Novel High Performance Copper Clad Laminates Based on Bismaleimide/Aluminium Borate Whisker Hybrid Matrix

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ABSTRACT: A novel method to prepare high performance copper clad laminates (CCLs) was developed by using bismaleimide/aluminum borate ($Al_{18}B_4O_{33}$) whisker hybrid as the matrix. The typical important properties such as thermal, mechanical, and dielectric properties of CCLs were investigated in detail. Results show that the incorporation of surface treated $Al_{18}B_4O_{33}$ whiskers with suitable concentration can noticeably increase the flexural strength and modulus, interlayer shear strength, the heat distortion temperature, as well as dielectric properties of the CCLs. Intensive studies show that the improved concrete strength of the matrix and that in the direction of thickness of CCLs are contributed to the noticeable improvement of mechanical properties of CCLs by using the hybrid matrix. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 103: 1325–1331, 2007

Key words: composites; whiskers; interfaces; hybrids; structure–property relation

INTRODUCTION

Copper clad laminates (CCLs) are the key base materials for electrical industries,[1] any product with integrate circuit boards must contain CCL, therefore, at present, almost all electric products need CCLs, from small products such as electric watches and calculators to big products such as computers and communication systems. Well, with the rapid worldwide development of electrical science and technology, high performance CCLs which can translate information by high frequency and high speed, designated as HPCCLs, have been gained increasing demands from the whole CCLs market, and the attentions of both scientists and engineering.[2–4] It is well known that the key properties including thermal, dielectric, and processing properties of a composite are mainly dependent on its matrix. Therefore, HPCCLs must be fabricated by using high performance matrix resins which have higher thermal stability, better dielectric properties (lower dielectric constant and lower dielectric loss), good processing characteristic, and acceptable price. That is, the traditional resins such as epoxy resins and phenol resins can not be used to produce HPCCLs. Some developed high performance matrices such as polyimide (PI),[5,6] polytetrafluouoethylene (PTFE),[7] and poly(phenylene ether) (PPE)[8,9] that have been developed to produce HPCCLs, but each of them has its additional disadvantages besides poor processing characterizations. For example, PTFE based CCLs have low glass transition temperature and stiffness as well as high price, in addition, they are easy to delaminate. PPE based CCLs have poor solvents resistance and stiffness. Therefore, developing high performance matrices have been the key to produce HPCCLs and also the key to meet the increasing demands by the worldwide CCLs market.

Bismaleimides (BMIs) are a leading class of thermosetting polyimides based on low molecular weight building blocks and terminated by reactive groups which undergo polymerization by thermal or catalytic means. They are of interest to the advanced composite industry for their stability at elevated tops and retention of properties in hot/wet environments as well as outstanding dielectric properties and flame retardancy. Because of their outstanding properties, some HPCCLs based on BMI resins have been developed. For example, BMI resin has been used to improve the properties of commercial FR-4 CCLs.[10] BT resin made up of BMI and cyanate ester is another classic example,[11] however, its price is somewhat higher, and its processing characteristics still need to be improved.

It is worthy to note that 4,4′-bismaleimidodiphenylmethane (BDM), based on 4,4′-diaminodiphenylmethane, is the most widely used building block of BMI family, because the precursor diamine is readily

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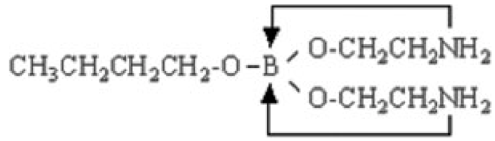
available at low cost. However, BDM has a relatively high melting point (~ 158°C) and a relatively low onset polymerization temperature (~ 160°C), therefore, it does not flow easily in the uncured state. Moreover, the cured product is extremely brittle because of its highly crosslinked structure. So all commercial resins based on BDM are modified BDM systems with improved processing characterization and toughness. Among them, a two-component high performance resin system based on BDM and o,o′-diallylbisphenol A (DBA), coded as BDM/DBA, has been proved to have good processing characteristics, outstanding thermal/mechanical properties, and toughness as well as good dielectric properties.[12–14] However, if the BDM/DBA system is directly used as the matrix for fabricating CCLs, its dielectric property still needs to be improved, and its property/price seems somewhat high. Therefore, how to find a novel way to further increase the integrate properties of BDM/DBA system and thus develop an effect method to prepare high performance matrix for HPCCLs is of great interest.

During the researching period, inorganic whisker/organic hybrid materials attract wide attention due to their benefits for merging the advantage of both inorganic and organic materials. Inorganic whiskers exhibit high stiffness and strength, and are almost free from internal flaws owing to their small diameter, and therefore their yield strength tends to approach the maximum theoretical value expected from the theory of elasticity.[15–18] Besides, inorganic whiskers also have superior thermal stability. There are more than 100 kinds of inorganic whiskers, but only about 10 kinds of them are commercial products. Generally, each kind has its advantage and disadvantage, so the property requirements for a concrete application should be considered when selecting the kinds of inorganic whiskers. Aluminum borate ($Al_{18}B_4O_{33}$) whiskers are a relatively new class of whiskers, and have attracted wide interest due to their attractively high property/price ratio.[19] Some valuable researches have been proved that appreciate amount of $Al_{18}B_4O_{33}$ whiskers with suitable surface treatment can greatly increase the mechanical and tribological properties of organic resins. Moreover, $Al_{18}B_4O_{33}$ whiskers with suitable content do not influence the processing characteristic of the original resin,[20] as stated by these authors their potential applications include tribological materials, biomaterials, and adhesives of advanced composites.

Therefore, there exists a great interest to invent a novel method to fabricate HPCCLs via designing inorganic whisker/organic hybrid resin as the matrix for CCLs, in detail, the CCLs based on BMI/$Al_{18}B_4O_{33}$ whisker hybrid matrix were prepared as models, and their integrate properties including thermal, mechanical, and dielectric properties were investigated to evaluate the possibility of developing HPCCLs via the method. Note that besides the reasons for selecting $Al_{18}B_4O_{33}$ whiskers to fabricate HPCCL as stated above, another important reason is that the boron atom in the whiskers tend to have good flame retard property,[21] which is an important and necessary property required by CCLs. To our best knowledges, no similar research has done before.



Scheme 1 Chemical structure of BE4.

EXPERIMENTAL

Materials

The 4,4′-bismaleimidodiphenyl methane (BDM) (mp = 156–158°C) was provided by the Northwestern Chemical Engineering Institute (China), o,o′-diallyl bisphenol A (DBA) was supplied by Northwestern Polytechnical University (China). $Al_{18}B_4O_{33}$ whiskers were obtained from Qihai Haixing Science and Technology Development (China), which was used as received, or surface treated by borate (designed as BE4, and its chemical structure is shown in Scheme 1) as described in previous literature,[22] the surface treated whiskers were coded as $Al_{18}B_4O_{33}$ (BE4) whiskers. The copper foil and the E-glass fiber cloth were supported by Ventec Copper Clad Laminates (China). Other solvents were all commercial products of industrial grade and used as received without any further purification.

Preparation of BMI/$Al_{18}B_4O_{33}$ hybrid resin

About 100 g of BDM and 60 g of DBA were placed in a flask equipped with a mechanical stirrer and thermometer. The mixture was heated to 110–130°C for 20 min, and appreciate amount of $Al_{18}B_4O_{33}$ whiskers or $Al_{18}B_4O_{33}$ (BE4) whiskers were then added to the mixture with thorough stirring. The resultant mixture, coded as BDM/DBA/$Al_{18}B_4O_{33}$ hybrid resin, was then used to prepare glue per procedure as described below.

For the property investigation of cured matrix, the obtained BDM/DBA/$Al_{18}B_4O_{33}$ hybrid resin was degassed in a vacuum oven at 120°C for 15 min and cast into a glass mold for curing and post curing per following procedures:

Curing: 150°C/2 h + 170°C/2 h + 190°C/2 h + 210°C/2 h.

Postcuring: 230°C/8 h.

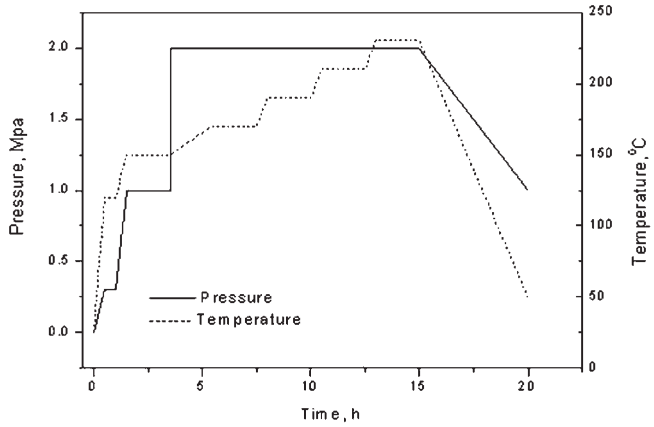


Figure 1 Preparation cycle for CCLs.

Preparation of prepregs and CCLs

The obtained BDM/DBA/$Al_{18}B_4O_{33}$ hybrid resin was added to acetone followed by thoroughly stirring to form a 35–45 wt % resin solution, namely as glue. The glue was poured into a clean plastic-cell to form a resin bath, E-glass woven fabric was dipped into the resin bath for 3–5 min followed by air-dried at 100°C for 10 min to form a prepreg (Half-cured sheet).

Prepregs were cut into square plies (100 × 100 cm^2). Eight plies covered with a piece of copper foil on each side were stacked or ''laid-up'' to yield a laminate of the required dimension. The entire lay-up includes the release film, copper foil, and prepreg plies. After the laminate lay-up was completed, it was cured per the procedure as shown in Figure 1. After that the cured laminate was demolded and postcured in an air oven at 230°C for 8 h.

Measurements

The flexural properties and interlaminar shear strength (ILSS) were tested according to GB/T3357-82 and GB/T4722-92, respectively. The heat distortion temperature (HDT) was measured according to GB 1634-79.

Dynamic mechanical analysis (DMA) was performed at a heating rate of 5 K/min using a DuPont 983 dynamic mechanical analyzer and with a constant strain of 20 μm. A single cantilever clamping geometry 17.5 mm and rectangular samples were tested at a fixed frequency of 1 Hz between 30 and 300°C. The glass transition temperature (T_g) is defined as the peak temperature of tan δ-temperature plot.

Hitachi *S*-570 scanning electron microscope (Japan) was used to observe the fracture morphology and interfacial adhesion of the samples. Cross sections of the samples were obtained by fracturing the sample during flexural testing.

Dielectric properties of CCLs were measured according to GB/T 4722-92.

RESULTS AND DISCUSSION

Mechanical properties of CCLs

It is known that ILSS of a composite is mainly depended on the strength of the matrix used and the interfacial adhesion between the matrix and reinforcements,[22] while the flexural property of a composite reflects the integrate properties of the composite due to its special stress loading and damage mechanic.[23] Therefore, ILSS and flexural properties have been selected as the typical properties to evaluate the integrate mechanical properties of a composite.

Figure 2 shows the dependence of the flexural strength on the $Al_{18}B_4O_{33}$ (BE4) whisker loading. It can be seen that the flexural strength increases greatly with the small incorporation of $Al_{18}B_4O_{33}$ (BE4) whiskers, and when the $Al_{18}B_4O_{33}$ whiskers loading is about 7.5 phr, the CCL has the maximum flexural strength (625 MPa), which is about 122% times the original value (512 MPa). Note that although there exists an optimum whisker content, the flexural strengths of all CCLs containing whiskers are much higher than that of the CCL without the whiskers, even when the whisker content is 15 phr. In detail, when the whisker content is higher than 7.5 phr, the flexural strength decreases, and then nearly levels off at 15 phr. When the whisker content is 15 phr, the flexural strength of the composites is 558 MPa, which is about 1.09% times that of the CCL without whiskers.

As shown in Figure 3, the dependence of ILSS on the $Al_{18}B_4O_{33}$ whisker concentration has the similar trend to that of flexural strength, moreover, when the $Al_{18}B_4O_{33}$ whisker loading is about 7.5 phr, the ILSS value reaches the maximum (60.3 MPa), which

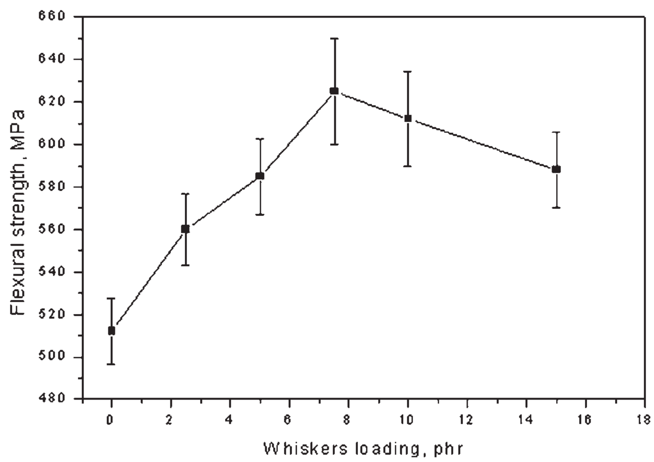


Figure 2 Flexural strength versus $Al_{18}B_4O_{33}$ (BE4) whiskers content for CCLs.

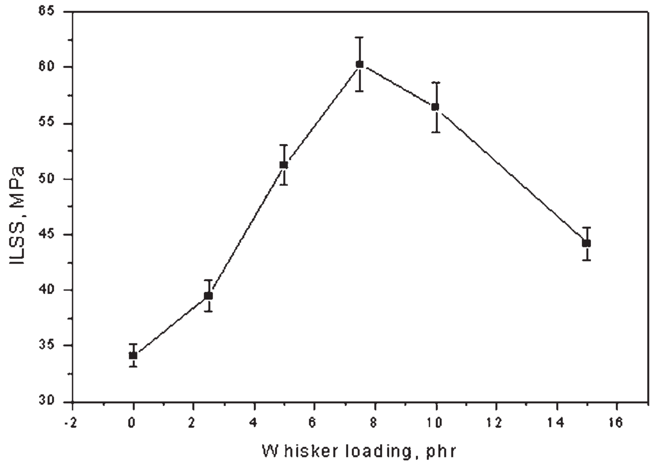


Figure 3 ILSS vs. $Al_{18}B_4O_{33}$ (BE4) whiskers content for CCLs.

is about 1.77 times of the original value (34.1 MPa). When the $Al_{18}B_4O_{33}$ whisker loading is greater than 7.5 phr, the ILSS value decreases. While when the $Al_{18}B_4O_{33}$ whisker loading is 15 phr, the ILSS value is 44.2 MPa which is about 1.30 times of the original value.

To explain the above results, the comparing study on the flexural strengths of two hybrid matrices and BDM/DBA were done. As shown in Figure 4, it can be seen that the incorporation of $Al_{18}B_4O_{33}$ (BE4) whiskers into BDM/DBA resin can greatly increase the flexural strength of BDM/DBA, and when the $Al_{18}B_4O_{33}$ (BE4) whisker loading is about 7.5 phr, the hybrid matrix has the maximum flexural strength, which is about 120% times of BDM/DBA resin. Similar results were also obtained in previous literatures.[24] Comparing two curves in Figures 2 and 4, it can be seen that the increase degree of $Al_{18}B_4O_{33}$ (BE4) whiskers on CCLs is bigger than that on matrix. The answer behind these phenomena maybe found from the SEM image of CCLs as shown in Figure 5. It reveals that although most whiskers are freely dispersed in the CCLs, there are some $Al_{18}B_4O_{33}$ whiskers between fiber layers, and their another ends are still embedded in the matrix, suggesting that whiskers and matrix have good adhesion, and more importantly, these whiskers act as ''nails'' hammered between fiber layers in the direction of thickness of CCLs. In other words, these whiskers change the CCLs from completely two-dimension to somewhat three-dimension, and thus improving the overall strength of CCLs. Conclusively, the improved concrete strength of the matrix and that in the direction of thickness of CCLs are contributed to the noticeable improvement of mechanical properties of CCLs by using the hybrid matrix.

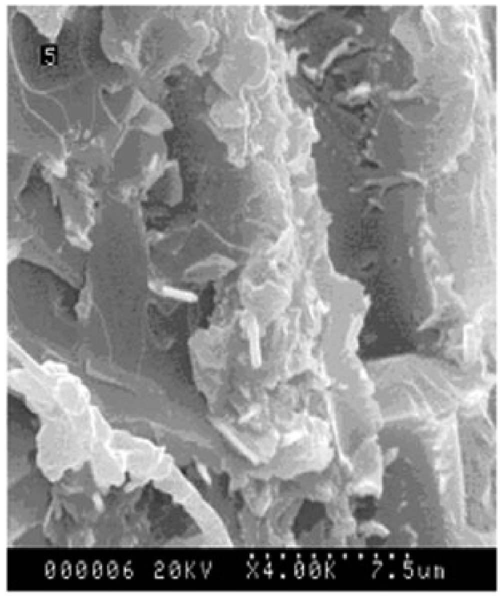


Figure 5 SEM images of CCLs with $Al_{18}B_4O_{33}$ (BE4) whiskers.

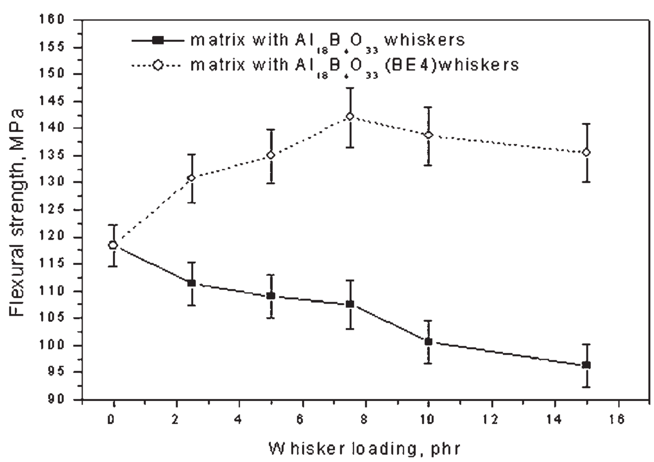


Figure 4 Flexural strength of BDM/DBA/$Al_{18}B_4O_{33}$ and BDM/DBA/$Al_{18}B_4O_{33}$ (BE4) hybrid matrix.

Thermal properties of CCLs

Heat distortion temperature (HDT) denotes the maximum temperature at which a polymer can be used as a rigid material, which maybe also considered as the upper temperature limit at which the material can support a load for any appreciable time. Thus, HDT value is a very practical and important property of a polymer.[25] Figure 6 gives the relationship of HDT of CCLs on the $Al_{18}B_4O_{33}$ (BE4) whisker loading, and the corresponding data of CCLs based on original $Al_{18}B_4O_{33}$ whiskers were also measured for comparison. It can be seen that the incorporation of $Al_{18}B_4O_{33}$ whiskers with or without surface treatment increases the HDT of CCLs, and higher the whisker loading, higher is the HDT value. In case of CCLs based on the matrix containing 15 phr

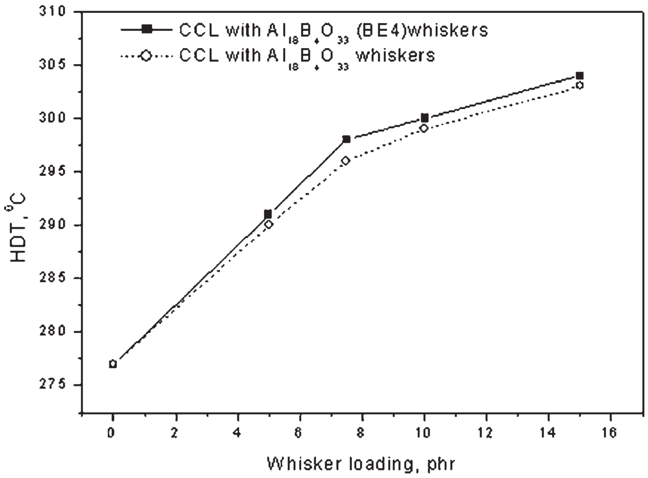


Figure 6 Relationship of HDT of CCLs on the whisker loading.

$Al_{18}B_4O_{33}$ (BE4) whiskers, its HDT value increases from 277 to 304°C.

It is known that fillers generally increase the HDT of a polymer, this increase is primarily due to the increase in modulus and the reduction of high creep rather than due to any large increase in the glass transition temperature.[25] To confirm whether it is true in present system, and to investigate the nature of the HDT increase of CCLs resulting from the incorporation of $Al_{18}B_4O_{33}$ whiskers, the flexural modulus of hybrid matrix and BDM/DBA resin, and glass transition temperature of different CCLs were measured. Figure 7 shows the dependence of the flexural modulus of the matrix on the $Al_{18}B_4O_{33}$ whisker loading. It is not surprising to find that the flexural modulus of BDM/DBA resin lineally increases greatly with the incorporation of $Al_{18}B_4O_{33}$ whiskers, similar results were also obtained by other researches.[26,27]

The glass transition temperature of CCLs without $Al_{18}B_4O_{33}$ whiskers and those containing different $Al_{18}B_4O_{33}$ whiskers such as original $Al_{18}B_4O_{33}$ whiskers or $Al_{18}B_4O_{33}$ (BE4) whiskers, respectively, were measured by DMA method which can be taken as the maximum of the curve tan δ versus temperature, as shown in Figure 8. It can be seen that T_g values of the two kinds of CCLs containing 15 phr whiskers are slightly higher than that without $Al_{18}B_4O_{33}$ whiskers, and the CCL with $Al_{18}B_4O_{33}$ (BE4) whiskers has the maximum T_g value (320°C), which is about 3°C higher than that of CCL without $Al_{18}B_4O_{33}$ whiskers, suggesting that $Al_{18}B_4O_{33}$ whiskers with or without surface treatment almost cannot greatly improve the glass transfer temperature of CCLs. In other words, the increase of HDT by the addition of whiskers is primarily due to the increase in modulus and the reduction of high creep rather than due to any large increase in the glass transition temperature.

Although the three CCLs have slight difference in their T_g values, they have significant difference in the shape broadness and intensity of tan δ peaks. As shown in Figure 8, the CCL without $Al_{18}B_4O_{33}$ whiskers and that with $Al_{18}B_4O_{33}$ whiskers have similar shape broadness and intensity of tan δ peaks, but, surprisingly, the CCL with $Al_{18}B_4O_{33}$ (BE4) whiskers has smaller shape broadness and lower intensity of tan δ peaks than the formers. According to Matsuoka,[28] the greater the motion associated with the transition, the greater the intensity of tan δ peak, Figure 8 suggests that $Al_{18}B_4O_{33}$ (BE4) whiskers decrease the molecular motion due to their interaction with BDM/DBA resin and thus causes a decrease in tan δ peak intensity.

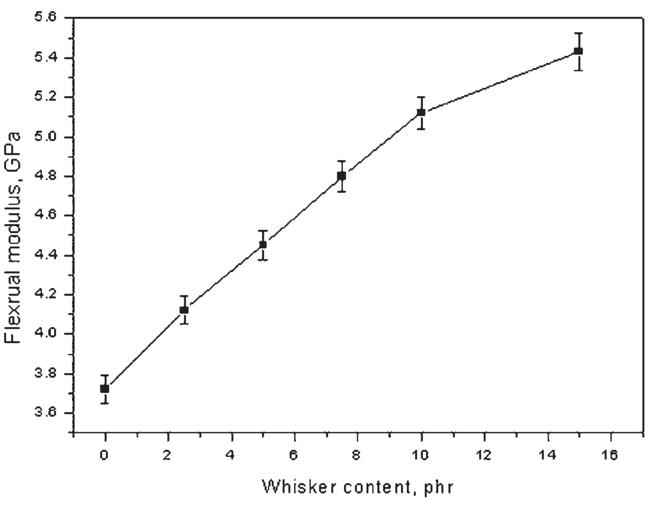


Figure 7 Flexural moduli of the BDM/DBA/$Al_{18}B_4O_{33}$ whisker hybrid matrices.

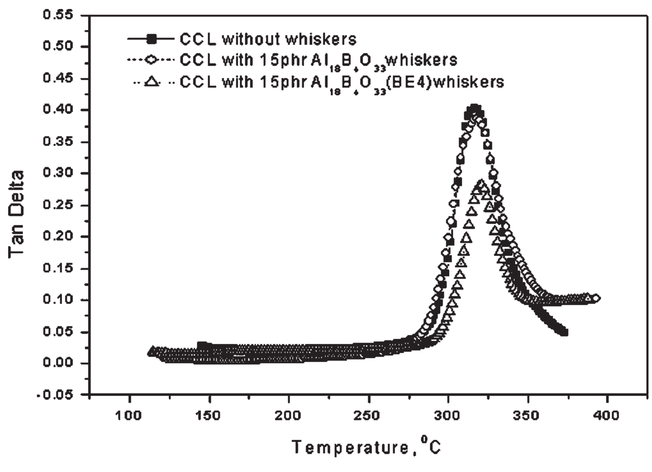


Figure 8 Tan δ-temperature curves from DMA tests of CCLs.

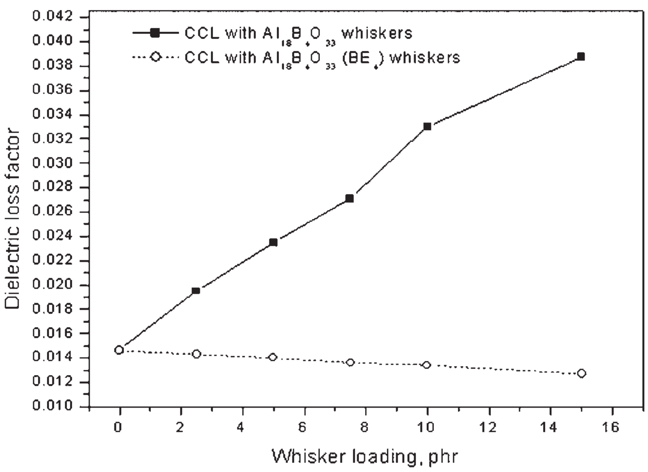


Figure 9 Dependence of dielectric loss factor of CCLs on the whisker loading.

The shape of tan δ peak may be used as a convenient indicator of the morphology state of the phases within multiphase materials.[29] The broadness of a relaxation indicates the complexity of the morphology of CCLs. Although only one damping peak is observed for all CCLs, the tan δ peak does tend to become strainer with the addition of $Al_{18}B_4O_{33}$ (BE4) whiskers but not with the incorporation of $Al_{18}B_4O_{33}$ whiskers without surface treatment, indicating that the hybrid matrix based on $Al_{18}B_4O_{33}$ whiskers and BDM/DBA has more complex morphology than that based on $Al_{18}B_4O_{33}$ (BE4) whiskers and BDM/DBA.

Dielectric properties

A necessary and important property of HPCCLs is their excellent dielectric properties including low dielectric constant (ε) and loss factor (tan δ). Comparing the dielectric loss factor of CCLs with whiskers and that without whiskers (Fig. 9), it can be seen that over the whole whiskers loading up to 15 phr, the CCL with nonsurface treated $Al_{18}B_4O_{33}$ whiskers shows the highest dielectric loss factor, while the CCL with $Al_{18}B_4O_{33}$ (BE4) whiskers possess the lowest dielectric loss factor, indicating that suitable surface treatment of whiskers is necessary to fabricate CCLs with improved dielectric properties.

It is known that dielectric loss factor of a reinforced polymer composite is determined by the tan δ of the matrix, reinforcement, and the interface polarization.[30] The surface treatment of whiskers leads to strong matrix/whiskers interface and whiskers/glass fibers interface, and thus reduces polarity of the CCLs. The strong matrix/whiskers interfaces are achieved through the chemical reactions between —NH_2 groups in BE4 molecular and —CH=CH— groups in the maleimide ring of BMI resin via Michael reaction, while the central boron atom in BE4 molecular leads to good physical absorption with $Al_{18}B_4O_{33}$ whiskers in addition of the reaction between the —OR groups in BE4 molecular and —OH groups in the surface of $Al_{18}B_4O_{33}$ whiskers. On the other hand, the strong whiskers/glass fibers interface are obtained through the —OR groups in BE4 molecular and —OH groups in the surface of glass fibers. These chemical reactions reduce the polarity groups in BDM/DBA phase and reinforcement phase. It is worthy noting that generally, when the interface is strong and has a low polarization, the influence of the interface on dielectric loss factor is negligible[30] (following discussion on dielectric constant will also confirm this point), but when the interface is weak, the influence of the interface on dielectric loss factor is not negligible, that is, in case of the CCL with $Al_{18}B_4O_{33}$ (BE4) whiskers and that with $Al_{18}B_4O_{33}$ whiskers, the interface polarization of the former is negligible but that of the latter is not negligible.

From above analyses, the dielectric loss factor of CCL with $Al_{18}B_4O_{33}$ (BE4) whiskers is therefore dominated by the ratio of glass fibers, BDM/DBA resin, and whiskers, and will decrease with increasing whisker loading because the whiskers have a much lower dielectric loss factor than that of BDM/DBA resin, while the dielectric loss factor of CCL with $Al_{18}B_4O_{33}$ whiskers polymer is dominated by not only the ratio of glass fibers, BDM/DBA resin, and whiskers, but also the interface polarization. Because of the above analyses, the overall polarization of the CCLs with $Al_{18}B_4O_{33}$ (BE4) whiskers are lower than that of CCL with $Al_{18}B_4O_{33}$ whiskers and CCL without $Al_{18}B_4O_{33}$ whiskers, therefore, the former has the lower dielectric loss factor than the latter.

The comparison of the data in Figure 10 shows that over the whole whisker loading range developed

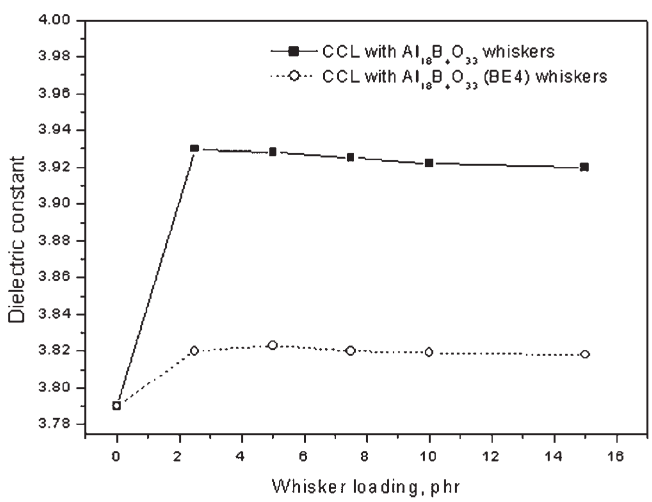


Figure 10 Dependence of dielectric constant of CCLs on the whisker loading.

in this study, the CCL without whiskers has the lowest dielectric constant, while the CCL with nonsurface treated $Al_{18}B_4O_{33}$ whiskers shows the highest dielectric constant, although which has higher dielectric constant than the CCL with $Al_{18}B_4O_{33}$ (BE4) whiskers. In addition, the dielectric constant of CCLs increases slightly with the whisker content up to 2.5 phr, then nearly levels off up to 15 phr whisker loading. It is known that the dielectric constant of a material is a function of its capacitance, which is proportional to the quantity of the charge stored on either surface of the sample; an applied electric field.[31] In case of a composite, its dielectric constant is dictated mainly by the polarity of matrix and those interfaces in subsurface (the area immediately next to the sample surfaces) of the sample. The small addition of whiskers, no matter surfaced treated or nontreated, increases the quantity of the accumulated charge because of an additional contribution from the polarization of matrix/whisker and whisker/fiber interfaces, although the additional contribution in CCL with surface treated whiskers is noticeably less than that in CCL with nonsurface treated whiskers and thus increases the dielectric constant. However, when the whisker content reaches a certain amount or even higher, the dielectric constant does not tend to further increase because the volume of the subsurface is certain, after the volume is fully occupied, the additional incorporated whiskers could not stay in the subsurface and thus can not have further effect on the dielectric constant of the material.

CONCLUSIONS

A system made up of BDM/DBA and $Al_{18}B_4O_{33}$ whiskers was chosen as the model to evaluate the method of fabricating CCLs by using polymer/inorganic whisker hybrid matrix, research results show that (1) inorganic whiskers should be surface treated to guarantee the good interfacial adhesion and thereby the improved mechanical, thermal, and dielectric properties of the resultant CCLs; (2) the concentration of whiskers has noticeable influence on the mechanical properties, dielectric properties, and HDT, but almost no effect on glass transition temperature. A 7.5 phr maybe the optimum content of $Al_{18}B_4O_{33}$ (BE4) whiskers in present study; (3) CCLs based on the polymer/whisker hybrid matrix exhibit an easy and potential way to produce high performance CCLs.

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