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Interface and its effect on the interlaminate shear strength of novel glass fiber/hyperbranched polysiloxane modified maleimide-triazine resin composites

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ABSTRACT

Interface is the key topic of developing advanced fiber reinforced polymeric composites. Novel advanced glass woven fabric (GF) reinforced composites, coded as GF/mBT, were prepared, of which the matrix resin was hyperbranched polysiloxane (HBPSi) modified maleimide-triazine (mBT) resin. The influence of the composition of the matrix on the interfacial nature of the GF/mBT composites were studied and compared with that of the composite based on GF and BT resin using contact angle, X-ray photoelectron spectroscopy (XPS), scanning electron microscope (SEM), and dielectric properties over wide frequency and temperature ranges. Results show that the interfacial nature of the composites is dependent on the chemistries of the matrices, mBT matrices have better interfacial adhesion with GF than BT resin owing to the formation of chemical and hydrogen bonds between mBT resin and GF; while in the case of mBT resins, the content of HBPSi also plays an important role on the interfacial feature and thus the macro-performance. Specifically, with increasing the content of HBPSi in the matrix, the interlaminate shear strength of corresponding composites significantly improves, demonstrating that better interfacial adhesion guarantees outstanding integrated properties of the resultant composites.

1. Introduction

Advanced fiber reinforced polymer composites (APCs) have attracted great interest of scientists and engineers since last century owing to their light weight, outstanding thermal resistance, high specific strength and modulus [1–3], and thus have been playing more and more important roles in modern industries including aerospace, aeronautics, electronic, transportation, etc. [4–6].

It is well known that the matrix is not only the basic component of a composite, but also the key factor for determining many important properties such as thermal resistance, dielectric property, and interlaminate shear strength as well as processing characteristics of a composite [7–9], so developing high performance matrices has been one key topic of APCs [10–13].

However, according to the principle of composites, whether the superior performance of the matrix can be fully exhibited in a composite is mainly dependent on the interfacial nature between the matrices and reinforcements [14–16]. In other words, it is necessary to intensively investigate the interface and its effect on the properties of the composites when developing any new matrix resin.

In this paper, a recently developed high performance resin system (coded as (mBT) maleimide-triazine) consisting of 4,4′-bismaleimidodiphenyl methane (BDM), *o,o*′-diallyl bisphenol A (DBA), 2,2′-bis (4-cyanatophenyl) isopropylidene (BADCy), and hyperbranched polysiloxane (HBPSi), was selected as the matrix resin owing to its attractive performances [17], which not only has outstanding integrated performance such as better dielectric, thermal and mechanical properties, but also meets the processing requirements of many techniques including traditional prepreg technique as well as Resin Transfer Molding. Therefore, the intensive investigation on the interfacial nature and its effect on the typical properties of composites based on mBT resin system are of great importance in both theory and actual applications. On the other hand, in order to discuss the interfacial nature induced by the composition of the matrix resin, the composite based on BT resin (a resin is made up of BDM, DBA, and BADCy) were also prepared for investigation.

2. Experimental

2.1. Materials

BDM was supplied by HuBei Fengguang Chemicals (China). DBA was purchased from Laiyu Chemical Factory (China). BADCy was the commercial product of Zhejiang Shengda Chemicals Co.,

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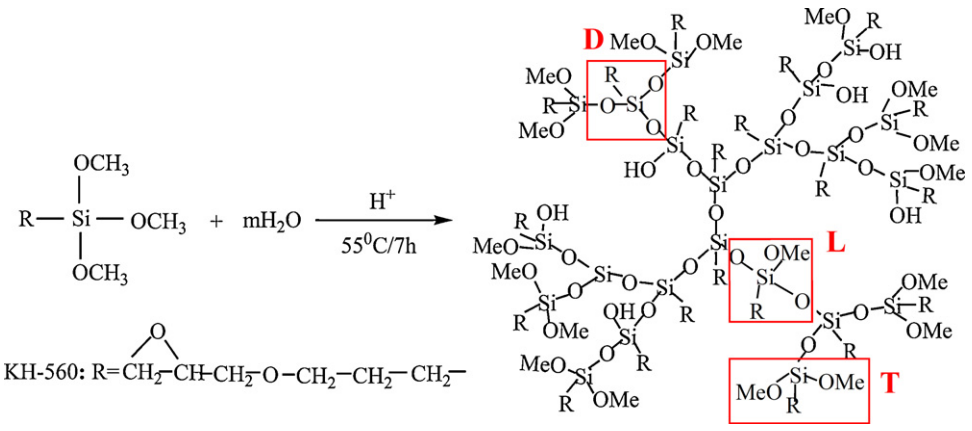

Scheme 1. The structure of hyperbranched polysiloxane.

Ltd. (China). E-glass woven fabric (MW-100) (ρ = 2.55 g/cm^3) was bought from Shaanxi Xingping Glass Fibers Factory (China).

HBPSi was synthesized via the hydrolysis of γ-glycidoxypropyltrimethoxysilane in our lab according to the method described in literature [17]. The structure of HBPSi is shown in Scheme 1. The ^{1}H NMR spectrum of HBPSi was recorded on a Bruker WM300 (Germany) with $CDCl_3$ as the solvent and internal standard, corresponding data are: 0.67 ppm (≡Si–C<u>H</u>$_2$–), 1.719 ppm (–CH$_2$–C<u>H</u>$_2$–CH$_2$–), 2.6 and 3.14 ppm (CH–O–C<u>H</u>$_2$–CH), 2.8 ppm (>C<u>H</u>–O–CH$_2$), 3.5 ppm (–O–C<u>H</u>$_3$, –O–C<u>H</u>$_2$–CH$_2$–), 3.75 ppm (–C<u>H</u>$_2$–O–CH$_2$–). The ^{29}Si NMR spectrum of HBPSi is shown in Fig. 1. HBPSi has three different chemical shifts at −67.99, −66.21, and −65.39 ppm, which can be assigned to dendritic (D), linear (L) and terminal (T) unit, respectively. The absolute molecular weight of HBPSi measured by multiangle laser light scattering (MALLS, DAWN HELEOS, USA) is 1500–2000, and the refractive index increment (dn/dc) value is 0.0772. Its atom ratios of Si/C and Si/O measured using energy dispersive X-ray spectroscopy (EDS, S-4700, Japan) are 0.14 and 0.30, respectively.

2.2. Preparation of BT and mBT resins

BDM and DBA with a weight ratio of 1:0.86 were put into a three-necked flask with a mechanical stirrer and a thermometer. The mixture was heated to 135 °C and maintained within that temperature with stirring until a clear and brown liquid was obtained. The mixture was maintained at that temperature for an additional 30 min to obtain a transparent liquid, which is BDM/DBA prepolymer.

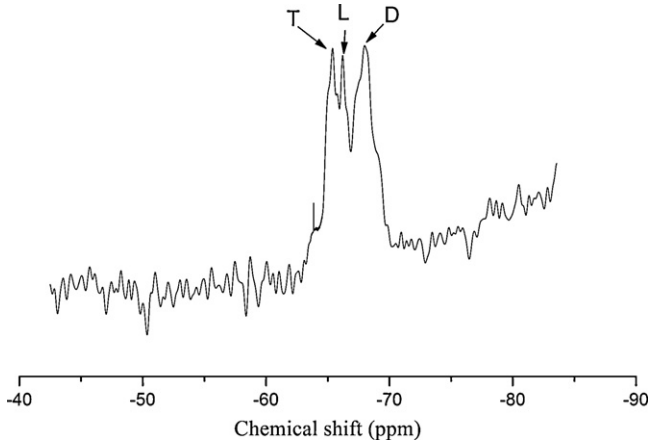

Fig. 1. The ^{29}Si NMR spectrum of HBPSi.

When the BDM/DBA prepolymer was cooled to 100 °C, preweighted BADCy (Table 1) was added into the flask with stirring for 30 min to obtain a brown transparent liquid, coded as BT resin.

For preparing modified BT resin, appropriate quantities of HBPSi, BADCy and BDM/DBA prepolymer according to the formulations listed in Table 1 were added into a flask with stirring for 30 min at 100 °C to obtain a brown transparent liquid. According to the weight ratio of components, the resultant resin is coded as mBT-1, mBT-2, or mBT-3, respectively.

The typical properties of mBT and BT resins are summarized in Table 2 [17].

2.3. Preparation of GF/mBT composites

BT resin was dissolved in acetone (the weight ratio of the resin vs. acetone was 1:1) followed by thoroughly stirring to form a transparent resin solution, named as glue. A sheet of glass fiber woven fabric was immersed in the glue, and then hung up for at least 24 h at room temperature to remove acetone, forming a prepreg. Ten prepregs were stacked one by one, and then put into a metallic mold for molding according to a programmed cycle of 150 °C/2 h + 180 °C/2 h + 200 °C/2 h under a pressure of 0.7 MPa. After that, the sample was postcured at 240 °C for 4 h in an air oven. The resultant composite is coded as GF/BT.

Other composites such as GF/mBT-1, GF/mBT-2, and GF/mBT-3 were also prepared by the same procedure mentioned above except that the matrix used were mBT-1, mBT-2, and mBT-3, respectively.

The fiber volume fraction of the composites was evaluated by an ablation method according to the Chinese Standard (GB2577-89). Results show that GF/BT and GF/mBT composites have similar volume fractions of fibers (59 ± 1.4 vol%).

2.4. Preparation of glass fiber samples for XPS tests

To detect the interfacial chemistry between glass fibers with a matrix resin (HBPSi, BT, or mBT-2), glass fibers were immersed into the resin/acetone solution, and then the solution was remained at 60 °C for 20 min (without the appearance of gelation). After that,

Table 1
The formulations of BT and mBT resins.

Resin	Weight (g)			
	BDM	DBA	BADCy	HBPSi
BT	100	86	25	0
mBT-1	100	86	25	10
mBT-2	100	86	25	20
mBT-3	100	86	25	30