | Epoxy + 30 wt % hBN + 4 wt % SF | D5  | Epoxy + 40 wt % hBN + 4 wt % SF |
| A6  | Epoxy + 10 wt % hBN+ 5 wt % SF | B6  | Epoxy + 20 wt % hBN + 5 wt % SF | C6  | Epoxy + 30 wt % hBN + 5 wt % SF | D6  | Epoxy + 40 wt % hBN + 5 wt % SF |

It has been observed in earlier investigation also that a relatively small quantity of filler can successfully remedy the defects from epoxy self-curing. However, when hBN content increases further, decreasing trend in tensile strength is observed. For maximum hBN loading of 40 wt. %, the tensile strength of composites decreased to 30.8 MPa.

Figure 1. Tensile strength of fabricated samples.

The reduction in tensile strength with high filler loading is due to fact that excessive fillers served as local stress-concentration points in the resin matrix causing in the weakening in tensile strength.
Again, when combination of sisal fiber and hBN is incorporated, it successfully enhances the tensile strength of epoxy matrix but only when the content of sisal fiber is limited. It is observed that when sisal fiber were added in combination with hBN in epoxy, tensile strength of the hybrid composite increases upto 3 wt. % of sisal fiber. When content of fiber increases further, decreasing trend in the tensile strength is observed. Incorporation of 3 wt. % of fiber in combination with 10 wt. % of hBN shows maximum tensile strength of 55.3 MPa. Further increase of sisal fiber decreases the tensile strength which reduces to 47.2 MPa for 5 wt. % sisal fiber.

3.2. Compressive strength
The variation in compressive strength of epoxy composites filled with different content of hexagonal boron nitride and sisal fiber is shown in figure 2. With hBN as single filler is incorporated in epoxy matrix, compressive strength increases from 85 MPa for neat epoxy to 108.7 MPa for maximum loading of 40 wt. % hBN in epoxy which is around 27.9 % increment in compressive strength. This increment increases further when sisal fiber were added in combination with hBN in epoxy matrix. In this case, when 5 wt. % of fiber were added in combination with 40 wt. % hBN in epoxy matrix, compressive strength increases from 108.7 MPa to 118.1 MPa which is an increment of 38.9 % over neat epoxy. Unlike tensile strength, compressive strength always show increasing trend with increase in filler content. The increasing behaviour in compressive strength is attributed to the favorable deformation processes which are facilitated due to the presence of hBN particles and short sisal fiber in the epoxy matrix.

3.3. Thermal conductivity
The variation in thermal conductivity of epoxy composites filled with different content of hexagonal boron nitride and sisal fiber is shown in figure 3. When hBN content was 40 wt. % in epoxy, the thermal conductivity was increased from 0.211 W/m-K for neat epoxy to 1.52 W/m-K. Further when sisal fiber is added in epoxy/hBN combination, the thermal conductivity decreases with increase in sisal fiber upto 2 wt. %. Later, the highest value of thermal conductivity is obtained with a combination of 30 wt. % hBN and 3 wt. % of sisal fiber. The value reported is 1.88 W/m-K. Also
improved value of thermal conductivity is obtained for 40 wt. % hBN and 3 wt. % of sisal fiber which is having a value of 1.55 W/m-K. Apart from the above combination, thermal conductivity decreases with sisal fiber content. This may be attributed to the fact that hBN was well distributed and ordered on the sisal fiber surfaces for a particular combination. This arrangement helped the hBN powder interconnection, resulting in a significant improvement in thermal conductivity [15].

Figure 3. Thermal conductivity of fabricated samples.

Figure 4. Glass transition temperature of fabricated samples.
3.4. Glass transition temperature
The variation in glass transition temperature of epoxy composites filled with different content of hexagonal boron nitride and sisal fiber is shown in figure 4. It is clear from the figure that compared to neat epoxy, glass transition temperature of its composites were higher. It can also be seen that the glass transition temperature depends upon the content of the filler. With single filler hBN, glass transition temperature increases from 73 °C for unfilled epoxy to 93.2 °C for 40 wt. % filled epoxy. Further, with incorporation of sisal fiber in the combination with hBN, glass transition temperature further increases and reaches maximum to 103.3 °C for a reinforcement of 40 wt. % hBN and 5 wt. % sisal fiber. The increment in glass transition temperature with the addition of the filler is because of the reduction in free volume of the polymer. With reduced availability of free volume, mobility of polymer chain is restricted and because of this restriction glass transition temperature of polymer composite increases [16].

3.5. Coefficient of thermal expansion
The variation in coefficient of thermal expansion of epoxy composites filled with different content of hexagonal boron nitride and sisal fiber is shown in figure 5. CTE value decreases with addition of either of the fillers into epoxy matrix. The decrement is attributed to constrained mobility which arises in the molecules of epoxy due to adsorption of filler surfaces. The CTE of the composite reduces from $68 \times 10^{-6}/\text{°C}$ to $40.3 \times 10^{-6}/\text{°C}$ with 40 wt. % of hBN and 5 wt. % of sisal fiber. The decreasing trend is gradual with filler content. With increase in content of either of the filler, there is a transition from loosely bound polymer to tightly bound polymer because of the reduction in the distance between filler content. This increase the hindrance inside the multi-phase system and supresses the expansion of polymer-filler combination.

4. Conclusions
The effect of micro-size hBN and short sisal fiber content on mechanical and thermal behaviour of epoxy based hybrid composites is investigated. Mechanical property analysis of epoxy/hBN/sisal fiber composites revealed that tensile strength increased with hBN content when 10 wt. % of hBN is added.
However further increase in hBN content decreases the tensile strength of the composite. Again, when combination of sisal fiber and hBN is incorporated, it successfully enhances the tensile strength of epoxy matrix but only when the content of sisal fiber is limited. With increase in either of filler content, compressive strength of the composites increases. Compressive strength increases from 85 MPa for neat epoxy to 108.7 MPa for 40 wt. % hBN in epoxy. It further increases to 118.1 MPa when 5 wt. % sisal fiber were added in combination with hBN in epoxy matrix. Thermal property analysis reflects that with increase in content of either hBN or sisal fiber, coefficient of thermal expansion and glass transition temperature increases and the increment is a function of filler content. Also, for a particular combination of filler, even insulative sisal fiber in combination with hBN help to increase the thermal conduction behavior of the epoxy matrix.

5. References

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