Mechanical and dielectric properties at liquid nitrogen temperature of epoxy/AlN composites modified with different contents of flexible amine

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Received: 31 January 2020 / Accepted: 15 April 2020 / Published online: 27 April 2020
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Abstract

Epoxy resin has been wide applied in aerospace, superconducting industries because of the high electrical insulation, excellent adhesive properties and great chemical resistance. In this paper, epoxy/AlN composites were prepared by changing the content of the flexible amine (D-230) and the diethyl toluene diamine (DETDA) from 0 to 100 wt%. The effects of different ratios of D-230 and DETDA were investigated on the mechanical and dielectric properties of epoxy/AlN composites at liquid nitrogen temperature (77 K) and room temperature (RT). The properties of the samples were characterized by flourier transformed infrared (FTIR), scanning electron microscopy and broadband dielectric spectrometer. FTIR results showed that AlN particles were successfully modified by the silane coupling agent (KH-560). Tensile testing showed that the addition of flexible diamines generally improved the elongation at break at both RT and 77 K and the tensile strength at 77 K. The introduction of D-230 led to a lower dielectric constant at cryogenic temperature than that at RT.

Keywords Epoxy/AlN composites · Mechanical property · Dielectric property · Cryogenic temperature

1 Introduction

The development of superconducting cryogenic temperature technology has brought extensive application of composite materials at cryogenic temperature. In recent years, many resistant epoxy resins have been used as binders and impregnating materials for cryogenic temperature applications [1–3]. Therefore, the research on the performance of epoxy resin under ultra-low temperature is gradually attracting many attentions [4–8]. Figure 1 is the molecular formula of epoxy resin E-51, showing that epoxy resin molecules contain a large number of extremely active epoxy groups, ether bonds and hydroxyl groups [9]. They can produce chemical bond or adsorption

between molecules, and cross-link with curing agents, so epoxy resin has high bonding strength and good moisture resistance as adhesive.

In recent years, polymer composites have been widely used in the field of electronic devices, including central processing units, high-integrated memory chips, batteries, displays and electronic packaging materials for their excellent adhesive properties, good electrical insulation behaviors, low cost, high electrical insulating characteristics, great chemical resistance [10–14]. However, epoxy resins also have disadvantages such as brittleness, low toughness, and poor impact resistance, which limit the wide application. Incorporating heat-conducting insulating inorganic particles into epoxy resins can improve the

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Fig. 1 Molecular formula and properties of epoxy resin

thermal conductivity and preserve the electrical insulation properties [15, 16]. Various kinds of fillers, such as metals and metal oxides [17] aluminum nitride (AlN) [18–20], boron nitride [21], and silicon carbide [22] have been applied to prepare thermally conductive polymer composites. In this paper, AlN is used as fillers to prepare epoxy/AlN composites. AlN is a diamond-like nitride with high intrinsic thermal conductivity (319 W/mK) [23], high electrical resistivity ($> 10^{14} \Omega \text{ cm}$) [24], low thermal expansion coefficient (theoretical value: 4.4 ppm/°C) [25], and low dielectric constant (8.9 at 1 MHz) [24]. However, due to the high surface energy, AlN is extremely easy to agglomerate into large particles and could not be uniformly dispersed in the epoxy resin matrix. Functional modification of organic molecules on the surface of AlN particles provides a feasible way to improve their dispersion in epoxy groups. The modification with functionalized organic molecules not only improves the compatibility of AlN particles with the resin matrix, but also enhances the interfacial force between AlN particles and the matrix, thereby obtaining high-performance epoxy/AlN composites.

In comparison with the researches that have been carried out on the mechanical and dielectric properties of the epoxy/AlN composites. Yang et al. [26] investigated the mechanical properties of modified epoxy resins with flexible diamines at different temperatures from 77 K to RT. In their study, diethyl toluene diamine (DETDA) cured DGEBA epoxy resin has been modified by two flexible diamines (D-230 and D-400). The results show that the addition of flexible diamines generally improves the elongation at break and impact strength at both RT and 77 K. Chia-Ching et al. [27] investigated the dielectric properties of epoxy/AlN composites. The effects of the content of AlN filler on the physical and dielectric properties of epoxy/AlN composites are developed. They found that as the content of AlN powder in the epoxy/AlN composites increases from 5 to 40 wt%, the dielectric constant increases from 6.52 to 7.28 (measured at 1 MHz). The loss tangent of epoxy/AlN composites is slightly increased as the measured frequency increases. Zhu et al. [28] investigated dielectric

properties of immiscible LDPE/epoxy blend filled with hybrid filler consisting of HGM and nitride particle. In their study, they found that the dielectric properties of the HGM and nitride powders filled composites with an immiscible LDPE/epoxy blend matrix depend on the volume ratio of LDPE to epoxy. With increase in the volume ratio of LDPE to epoxy, the D_k continually decreases, and the D_f first decreases then increases. At a volume ratio of LDPE to epoxy of 3:7, the composite shows an optimum combination of dielectric properties.

In this work, a novel method for epoxy/AlN composites preparation was investigated, and the effects of different ratios of flexible amine (D-230) and diethyl toluene diamine (DETDA) were studied on the mechanical and dielectric properties of epoxy/AlN composites at liquid nitrogen temperature (77 K) and room temperature (RT). It was found that by changing the content of D-230, composites could be obtained with better mechanical and dielectric properties at 77 K than RT, which laid a foundation for the application of epoxy resin at cryogenic temperature.

2 Experimental

2.1 Materials

AlN with an average diameter of 500 nm was obtained from Hefei Kaier Nanometer Technology Development Co. Ltd (China). Bisphenol A epoxy resin (E-51) and the cycloaliphatic epoxy resin were purchased from Shandong Uself chemical technology Co. Ltd (China). The D-230 and the DETDA were used as hardener and purchased from Chengdu Yuanda Chemical Co. Ltd (China). The mass ratios of resin, hardener and AlN were kept at 10:3:2. The addition amount of AlN nanoparticles remained consistent across all samples.

2.2 Preparation of epoxy/AlN composites

Surface modification of AlN nanoparticles was performed by using a wet method according to previous report [29]. Briefly, AlN and KH-560 (mass ratio 1:20) were dispersed in acetone, magnetically stirred for 2 h, ultrasonically dispersed for 1 h, washed and placed in a vacuum oven at 80 °C for 24 h.

The epoxy/AlN composites were prepared by pre-heating E-51 and cycloaliphatic epoxy resin in an oven at 60 °C for 0.5 h to reduce the viscosity, followed by adding AlN particles under continuous stirring to obtain homogeneous mixtures. Then D-230 and DETDA in different ratios were mixed under magnetic stirring for 2 h, ultrasonic dispersion for 1 h and vacuum defoaming, followed by pouring into a mold for curing. Tensile specimens were prepared according to ASTM D638-96 standards. The dimensions of impact specimens are 4 mm × 10 mm × 80 mm with a 0.2 mm v-notch. The curing process was performed according to 80 °C /20 h and 130 °C /10 h, then the molds were left in the oven and cooled gradually to room temperature.

2.3 Characterization

The flourier transformed infrared (FTIR) spectrum of AlN particles was obtained using a Paragon 1000 (Perkin Elmer Inc., USA) with the scanning range from 4000 to 500 cm⁻¹.

The tensile properties were tested using a tensile testing machine (Reger Instruments Ltd., China) under tensile rates of 5 mm/min at RT and 2 mm/min at 77 K. The testing at 77 K was performed within seconds after the specimens were dipped in a liquid nitrogen filled cryostat for 20 min. The tensile strength was calculated based on Eq. (1):

$$\sigma = \frac{p}{b \times d} \quad (1)$$

where σ was the tensile strength (MPa), p was maximum load on specimen (N), b was sample width (mm), and d was sample thickness (mm), respectively.

The impact strength of the samples was measured with a REGER Impact Tester, and the test was according to ASTM D-256. The dimensions of the samples for impact test were 4 mm × 10 mm × 80 mm with a 0.2 mm v-notch. Impact test at 77 K was performed with the specimens dipped in a cryostat filled with liquid nitrogen for 20 min, and the test was completed in seconds after taking the specimens out of the cryostat. At least three specimens were tested for each composition. The tensile strength was calculated based on Eq. ((2)):

$$A_k = \frac{E_c}{b \times d} \times 10^3 \quad (2)$$

where A_k was the impact strength (KJ/m²), E_c was fracture absorption energy (J), b was sample width (mm), and d was sample thickness (mm), respectively.

A field emission scanning electron microscope (FE-SEM, JSM-7800F, Japan) was used to observe the tensile fracture section of epoxy/AlN composites. For SEM observation, the fractured surface was sputtered with thin layers of gold to avoid the accumulation of charge.

The dielectric properties were tested on a broadband dielectric spectrometer (Novocontrol Concept 80, Germany) with a test frequency range of 10¹ – 10⁷ Hz. The sample was placed in the middle of two circular electrodes with a diameter of 10 mm, and a voltage of 1 V was applied to both surfaces of the sample. Before the test, the samples were divided into two groups. One group was the control group. The two sides of the samples were uniformly coated with a layer of silver paste, then placed in an vacuum oven for 12 h. Another group was the experimental group. First, the samples were placed in liquid nitrogen for 30 min. Then, the other steps were same as the control group. The control group was named as the dielectric constant of the epoxy/AlN composites with different D-230 content at RT. The experimental sample was named as the dielectric constant of the epoxy/AlN composites with different D-230 content at 77 K. The dielectric properties of the samples after liquid nitrogen treatment were analyzed.

3 Results and discussion

3.1 Modification of AlN particles

The FTIR spectrum of as received AlN and KH-560 treated AlN are shown in Fig. 2. The FTIR spectra were substantially similar for AlN after modification by 5 wt% and 6 wt% KH-560. As shown in Fig. 2, the strong absorption at 3430 cm⁻¹ was attributed to hydroxyl group (–OH) stretching, and that at 1631 cm⁻¹ was attributed to –NH– group vibration. It was indicated that AlN particles had higher affinity and reactivity to moisture and water [30]. The peak at 740 cm⁻¹ was ascribed to the characteristic absorption peak of AlN. At the same time, new absorption peaks at 2921, 2854, and 1040 cm⁻¹ and those at 2921 and 2854 cm⁻¹ were characteristic of asymmetric and symmetric –CH₂ stretching vibrations, respectively. The peak at 1040 cm⁻¹ was contributed to Si–O stretching vibration. The changes in the infrared absorption spectra of the raw AlN particles and KH-560-treated AlN particles indicated that the surface of AlN particles was successfully grafted with silane coupling agents.

Figure 3 is the schematic representation of reaction among AlN, KH-560, and epoxy resins. After surface treatment of AlN by KH-560, there is better interfacial compatibility between treated AlN and resin matrix, which is beneficial for enhancing the interface adhesion strength of epoxy/AlN composites [31]. Furthermore, the surface

treatment of AlN can improve the uniform dispersion of AlN in resin matrix, which finally increases the mechanical properties of the composites.

Mechanical properties of epoxy/AlN composites.

Figure 4a shows the tensile strength of epoxy/AlN composites at RT and 77 K. The tensile strength under 77 K was

Fig. 2 **a** FT-IR spectra of as received AlN and **b** AlN after modification with 5 wt% and **c** 6 wt% KH-560 and molecular formula of silane coupling agent KH-560

Fig. 3 Schematic representation of reaction among AlN, KH-560, and E-51

Fig. 4 **a** Tensile strength and **b** impact strength of epoxy/AlN composites with different D-230 contents at RT and 77 K

higher than that at RT. When the D-230 content was 80 wt%, the tensile strength of the epoxy/AlN composites at 77 K was 83.2 MPa, which was 48.3% higher than that containing 80 wt% of D-230 at RT.

When the filling amount of D-230 was less than 20 wt%, the tensile strength of composites increased with the increase of D-230 content at RT. When the addition amount of D-230 was 20 wt%, the epoxy/AlN composites had the highest tensile strength at RT. As the D-230 content increased further, the tensile strength of the composites began to decrease. In general, the addition of soft matters like rubber and polyurethane to epoxy resins resulted in reductions of the tensile strength at RT [32]. The solidification of the epoxy/AlN composites at high temperatures produced internal stress, resulted from the bulk shrinkage from high temperature to room temperature during the cooling process. Due to the internal stress, micro-cracks and voids were generated inside the composites, which greatly affected the performance of the final product. Conversely, the introduction of soft segments like D-230 into brittle epoxy can significantly reduce the internal stress [26, 33]. When a proper amount of D-230 was added, the internal stress of the sample was reduced, and the crack resistance of the system was improved, so that the tensile strength was increased by adding an appropriate amount of D-230. On the other hand, as the content of the flexible diamine increased, the crosslink density of the system decreased, thereby lowering the cohesive strength, so the tensile strength decreased as the relative content of D-230 increased.

When the addition content of D-230 increased from 0 to 80 wt%, the tensile strength of the composites at 77 K

increased with increasing the D-230 content. When the addition amount was 80 wt%, the epoxy/AlN composites had the highest tensile strength at 77 K. The higher tensile strength of epoxy resin composites at 77 K was resulted from the increased cohesive strength caused by the cold shrinkage of the chemical bond and the network structure. The pure diethyl toluene diamine curing system frozen and became more brittle at 77 K, while D-230 was free to move at cryogenic temperatures due to the rotatability of the ether bond [26]. The spacing between molecular chains became smaller, and the molecular chain arrangement became denser. The anti-cracking ability of the system increased, so the tensile strength of the composites containing D-230 were larger than that of pure DETDA system at 77 K, and the contribution to the tensile strength increased with the increase of the content of the flexible component. When the D-230 content was 100 wt%, the tensile properties of the composites were slightly decreased.

Figure 4b shows the impact strength of epoxy/AlN composites at RT and 77 K. In general, the impact strength increased with increasing D-230 content at RT and 77 K. At RT, the increased impact strength can be attributed to the reduction of internal stress due to the stress relaxation by flexible molecular chains of flexible diamines D-230. At 77 K, ether linkages in D-230 could still be flexible via changing the main chain bond angles since they could undergo deformation at cryogenic temperature. Therefore, as the content of D-230 increased, the impact strength of the epoxy/AlN composites gradually increased at 77 K.

Figure 5 represents the curing network structures of unmodified and D-230 modified epoxy resins. There

Fig. 5 Schematic representation of curing network structures of D-230 modified epoxy resin system and molecular formula of DETDA and D-230

was more space between the D-230 modified epoxy/AIN composites, leading to a decrease in the network structure density. Therefore, after adding more D-230, there was more space between the composites, so the tensile strength tended to decrease. As shown in Fig. 5, the difference in chain length between the two curing agents would promote the mobility of the macromolecule to a certain extent. In other words, the DETDA and the D-230 had a better synergistic effect in improving the toughness of epoxy/AIN composites compared to pure DETDA. In addition, the tensile strength at 77 K was much higher than the tensile strength at RT, but the tensile strain at 77 K was much lower than that at RT, due to the uppressed macro-molecular mobility at 77 K.

SEM observation.

Figure 6 shows the morphology of the fractured surfaces for the epoxy/AIN composites used for tensile experiments. The fractured paths of river line patterns could be observed, indicating a brittle fracture. As shown in the Fig. 6b and d, most of the nanoparticles were embedded in the epoxy matrix, indicating good

compatibility and interfacial bonding between AIN and epoxy resin.

In the pure diethyl toluene diamine curing system, the crack tip of the tensile section at 77 K and RT continued to expand until fracture and without crack termination, and presented a typical brittle fracture morphology. The fracture morphology at 77 K was smoother than that at RT, indicating that the brittleness at 77 K was greater than that at RT. Meanwhile, surface-modified AIN nanoparticles could improve the interaction of two-phase interfaces. The particle size of AIN was small and uniformly dispersed in the resin matrix. In addition, there were many chemical bonds on the AIN surface, which could easily interact with the resin molecules to form stronger interfacial bonds. Thus, when subjected to impact, more silver streaks and plastic deformation would be produced to absorb energy.

Dielectric properties of epoxy/AIN composites.

Figure 7a and b show the frequency dependence of the dielectric constant of epoxy/AIN composites. The dielectric properties of composites after liquid nitrogen treatment were analyzed. The dielectric constant of the

Fig. 6 **a** SEM images ($\times 5000$) of epoxy/AIN composites without D-230 at RT and **b** 77 K and **c** composites containing 80 wt% D-230 at RT and **d** 77 K

Fig. 7 Frequency dependence of dielectric constant for epoxy/AlN composites containing different D-230 content at **a** RT and **b** 77 K

epoxy/AlN composites slightly decreased with increasing the frequency, and the introduction of D-230 led to a slight increase in the dielectric constant. With the introduction of D-230, the dielectric constant at 77 K was lower than that at RT.

When the polymer was filled with an inorganic filler, the dielectric constant generally increased because of the higher dielectric constant of fillers by nature than the polymer matrix and interfacial polarization [23]. In this paper, the content of D-230 in all samples was same, so the different dielectric constant of the composites was due to the interfacial polarization effect. The polarization appeared in heterogeneous media consisting of phases with different dielectric permittivity, attributed to the accumulation of charges at the interfaces [34]. The dielectric constant of epoxy/AlN composites was closely associated with the dipolar polarization. Here, D-230 was introduced to reduce the rigidity of the epoxy resin, which could promote the dipolar movement and enhanced the orientation polarization and thus resulting in higher dielectric constants. At low frequencies, all the free dipole groups in the epoxy molecular chains could orient themselves, resulting in higher dielectric constant at these frequencies. As the electric field frequency increased, the polarization failed to settle itself completely. The contributions of these dipole groups to the dielectric constant was continuously reduced, resulting in a decrease in the permittivity of the epoxy system at higher frequencies, so the dielectric constant of epoxy resin dropped approaching at high frequencies [35]. The composites had a lower dielectric constant at 77 K with the same content of D-230 because the dipolar

movement and the orientation polarization were limited at 77 K.

As shown in Fig. 8a and b, the dielectric loss tangent exhibited initially slight decreases with increasing the frequency, followed by an increase to a certain frequency of 10^7 Hz. The addition of D-230 caused a slight increase in dielectric loss tangent. The dielectric loss values of all samples were less than 0.04, indicating great dielectric properties.

Dielectric loss depended on the electrical conductivity in the epoxy composites. The electrical conductivity in turn depended on the number of charge carriers in the bulk of the material, the relaxation time of the charge carriers and the frequency of the applied electric field [35]. The increase in the value of dielectric loss tangent could be attributed to dipole polarization, since dipoles had less time to orient themselves in the direction of the alternating field with increasing frequency. So, the dipole polarization effects reduced when the frequency increased further, and the value of the dissipation factor declined accordingly. Furthermore, the decrease in dielectric loss at low frequency also can be ascribed to the dc conductivity. However, the dielectric loss tangent tended to increase further, which was an obvious relaxation loss process related to the epoxy resin. Because AlN particles were well dispersed in the matrix, the composites revealed a lower loss tangent. The dipolar polarization motion was frozen at 77 K, so the dielectric constant and the dielectric loss constant of the epoxy/AlN composites after liquid nitrogen treatment were reduced, indicating that the composites had better dielectric properties at cryogenic temperature, which laid

Fig. 8 Frequency dependence of dielectric loss constant for epoxy/AlN composites containing different D-230 content at **a** RT and **b** 77 K

a foundation for the application of epoxy resin in the field of cryogenic temperature and insulation.

4 Conclusions

The cryogenic tensile strength increased with the increasing flexible diamine content except the D-230 content of 100 wt%. When the D-230 content was 80 wt%, the tensile strength of the epoxy/AlN composites at 77 K was 83.2 MPa, which was 48.3% higher than that containing the same content of D-230 at RT. The impact strength increased with increasing D-230 content at RT and 77 K. The dielectric constant and dielectric loss of the epoxy/AlN composites increased slightly with increasing the D-230 contents, but it still remained at a low level. With the introduction of D-230, the dielectric constant of the composites at 77 K was lower than that at RT. The dielectric losses at both RT and 77 K were less than 0.04, indicating that the obtained composites had excellent dielectric properties. In conclusion, when the D-230 content was 80 wt%, epoxy/AlN composites could be obtained with better mechanical and dielectric properties at 77 K.

Acknowledgements The authors are grateful for the financial support of the National Key Research and Development Plan (Grant No. 2017YFE0301401), the Funds of Science and Technology Department of Sichuan Province (Grant Nos. 2017JY0057, 2018JY0003).

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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