

Preparation and Dynamic Mechanical Properties of Polyurethane-Modified Epoxy Composites Filled with Functionalized Fly Ash Particulates

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Received 19 July 2006; accepted 21 December 2006

DOI 10.1002/app.26146

Published online 10 April 2007 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: Fly ash (FA) is a porous ceramic, which has proved to have some favorable functions. In this article, we have prepared a series of fly ash filled composites, where the polyurethane-(PU) modified epoxy (EP) was acted as the matrix. With the purpose of characterizing the effect of the content of PU in the matrices and the surface-treatment (ST) of fly ash particulates on the dynamic mechanical properties of composites, Fourier transform infrared spectral analysis (FTIR), microstructure observation, impact property test as well as dynamic mechanical analysis (DMA) were systematically investigated. With FTIR test, it can be found that the chemical reactions occur, and new chemical structures are formed between EP and PU, and the functionalized groups existing on the surface of fly ash particulates can also be detected. In fractographs

observation of composites, it can be seen that the silane coupling agent improves the interfacial bonding conditions between fly ash particulates and the matrix. Impact property test result reveals that PU improves the toughness of EP. Dynamic mechanical analysis of the composites is carried out from -40 to 150°C using a tension-compression mode. Results show that the composites, whose matrices are with 10 and 20 wt % PU, may possess better dynamic mechanical properties as compared with the other composites. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 105: 1118–1126, 2007

Key words: dynamic mechanical properties; epoxy composites; fly ash; silane coupling agent; glass transition temperature

INTRODUCTION

In aerospace and many other lightweight structures, there are many vibrational inputs that can lead to resonance, so it is necessary that damping of vibrations has become an important requirement in the design of aerospace and lightweight structures. To control the resonance response, it is also necessary to understand the damping mechanisms in the materials, especially the contributions to damping in structural materials.¹ The development of materials for vibration and acoustic damping has been focused on metals and polymers in recent years,^{2,3} and most of these materials are functional materials rather than structural materials due to their high cost, high density, and limited acclimatization, and so on.³ Therefore, an advanced polymer composite has a possible way of avoiding some of these problems.^{4,5} Owing to the high cost of fiber-reinforced polymer composites that have high specific strength, high modulus and light weight, many researchers have paid great

attention to the inexpensive polymer/ceramics composites.⁶ Shukla et al.⁷ investigated the polymer/Cu-coating cenosphere fly ash composites, and it was noticed that such composites could be used in EMI-shielding fields and wave absorption. Malhotra et al.⁸ studied the effect of fly ash particles on the structural, mechanical, and frictional behaviors of brake composites, and noted that the strength went down as the concentration of fly ash increased from 0 to 20 vol %, but the case was reverse as the concentration was more than 20 vol %, and 30 vol % fly ash in the frictional composites had a deleterious effect on their frictional behavior. Valeria et al.⁹ investigated the hybrid composite of epoxy resin modified with carboxyl terminated butadiene acrylonitrile copolymer (CTBN) and fly ash microspheres. The result indicated that the impact strength of CTBN-modified epoxy resin was superior to that of pure epoxy, also the addition of fly ash to EP composite showed a negative effect on this property but a small amount may be considered. Thus, it can be seen that there were several researches on polymer/fly ash composites, but only few on the dynamic mechanical properties of such composites.

Epoxy resin, as a widely applied thermosetting macromolecular material, is extensively used in

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industry because of its excellent heat, moisture, and chemical resistance as well as good dynamic mechanical property.^{5,10} It, however, cannot meet the requirements especially for the applications in structural encapsulations because of its severe brittleness and high coefficients value of thermal expansion (CTEs) compared with ceramic particles. Thus, thermoplastic PU was employed to modify epoxy resin to improve the toughness. In the previous work, Xiao and Gao¹¹ studied the thermoplastic polyurethane/epoxy resin blends, which possessed excellent elasticity, wear resistance, and toughness. Ho and Wang¹² studied the applications of thermoplastic polyurethane-modified epoxy resins in electronic encapsulation, and results indicated that PU-modified ERs improved the toughness and reduced the stress of cured ERs. Wei et al.¹³ investigated liquid crystalline polyurethane as the toughening agent to modify epoxy. The results showed that not only the impact strength of modified epoxy was enhanced too much, but also the mechanical properties, such as tensile strength and tensile modulus, were also improved. Furthermore, the dynamic mechanical properties of modified epoxy were also improved.

Fly ash as an inorganic rigid material, which has a rather poor wettability with the polymer materials, it must be surface-treated by a coupling agent when it was filled in the polymer. Demjen et al.¹⁴ found that silane with different functional groups had different effects on the mechanical properties of PP/CaCO₃ composites. The amount of silane coupling agent creating a monolayer coverage changed between 0.3 and 1.0 wt % as calculated for the CaCO₃. Zhang et al.¹⁵ studied the effect of silane coupling agent on the hollow glass spheres and resin. The results showed that the strength and modulus of filled system were both enhanced with a treatment by silane coupling agent. Suzuki et al.¹⁶ studied the relationship between the interphase consisting of physisorbed and chemisorbed silane on glass fibers and the resultant composite Mode I delamination fracture toughness in glass fiber fabric laminate. By total carbon analysis, it was found that the physisorbed silane migrated into the resin matrix and influenced the mechanical properties and the interlaminated fracture of laminated specimen.

In this article, a series of polyurethane-modified epoxy composites filled with functionalized fly ash were introduced. The purpose of this study is to investigate the effect of the content of PU in the matrices on the dynamic mechanical properties of composites, especially on the loss factors and glass transition temperature regions. In addition, it is also introduced how the surface-treatment of fly ash particulates effect the interfacial bonding behaviors and dynamic mechanical properties of the composites.

EXPERIMENTAL

Materials

Epoxy resin (A-component: E-51; B-component: solidified agent, catalyst, accelerator, etc) was employed as the matrix, and polyurethane (A-component: polyethylene glycol; B-component: Toluene-2, 4-diisocyanate, and polyethylene glycol, catalyst, accelerant) was employed as the flexibilizer and a part of the matrix.

Ethyl acetate and epoxy propane butyl ether were used as the diluting agent, respectively. Moreover, the fly ash particulates were applied as the filler and were surface-treated by the γ -aminopropyl triethoxy silane, which acted as a coupling agent.

Fly ash samples

The fly ash originated from coals typical of those burned in electric power plants. Their properties, which includes mineral transformation during combustion, have been studied extensively.¹⁷ The phase compositions and contents of chemical element in fly ash have been obtained with the help of the X-ray diffraction (XRD) and the X-ray fluorescence (XRF) equipments, as depicted in Figure 1 and Table I. From the Figure 1, it can be known that the phase compositions of as-received fly ash are α -SiO₂ and 3Al₂O₃·2SiO₂.

Surface-treatment of fly ash

The fly ash particulates were functionalized stoichiometrically by coating the surface with silanol groups, where the weight ratio of γ -aminopropyl triethoxy silane to fly ash particulates was controlled to 2 wt %, and the dry temperatures and time were 50°C for 0.5 h and 100°C for 3 h, respectively.

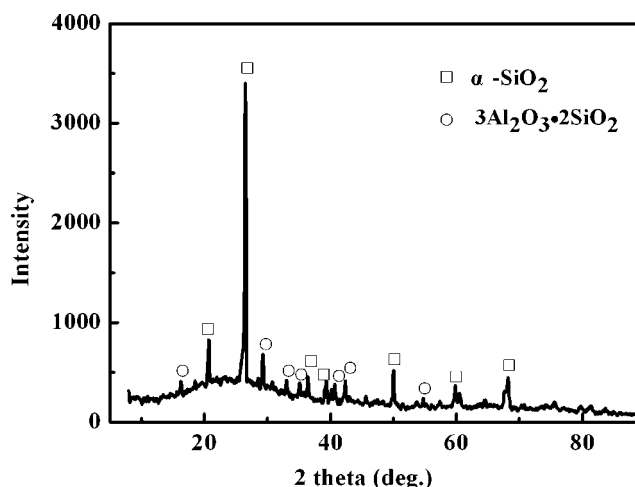


Figure 1 Phase compositions of fly ash particulates.

TABLE I
The Chemical Composition (wt %) of Fly Ash Particulate Sample

Elements	Content (wt %)
O	45.32
Si	29.38
Al	13.82
Fe	5.27
Ca	0.52
Mg	0.95
K	2.45
Ti	0.56
Na	0.78
Mn	0.42
Other	0.53

Composites preparation

Predetermined proportion (30 vol %) of fly ash particulates, kept under a dry condition, were added into a series of EP/PU blends matrices, and distributed into the matrices with agitating at 60°C, and then dispersed with ultrasonic instruments. After mixing homogeneously, the mixtures were degassed at 60°C until most of entrapped bubbles were removed completely. Finally, the mixtures were cured in a preheated mold at 60°C for 5 h and then postcured at 80°C for about 10 h.

Characterizations

To identify chemical reaction course of toughening of epoxy resin and the surface functional groups of fly ash particulates with treatment, spectroscopic analysis method was applied. Infrared spectrum analysis was performed on a Perkin-Elmer Spectrum one series FTIR spectrometer with a 4 cm⁻¹ resolution. The materials were mixed with dried powdery KBr, pelletized under pressure and then scanned. The scanning speed was 0.2 cm s⁻¹, and the samples were measured in the range of 4000–370 cm⁻¹ as KBr pellets.

A scanning electron microscope (SEM, Hitachi S-4700) was used at an accelerating voltage of 15 kV to examine the morphology of composites. Samples were notched by razor blade and then fractured in the liquid nitrogen. Because of the poor conductivity of polymer, the fractured surfaces were coated with a thin layer of gold. This coating was carried out by placing the samples in a high-vacuum evaporator, sputtering the gold metal held in the heated tungsten basket.

Impact toughness was measured with a drop hammer impact test machine (Instron 9250HV, USA). The falling velocity of the drop hammer was 1 m/s, and the mass of the drop hammer was 4.2 kg. The dimensions (length, width, and thickness) of specimen were 55, 5, and 4 mm, respectively.

The glass transition temperature (T_g) and dynamic mechanical property were measured by a Dynamic

Mechanical Thermal Analyzer, the Mak-04Viscoanalyser (France). The measurements were performed in a temperature range from -40 to 150°C with a heating rate of 3°C min⁻¹ at a fixed frequency of 30 Hz, in an argon atmosphere. A tension-compression mode was chosen and the dimensions (length, width and thickness) of specimen were 30, 5, and 1.5 mm, respectively. Figure 2 is the sketch map of Mak-04Viscoanalyser.¹⁸

RESULTS AND DISCUSSION

FTIR test

Toughening of epoxy resin

Figure 3 shows the FTIR spectra for the reaction course of epoxy resin and polyurethane. In spectrum (a), the band lying in 916 cm⁻¹ is the characteristic peak of epoxide group ($\begin{matrix} \text{---CH} & \text{---CH}_2\text{---} \\ & \diagdown \quad / \\ & \text{O} \end{matrix}$), and the band lying in 2278 cm⁻¹ is the characteristic peak of isocyanic group (---NCO---), which reveals that epoxy and Toluene-2, 4-diisocyanate both maintain their own structure characteristics.

In spectra (b) and (c), with the process of curing, the peak of isocyanic group (---NCO---) lying in 2278 cm⁻¹ is vanished, which indicates that a reaction occurs between the isocyanic group (---NCO---) and the hydroxyl group (---OH), thus creating the carbamate group (---NHCOO---).¹⁹ Also the peak of epoxide group lying in 916 cm⁻¹ disappears, and a new absorbing peak is generated in 1386 cm⁻¹, which illuminates that a reaction also existed between the isocyanic

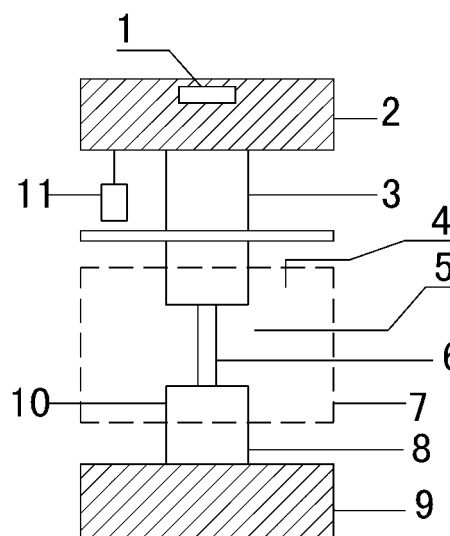


Figure 2 Sketch map of Mak-04Viscoanalyser: 1, displacement indicator; 2, lift platform; 3, vibration generator; 4, furnace temperature probe; 5, sample temperature probe; 6, sample; 7, furnace; 8, force sensor; 9, fixed platform; 10, sample clamp; 11, displacement sensor.

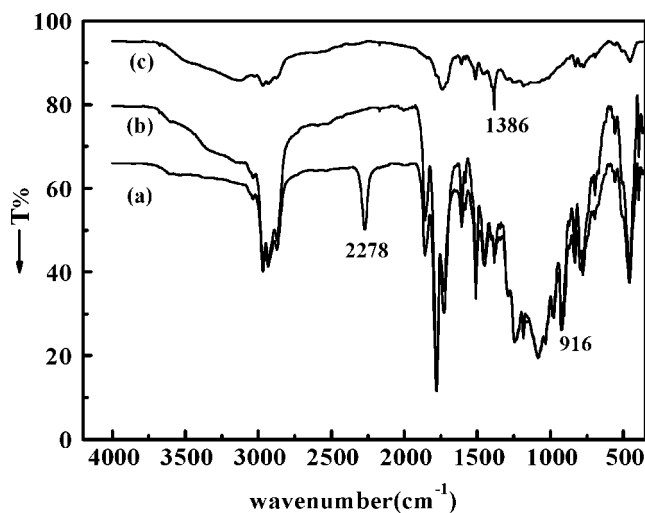
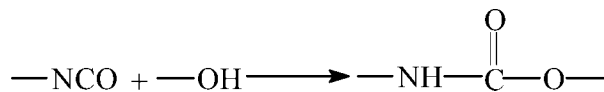


Figure 3 FTIR spectra for curing process (a) before curing; (b) after curing 5 h at 60°C; (c) after curing completely.

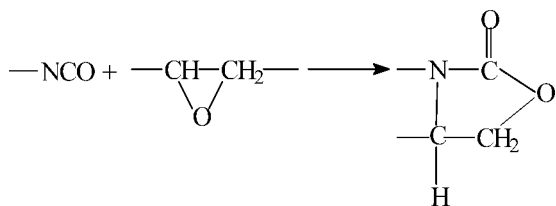
group ($-\text{NCO}-$) and the epoxide group. These results show that the isocyanic group ($-\text{NCO}-$) is an awfully active group, which is easy to react with the active hydrogen and the epoxide group.

According to the analysis above, we predict several potential chemical reactions:

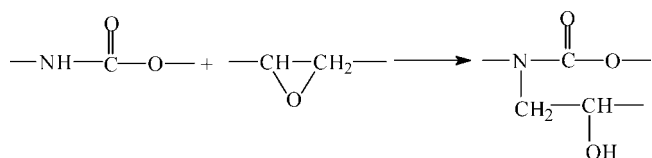
1. Reaction between the isocyanic group ($-\text{NCO}-$) and the hydroxyl group ($-\text{OH}$).²⁰



2. Reaction between the isocyanic group ($-\text{NCO}-$) and the epoxide group ($-\overset{\text{O}}{\text{C}}\text{H}-\text{CH}_2-$).²¹

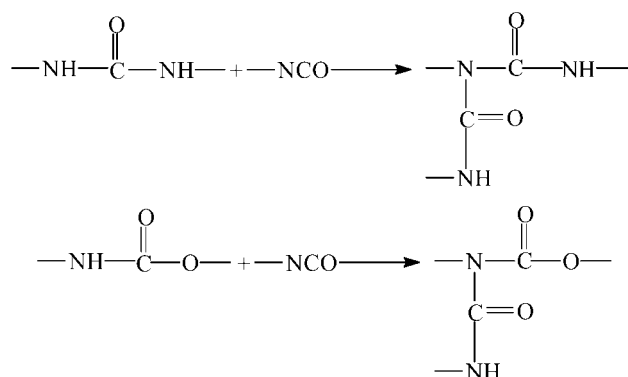


3. Reaction between the active hydrogen in carbamate group ($-\text{NHCOO}-$) and the epoxide group ($-\overset{\text{O}}{\text{C}}\text{H}-\text{CH}_2-$).²²



4. Reactions between the active hydrogen in carbamate group ($-\text{NHCOO}-$), urea group

($-\text{NHCONH}-$) and the isocyanic group ($-\text{NCO}-$).²²



Through the earlier discussion, it can be concluded that the chemical reactions occur, and new chemical structures are formed between EP and PU, which may be in favor of improving the compatibility of these two phases and toughening of the epoxy resin further.^{21,22} Moreover, the carbamate group ($-\text{NHCOO}-$) and the hydroxyl group ($-\text{OH}$) of epoxide group can generate a spot of hydrogen bonds, which can also effectively improve the systemic compatibility.

Surface-treatment of fly ash particulates

Figure 4 presents the FTIR spectra for γ -aminopropyl triethoxy silane, fly ash particulates without and with surface-treatment. As shown in spectrum (a), the curve denotes the characteristic peak of γ -aminopropyl triethoxy silane. The band lying in around

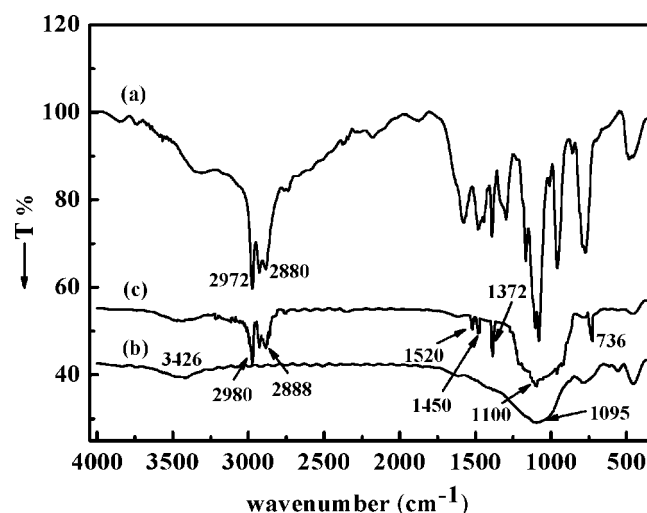


Figure 4 FTIR spectra for (a) γ -aminopropyl triethoxy silane; (b) fly ash without surface-treatment; (c) fly ash with surface-treatment.

2978 cm^{-1} represents a vibration absorbing peak of C—H bond that belongs to $-\text{CH}_3$. From spectrum (b), it can be seen that the fly ash particulates without surface-treatment have a very intensive band related to the vibration absorbing peak of the $-\text{Si}-\text{O}-\text{Si}-$ group at 1095 cm^{-1} ,^{5,23} and the absorbing peak lying in 3426 cm^{-1} is caused by the flexible vibration of hydroxyl group ($-\text{OH}$).⁵

In spectrum (c), there are a few new absorbing peaks of fly ash particulates with surface-treatment occurring in 2980, 2888, 1520, 1450, 1372, and 736 cm^{-1} , but these peaks are absent in spectrum (b), which suggest that these peaks correspond to characteristic peaks of γ -aminopropyl triethoxy silane. Herein, the peak lying between 2888 and 2980 cm^{-1} represents the characteristic peak of C—H bond. Additionally, the peak lying in 3426 cm^{-1} is weakened, which indicates that the amount of hydroxyl group ($-\text{OH}$) reduces, and the reactions must occur between the γ -aminopropyl triethoxy silane and hydroxyl group on the surface of fly ash particulates.

The basic principle of surface-treatment with the γ -aminopropyl triethoxy silane can be interpreted as follows: the γ -aminopropyl triethoxy silane^{24,25} with a general chemical structure $(\text{RO})_3\text{SiY}$, where $-\text{RO}$ is an alkoxy group and $-\text{Y}$ is an organic-functional group. During the surface-treatment of fly ash particulates with γ -aminopropyl triethoxy silane, the alkoxy group hydrolyzes in an aqueous environment, producing the hydroxyl group, one or more of which condense with the hydroxyl groups commonly found on the surfaces of fly ash particulates. Subsequent drying leads to a formation of both covalent bond linkages with the surfaces of fly ash particulates and development of a crosslinked silane film, which may be in favor of improving the resin-wettability. The idealistic view of a crosslinked silane film is depicted in Figure 5. Thus, it can be concluded that the effect of the silane coupling agent on the interfacial bonding conditions between fly ash particulates and the matrix is quite significant.

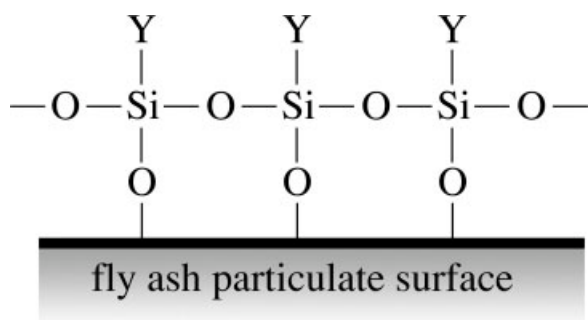


Figure 5 Idealistic view of the silane coupling-treated surface.

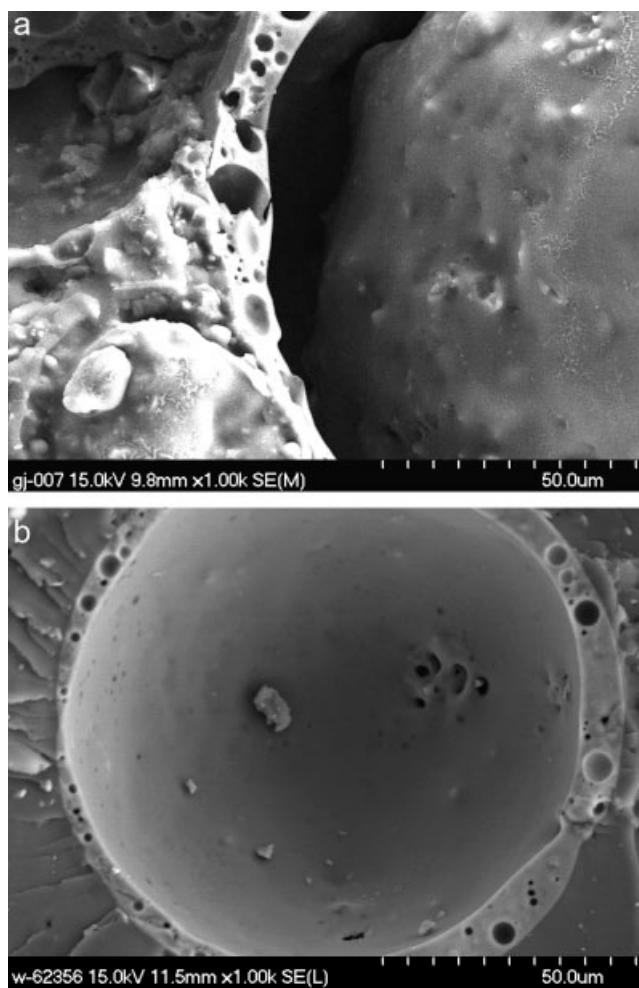


Figure 6 SEM micrographs of the fractured composites filled with: (a) fly ash without surface-treatment; (b) fly ash with surface-treatment.

Morphological investigation of composites

Fracture surfaces of the composite samples can show how the fly ash particulates are dispersed and distributed into the modified epoxy matrix. The SEM micrographs for the composites filled with fly ash without and with surface-treatment, are shown in Figure 6(a,b). From Figure 6(a,b), it can be seen clearly that there is an obvious interface desquamation between the matrix and fly ash particulates without surface-treatment, and the interfacial gap is about 5–8 μm . But the fly ash particulates, which were treated by the silane coupling agent, can combine well with the matrix and have no visible interface desquamation because of a favorable resin-wettability. These results indicate that the interfacial conditions of the latter are better than those of the former, which must be in favor of enhancing the dynamic mechanical properties of such composites.

Figure 7 illustrates SEM fractographs for the composite with 10 wt % PU. From Figure 7(a), it can be

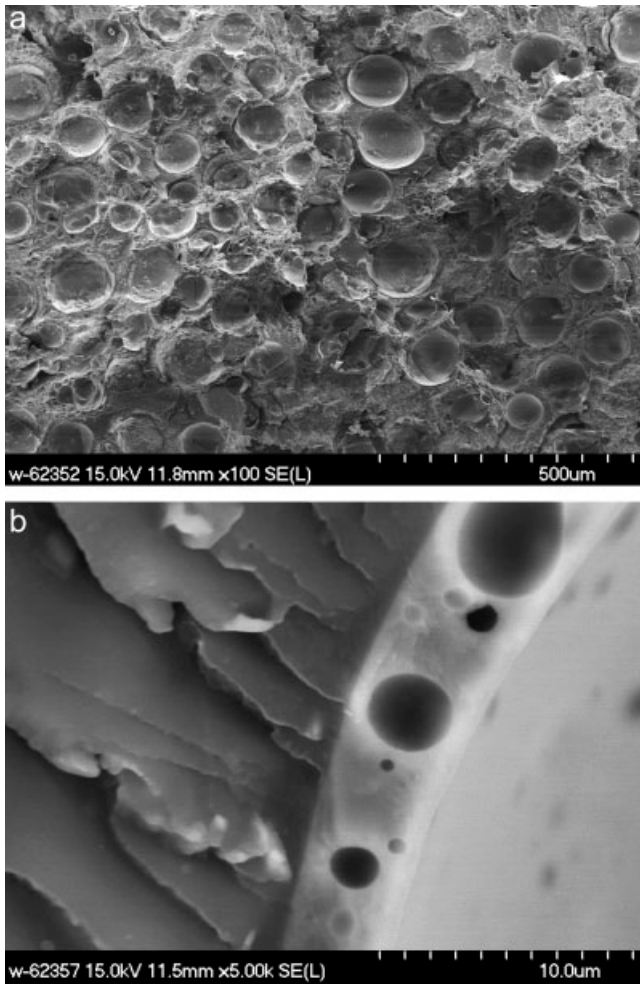


Figure 7 SEM fractographs of the composites filled with surface-treated fly ash: (a) particulates distribution; (b) interface.

seen that the fly ash particulates are distributed homogeneously into the matrix except a few high particulates rich regions. At a higher magnification observation of the interface [Fig. 7(b)], the interfacial bonding condition can be examined more clearly. A well interfacial bonding is observed between the matrix and fly ash particulates with surface-treatment. Moreover, the thickness of the spherical shells of fly ash particulates is about 3–5 μm , and there are some micropores in the spherical shells, which may result from the partial expansion of air in the spherical shells during the high-temperature combustion of coal. And in fact, these micropores may be beneficial to improve the dynamic mechanical property.

Impact strength of composites

To verify the function of PU toughening the EP, the impact toughness of composites with different content of PU in the matrices were measured. Figure 8 presents the impact strength varying with the PU

content. From the figure, it can be seen that the impact strength of pure EP is only 11.12 kJ/m^2 , and the impact strength is more than 11.12 kJ/m^2 after the addition of PU into EP. The impact strength enhances with the increase of PU content in the matrices, and the increasing degree of impact strength also enhances slightly with the increase of PU content. This result suggests that the toughness of EP has been improved through the modification of PU, which may be in favor of improving the dynamic mechanical properties of such composites. And in fact, these micropores may be beneficial to improve the dynamic mechanical property.

Dynamic mechanical properties of composites

DMA curves

Damping (loss factor) expresses an ability of converting the mechanical energy into the heat energy when material is subjected to an external loading. It is generally defined as eq. (1), and the eq. (2) is the expression of complex dynamic modulus (E^*):

$$\tan \delta = E''/E' \quad (1)$$

$$E^* = E' + iE'' \quad (2)$$

where δ is the phase angle between stress and strain, and E' and E'' are the elastic storage modulus and elastic loss modulus, respectively. As a result, $\tan \delta$ is an important parameter characterizing material's viscoelasticity.^{26,27}

Figure 9 shows the dependence of E' , E'' and loss factor, $\tan \delta$, for the composite with 10 wt % PU. In the temperature range from -40 to 0°C , the elastic storage modulus (E') drops sharply with a rise in temperature, but the elastic loss modulus (E'')

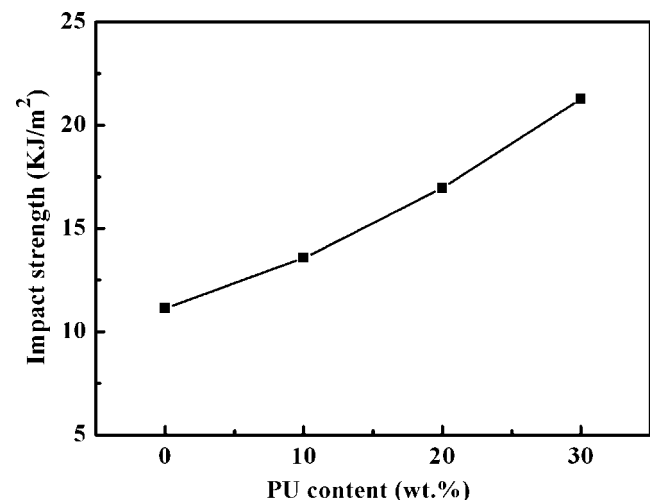


Figure 8 Effect of PU content on the impact strength of composites.

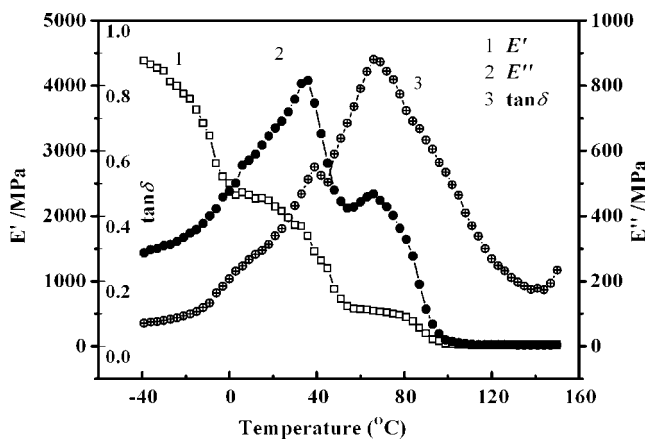


Figure 9 DMA curves of the composite, in which matrix with 10 wt % PU.

increases drastically. Within the temperature range from 0 to 100°C, E' of the composite also reduces sharply and reaches a small platform in the temperature range from 55 to 65°C with the increasing temperature. The curves of the temperature dependence of the E'' and $\tan \delta$ both reach a peak (the peak of the $\tan \delta$ curve is not obvious) around 40°C, and then decrease. This transition temperature is attributable to the noncrystalline phase in the matrix. Additionally, the curves of the temperature dependence of the E'' and $\tan \delta$ both show other two obvious peaks at 68°C, corresponding to the glass transition temperature (T_g) of the system, which is closely related to the T_g of the matrix. Furthermore, the values of the E'' and E' are prone to zero when the temperature increases over 100°C.

These may be interpreted as follows: In general, there are three states in the polymer materials (glass state, glass transition state, and high elastic state). The dynamic mechanical behavior has obvious discrepancy in the three different states. In the glass state, the movement of chain segment is mostly "frozen," so the material is prone to store more energy, and the energy dissipation is very little, leading to a high elastic modulus (E') and a low loss factor ($\tan \delta$). However, the movement of chain segment progresses enough and the elastic modulus reduces remarkably in the high elastic state. There is an obvious deformation when material is subjected to a minor stress, but the deformation can restore rapidly when the stress is removed. So the elastic storage modulus, elastic loss modulus, and loss factor are all very low in the high elastic state. Only in the glass transition state, the elastic modulus reduces and the loss factor reaches a peak value. Here, the temperature corresponding to the peak value of loss factor is the glass transition temperature (T_g) of material. Thus, it can be seen that the glass transition temperature region (damping temperature region) is one of

the important factors evaluating the dynamic mechanical property in our current work.

Effect of the content of PU on dynamic mechanical properties

The curves reconfirm that the entire properties of composites are greatly affected by the content of PU in the matrix and operating temperature. Figure 10 shows the curves of the temperature dependence of $\tan \delta$ for the composites investigated. From the Figure 10, we can see that the values of $\tan \delta$ increases first and then decreases with increasing temperature, which means that the increase of E' is slower than that of E'' when temperature is under T_g , but the case is reverse when temperature increases over T_g .

The constitution of different system is very complex, and is usually influenced by the component, polymeric conditions, and so on. Be similar to the graft, the entangled interpenetration may also produce phase separation. Because the value of mixed entropy (S) is low in the initial stage of interaction, but with the process of interaction, the increase of molecular chains results in the reduction of the mixed entropy ($\Delta S_m < 0$), and the mixed enthalpy (ΔH_m) also has a positive value. According to the equation of the Gibbs free energy (ΔG_m) of isothermal course,

$$\Delta G_m = \Delta H_m - T\Delta S_m \quad (3)$$

where the ΔG_m must be prone to be positive, making phase separation occur in the system. However, the entangled molecular chains restrict such separation because the entangled interpenetration is consecutive, consequently forming the intercross linking structures in the two-phase edge.

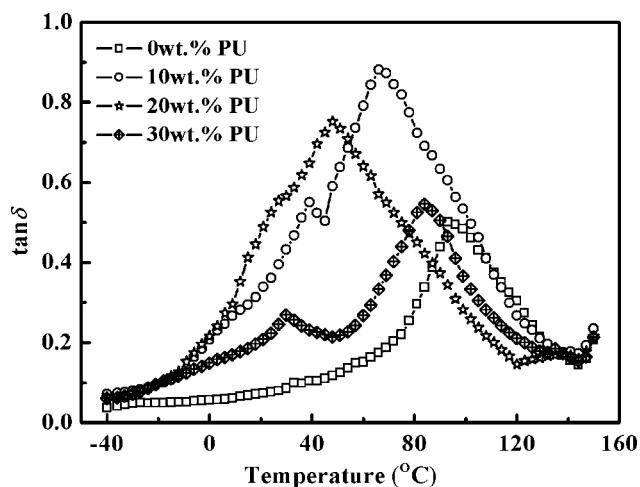


Figure 10 Temperature dependence of the $\tan \delta$ for composites.

As shown in Figure 10, the values of $\tan \delta$ for composites with different content of PU in the matrices, are higher than that of composite with pure epoxy, which proves that the polyurethane toughening of epoxy resin authentically enhances the dynamic mechanical property of epoxy resin. The peak value of $\tan \delta$ (0.892) appears at 68°C when 10 wt % PU exists in the matrix of composite, which is higher than those of other three composites. Furthermore, each of the curves of composites with 10 and 20 wt % PU has only one damping peak, but the composite with 30 wt % PU, has two damping peaks. Here, the short peak stands for the characteristic of PU, and another visible one stands for the characteristic of EP, which illuminates that there is two-phase separation for the composites with 30 wt % PU. The composites with 10 and 20 wt % PU, have fine dynamic mechanical properties because of the fact that two-phase compatibility of these two systems above is favorable, causing the damping peaks of EP and PU move to the low temperatures direction and high temperature direction, respectively, and both nearly form one high damping peak ultimately. However, the two-phase compatibility of the composite with 30 wt % PU is relatively poor. Thus, the phase separation between EP and PU hinders amalgamation of the two damping peaks, consequently leading to a poor dynamic mechanical property. Additionally, the effect of polyurethane toughening of epoxy resin can absorb and dissipate more energy, avoiding material's rupture caused by the energy accumulation instantaneously inside the materials, and thus improving the dynamic mechanical properties of composites. As shown in this figure, the high damping temperature regions (a temperature range of $\tan \delta > 0.5$) of composites, in which matrices with different content of PU, are 9, 69, 57, and 21°C in turn. It is obvious that the glass transition temperature regions of the composites with 10 and 20 wt % PU is wider than those of the other two composites. This can also prove that the composites with 10 and 20 wt % PU have better dynamic mechanical properties. As a straightforward consequence to these discussions, it can be concluded that the content of PU in the matrices is one of the important factors, which remarkably affect the dynamic mechanical properties of composites.^{28,29}

Figure 11 presents the temperature dependence of $\tan \delta$ for composites filled with fly ash with and without surface-treatment. As shown in this figure, the peak value of $\tan \delta$ for the former is 0.892, which is obviously higher than that of the latter (0.578), and the glass transition temperature region of the former is much wider. This is because the former has a better interfacial bonding condition compared with the latter, which can increase the damping contributions coming from the interface and spherically

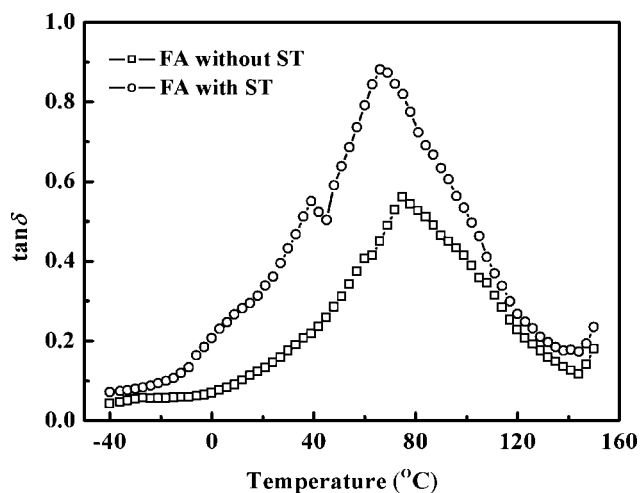


Figure 11 Effect of the surface condition of FA on the $\tan \delta$.

hollow structure. A better interfacial bonding condition may result in the movement of air and gas in the interior of micropores when material is subjected to a temperature loading, thus there must be friction between air and the spherical shells of fly ash. Furthermore, the air and gas close to the inner spherical shells will not be prone to move because of the impediment of spherical shells. As a consequence, both the sticky force among the air molecules and the frictional effect convert the mechanical energy into the heat energy, which can be dissipated. At the same time, there is a large shear strain in the interface phase, which can also increase the energy dissipation and enhance loss factor. Thus, it can be concluded that a better interfacial bonding condition can increase the damping contribution of interface and micropores.

CONCLUSIONS

The polyurethane-modified epoxy composites filled with fly ash are investigated in this article. The following conclusions can be drawn from the present study:

1. In fly ash reinforced modified epoxy composites, the damping is principally determined by the matrix viscoelasticity, and also includes the particles boundary sliding, interfacial sliding friction, and the energy loss behavior related to the spherically hollow structure.
2. The polyurethane toughening of epoxy resin enhances the impact toughness of epoxy resin, and the dynamic mechanical properties of composites with 10 and 20 wt % PU, are relatively better than those of the other two composites. Therefore, the content of PU in the matrices is one of the important factors, which remarkably

affect the dynamic mechanical properties of composites.

3. FTIR spectra illuminate that the chemical reactions occur and new chemical structures are formed, which are in favor of improving the compatibility between epoxy resin and polyurethane, and toughening of epoxy resin further.
4. FTIR test also reveals that there are several functionalized groups of γ -aminopropyl triethoxy silane on the surface of fly ash particulates with surface-treatment, which improves the interfacial bonding conditions between fly ash particulates and the matrix, and thus enhancing the dynamic mechanical properties of composites.
5. The SEM fractographs show that the fly ash particulates with surface-treatment are distributed homogeneously into the matrix, that combine well with the matrix and have no visible interface desquamation. These indicate that the effect of silane coupling agent on the interfacial bonding conditions between fly ash particulates and the matrix is quite significant.

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