Interaction and properties of epoxy-amine system modified with poly(phthalazinone ether nitrile ketone)

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ABSTRACT: An epoxy based on the tetraglycidyl 4,4′-diaminodiphenyl- methane (TGDDM)/bisphenol A type novolac(F-51) cured with 4,4′-diaminidiphenysulfone (DDS) has been modified with Poly (phthalazinone ether nitrile ketone)(PPENK). The interaction between the PPENK and epoxy resin have been investigated by differential scanning calorimetry (DSC), FT-IR, and dynamic mechanical analysis (DMA). The thermal and mechanical properties were characterized by thermogravimetric analysis (TGA), thermomechanical analysis (TMA), flexural, impact strength, and the critical stress intensity factor tests. The results showed that a large number of physical crosslinks formed by intermolecular and intramolecular hydrogen bonding indeed existed in the TGDDM/F-51/PPENK blends. These interactions gave good compatibility between PPENK and epoxy resin. So that any phase separation had not been detected by DMA and scanning electron microscope (SEM). Beyond that the interaction could also be a benefit to the thermal and mechanical properties. Compared with the neat epoxy resin, the critical stress intensity factor values reached the maximum at 10-phr PPENK, as well as the impact strength. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 42938.

INTRODUCTION

It is well known that cured epoxy resins possessing excellent dimensional stability, high modulus, and excellent creep properties, combined with the low melt viscosities of their precursors, are suitable for use as coatings, structural adhesives, and matrix resins in fiber–reinforced composites.[1–3] However, their high crosslink density is typically brittle to give them poor damage tolerance which is one of restrictions in the use of epoxy resins. Many methods have been developed to successfully enhance their fracture toughness.[4–8] Among these methods, although rubbers can be effective as toughening additives for epoxy resins, their drawbacks such as reduction in overall resin modulus and reduction in epoxy end-use temperature limit their application in aerospace. In addition, for highly crosslinked epoxy resins (trifunctional and tetrafunctional) commonly used in aerospace composites, rubbers are ineffective toughening additives due to a decreasing capability of the highly crosslinked resins to be plastically deformed.[9–11]

High-performance engineering thermoplastics have been selected as eligible modifiers for epoxy resins from the viewpoint of the maintenance of mechanical and thermal properties of the matrices. Therefore, toughening epoxy resins by blending with various thermoplastics, such as polysulfone (PSF),[5,12] poly(ether sulfone)

(PES),[13,14] poly (ether imide) (PEI),[15,16] poly(amide-amidic acid) (PAA),[17] and poly (ether ether ketone) (PEEK),[18,19] has been concentrated to develop and then widely applied in industry. The resulting morphology and extent of phase separation have been proved to affect the mechanical properties of the cured blend system. The dominant toughening mechanism of reaction-induced phase separation has been found in most of engineering thermoplastics/epoxy blend compositions.[14,15] However, in the miscible system like 3-diethylaminopropylamine-cured epoxy resin with PEI, the critical stress intensity factor (K_{IC}) and critical strain energy release rate (G_{IC}) decreased with the addition of PEI.[20] Guo *et al.* reported the homogeneous 4,4′-diaminodiphenylethane-cured epoxy/phenolphthalein poly(ether ether ketone) (PEK-C) blends. Their study result also showed that incorporation of PEK-C slightly decreased both the fracture toughness (K_{IC} and G_{IC}) and flexural properties.[18] As a result, phase separation induced by curing reaction of epoxy resin is expected. However, the complex interrelationship between phase behavior and mechanical properties of the toughening blends has not yet been clarified and is currently the subject of toughening thermosetting system.

Poly(phthalazinone ether nitrile ketone) (PPENK) is a recently developed high-performance engineering thermoplastic.[21]

PPENK is an amorphous polymer with a glass transition temperature (T_g) of 280°C, and its wholly aromatic twisted noncoplanar structure gifts it outstanding thermal stability and excellent mechanical properties as well as good solubility. Thus our attention has been paid to modify the fracture toughness of thermosetting resin by PPENK as modifier. In our previous work, we studied the fracture toughening and phase behavior of diglycidyl ether of bisphenol A (DGEBA)/PPENK cured with p-diaminodiphenylsulfone (DDS).[22] Moderate increase in the fracture toughness as estimated by notched impact strength was observed, although the DGEBA/PPENK blends formed homogeneous phase. This phase structure is different from most of engineering thermoplastics/epoxy blend compositions, thus it is necessary to research the interaction between PPENK and epoxy resins. In this article, we further examine PPENK as modifier to toughen polyfunctional epoxy resin. Tetrafunctional epoxy resin tetraglycidyl 4,4′-diaminodiphenyl- methane (TGDDM), cured with 4,4-diaminodiphenylsulfone (DDS), one of the most widely employed matrices for high performance fiber-reinforced composites and high-temperature coatings in aerospace applications,[23] was selected as matrix. And bisphenol A type novolac epoxy resin (F-51) was also used in order to decrease the crosslinking density of TGDDM/DDS. Our particular attention was paid to the interaction between the PPENK and TGDDM/F-51 epoxy resin. In addition, the thermal and mechanical properties, phase morphology, and toughening mechanisms were also investigated.

EXPERIMENTAL

Materials

There are two kinds of epoxy resins used in this study. One was tetraglycidyl 4,4′-diaminodiphenylmethane(TGDDM), a multifunctional, low viscosity, amber liquid sold as ERDM-434L (Shanghai Zhongsi industry, China). The other one was bisphenol A type novolac epoxy resin (F-51), a multifunctional, low
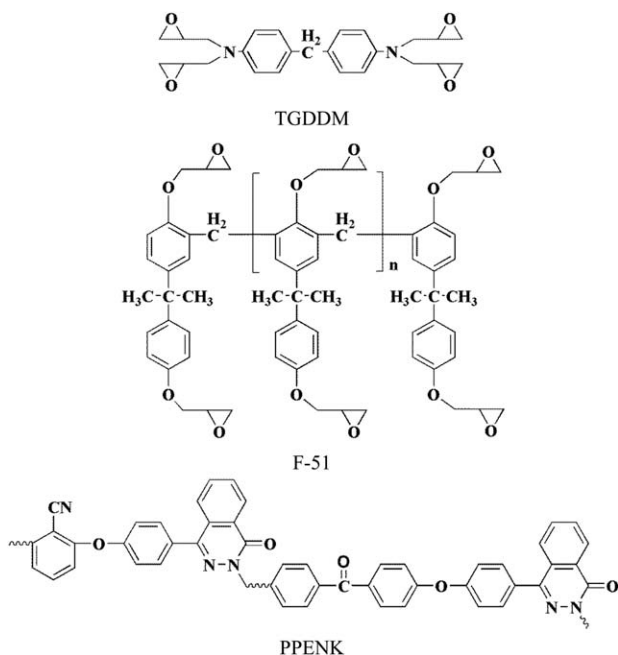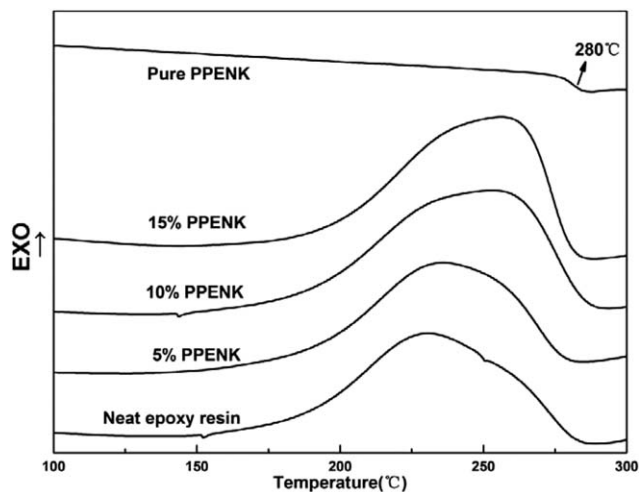
Figure 2. DSC thermograms of TGDDM/F-51/PPENK blends.

viscosity, light yellow liquid (Jinan Tianmao Chemical, Shandong, China). The curing agent was 4,4′-diaminodiphenylsulfone (DDS), a white powder, supplied by Sinopharm Chemical Reagent (Shanghai, China). The high-performance engineering thermoplastic was poly (phthalazinone ether nitrile ketone) (PPENK) ($\eta = 0.45$, $M_n = 1.8 \times 10^4$, supplied by Dalian Polymer New Materials Company, China). The chemical structure of TGDDM, F-51 and PPENK are shown in Figure 1.

Preparation of TGDDM/F-51/PPENK Blending System

The typical preparation of blending systems was described as follows. The mixed mass ratios of TGDDM, F-51, and the curing agent DDS were given as 60:40:45. All the blends were prepared using the solution mixing technique. The blending solution was first prepared with 0, 5, 10, 15 phr of PPENK in the TGDDM/F-51 chloroform solution, where phr represented the weight parts of PPENK per hundred parts of the total amount of TGDDM and F-51. The transparent homogeneous solution was then stirred at 100°C to remove the chloroform. Subsequently, the stoichiometric curing agent DDS was added into the viscous mixture. The resulting mixture was continuously stirred at 100°C until a homogeneous mixture was obtained.

Each sample was poured into a preheated chrome steel mould and degassed under vacuum at 100°C, then cured in an air circulating oven in curing procedures 130°C for 1 h plus 180°C for 2 h plus 200°C for 2 h. The cured substance was slowly cooled to room temperature to avoid cracking.

Figure 1. Chemical formulas of the materials.

Table I. Curing Characteristics of PPENK/TGDDM/F-51 Blend with DDS Hardener

phr-PPENK	$\triangle H$ (J g^{-1})	T_i (°C)	T_p (°C)	T_f (°C)
0	488	131	231	292
5	432	131	236	289
10	455	137	253	292
15	461	145	257	291

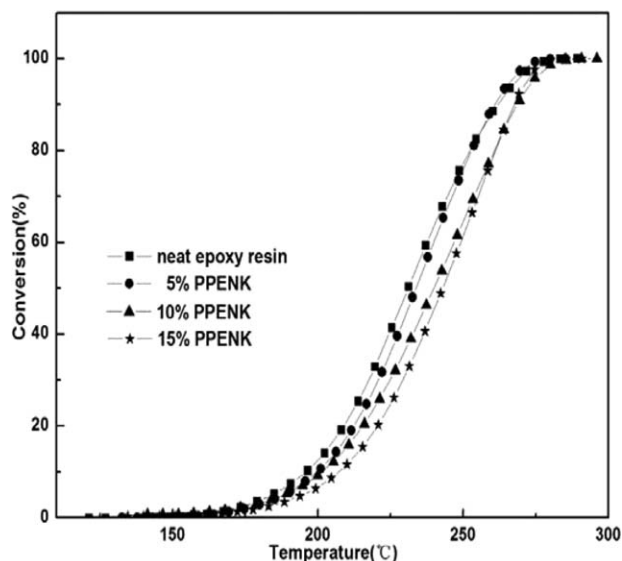**Figure 3.** Temperature dependence of conversion rate for varying content of PPENK.

Characterization

FT-IR Analysis. The analysis was conducted in a FTIR spectrometer (Nexus-470, Thermo Nicolet, USA) using samples in KBr pellets over the range of 400–4000 cm^{-1}.

DSC Analysis. An 8 $\pm$1 mg of samples were sealed in several alumina crucibles, and followed by scanning in a differential scanning calorimeter (Q20, TA instrument, USA) at a heating rate of 10°C min^{-1} from 50 to 350°C, with a stream of nitrogen at a flow rate of 40 mL min^{-1}.

TGA Analysis. A thermogravimetric analysis (Q500, TA instrument, USA) was applied to investigate the thermal stability of the cured samples at a heating rate of 20°C min^{-1} in nitrogen atmosphere (flow rate 40 mL min^{-1}).

DMA Analysis. Dynamic mechanical tests were carried out by a dynamic mechanics analyzer (Q800, TA instrument, USA) in
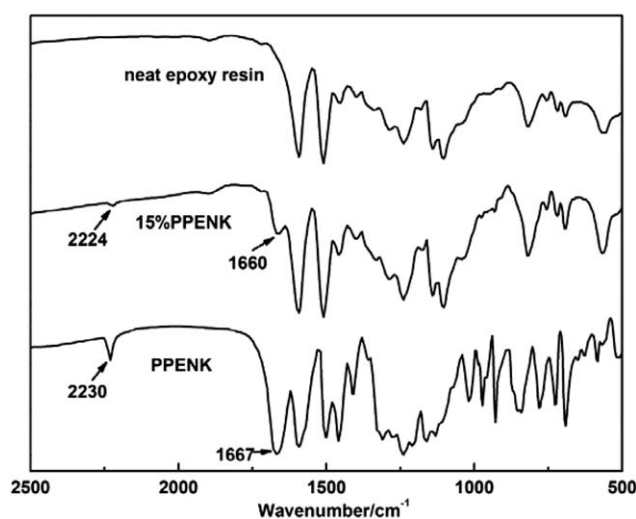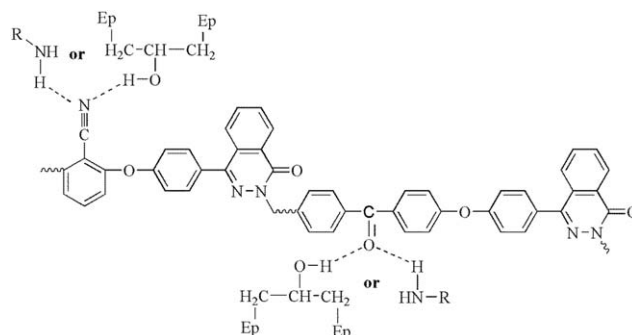

Figure 5. Schematic diagram of hydrogen bonding between PPENK and amine or hydroxyl groups.

the scanning temperature mode, in the range of 50–350°C, at a heating rate of 3°C min^{-1}, with an oscillating frequency of 1 Hz. The geometry of deformation was the clamp dual cantilever mode. The rectangular specimens with dimensions of $\sim$60 $\times$ 10 $\times$ 4 mm^3 were tested. We interpret the peak in tan δ as the glass–transition temperature (T_g).

TMA Analysis. Investigate the coefficient of thermal expansion of cured materials by using a thermomechanical analyzer (TMA, Q400, TA instrument, USA) at a scanning rate of 10°C min^{-1}. The test temperatures ranged from room temperature to 300°C at a frequency of 1 Hz.

Fracture Surface Morphology. The fracture surfaces of the failed specimens from impact measurement were analyzed with a scanning electron microscope (SEM, QUANTA 450, FEI, USA). The fractured specimens were etched with chloroform for 48 h, and were dried in vacuum for 24 h to remove the solvent. All specimens were sputter coated with a layer of gold before they were observed.

Mechanical Properties. A charpy impact strength measurement set (XCJ-4) was used to measure the impact property of the cured blends according to the Chinese standard GB/T 1843-2008. The specimen dimensions were (80 $\pm$ 2) mm $\times$

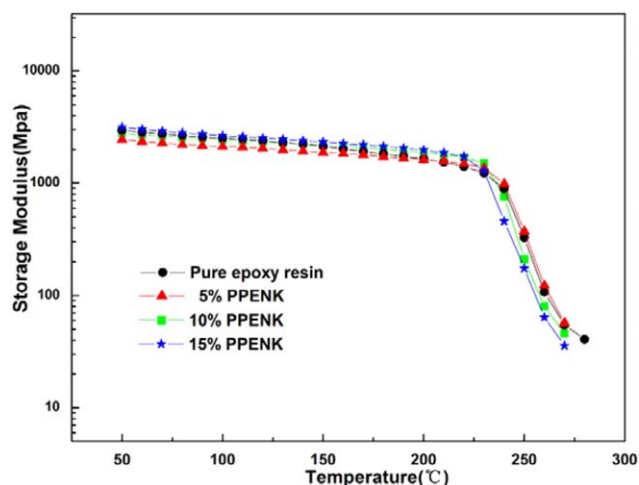**Figure 4.** FT-IR spectra of cured blends.

Figure 6. Dynamic mechanical spectra in terms of E′ for PPENK-modified TGDDM/F-51. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
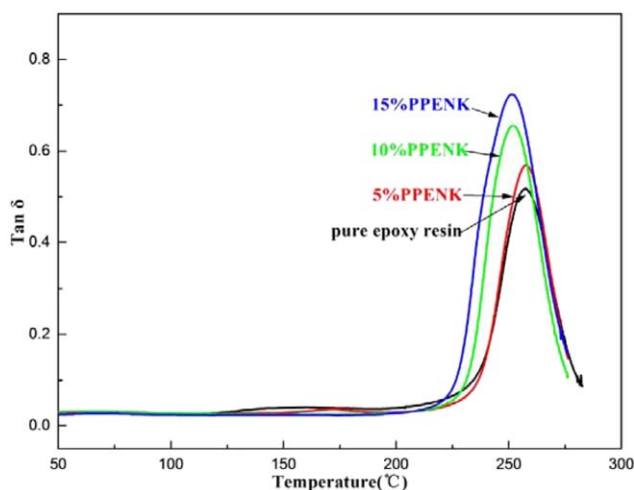
Figure 7. Dynamic mechanical spectra in terms of tan δ for PPENK-modified TGDDM/F-51. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
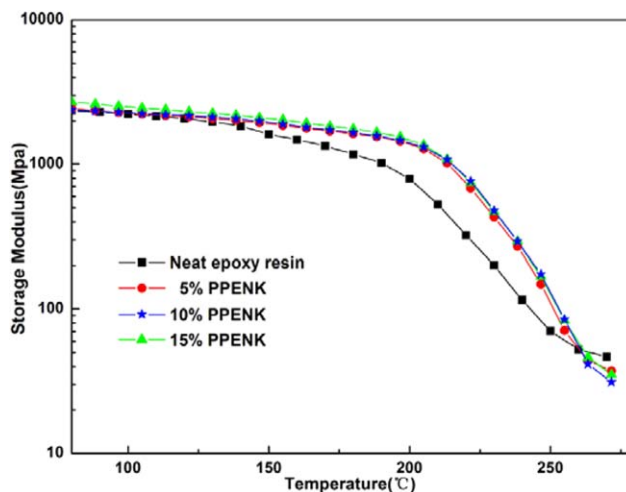

Figure 8. The storage modulus (E') curve of boiled samples. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

(10 ± 0.2) mm $\times$ (4 ± 0.2) mm without notch. At least five samples were used to measure the average impact strength.

The flexure properties of the cured blends were tested according to the Chinese standard GB/T 9341-2000. The specimen dimensions were (80 ± 2) mm $\times$ (10 ± 0.2) mm $\times$ (4 ± 0.2) mm. Three-point bending experiments on such specimens were performed at $23°C \pm 1°C$ on a WSM-50KN Electric Universal Testing Machine (Intelligent instrument equipment, Changchun, China).The displacement rate was 2 mm min^{-1}, and the flexural span was 64 mm.At least five specimens were used for every condition.

The critical stress intensity factor (K_{IC}) was determined according to ASTM standard D5045. The dimensions of specimens were 40 mm $\times$ 8 mm $\times$ 4 mm. A single-edge-notch three-point-bending test was used to obtain the K_{IC}. A natural crack was made by pressing a fresh razor blade into the notch. Five specimens were used to determine the K_{IC} of the materials.

RESULTS AND DISCUSSION

Interaction between PPENK and TGDDM Epoxy Resin
DSC experiments were first performed in the temperature range from 30 to 300°C at 10°C min^{-1} for the TGDDM/F-51/DDS matrix and for the toughened mixtures containing 5, 10, and 15 wt % PPENK contents, respectively, to explore the effect of adding toughening agent PPENK on the curing reaction of the epoxy system chosen in this research. The resulting DSC thermograms for the PPENK-modified epoxy mixture were shown

Table II. The Data of α-relaxation Peaks from the tan δ Curves of PPENK-modified TGDDM/F-51

phr-PPENK	Position (°C)	Height	Half width (°C)
0	258	0.52	26
5	258	0.57	25
10	252	0.65	26
15	252	0.72	31

in Figure 2. The integrated area of the exothermic curve was used to calculate the overall reaction heat ($\triangle H$). And the onset temperature (T_i), the peak exotherm temperature (T_p), and the terminal temperature at the curing reaction (T_f) were drawn from Figure 2 and presented in Table I. The conversion, defined as the ratio of partial heat $\triangle H_T$ at T to the overall heat $\triangle H$, versus temperature curves for neat TGDDM/F-51 and PPENK-modified TGDDM/F-51 were reported in Figure 3.

This system displayed the same tendency in the curing reaction as reported.[24,25] The addition of thermoplastic PPENK to TGDDM/F-51 hindered the epoxy curing reaction. For instance, from Figure 2, the curing temperatures like T_i and T_P were shifted to the higher temperature, respectively, with the increase of PPENK concentration in the epoxy mixture. In Figure 3, the blend system containing more PPENK, such as TGDDM/F-51/ 10% PPENK, attained lower conversion than neat epoxy TGDDM/F-51 at the same curing temperature. The delay in curing reaction should mainly correspond to physical reasons
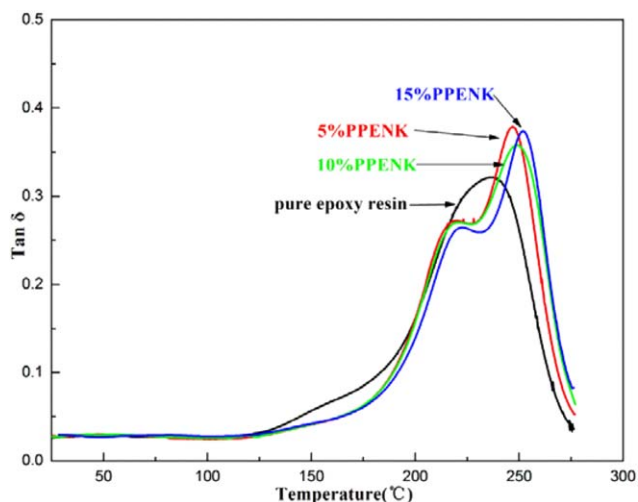

Figure 9. Tan δ curve of boiled samples. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]
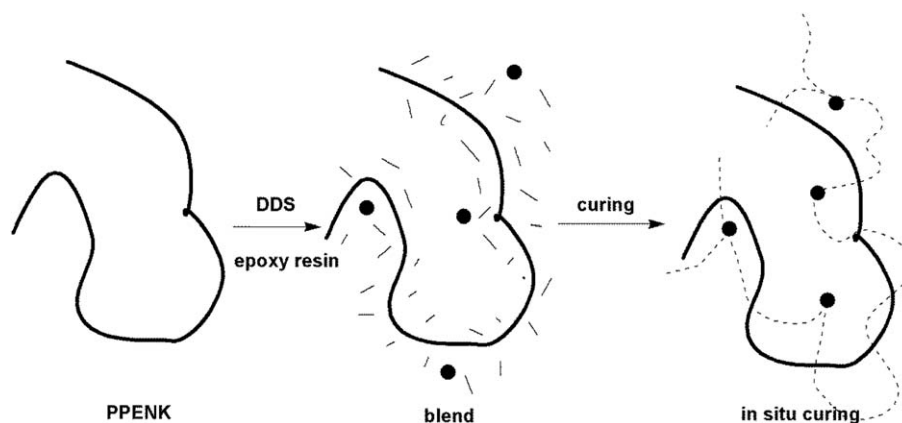
Figure 10. Schematic diagram of *in situ* curing mechanism.

such as dilution effect and/or viscosity increase because of thermoplastic addition,[26] or decrease of the density of reaction groups.[27] Thus the value of $\triangle H$ of TGDDM/F-51 modified with 5, 10, 15 phr PPENK were all less than that of the neat TGDDM/F-51. It has been reported that strong specific interactions, such as intermolecular hydrogen bonding, dipole–dipole interaction, and acid–base reaction etc., can produce a favorable mixing enthalpy in the blend.[28–30] It was seen from Table I that in TGDDM/F-51/PPENK system, their values of $\triangle H$ were updated with increasing concentration of PPENK in the system. The results implied that there were dozens of interactions between PPENK and TGDDM.

FT-IR analysis was carried out to further detect the interactions between the toughening agent PPENK and the epoxy TGDDM/F-51, and the results were shown in Figure 4. Compared to the typical band at 1667 cm^{-1}, attributed to the stretching vibrations of carbonyl groups, and the absorption at 2230 cm^{-1}, belonging to characteristic symmetrical stretching of —CN group in PPENK, they all shifted to lower wave numbers (1667cm^{-1}→1660cm^{-1} and 2230cm^{-1}→2224cm^{-1}) in the

cured TGDDM/F-51/15% PPENK system. It has been proved that the formation of hydrogen bond results in a decrease of the vibration frequency in IR spectroscopy.[28] These vibration frequency shifts in our system also indicated that the interaction between PPENK and TGDDM existed mainly via hydrogen bonding. The hydrogen bonding may form between the pendent cyano-group or ketone group in PPENK and amine or hydroxyl groups in TGDDM (Figure 5).

Dynamic mechanical analysis (DMA) was also used to demonstrate this interaction in our toughening system. The storage modulus (E') and loss factor (tan δ), derived from the DMA data, as a function of temperature were graphed in Figures 6 and 7, respectively.

The storage modulus (E') of blends offers a measurement of material stiffness under shear deformation at the corresponding temperature. As shown in Figure 6, the PPENK-modified TGDDM/F-51 exhibited very similar dynamic mechanical behavior to neat TGDDM/F-51 epoxy resin from 50 to 220°C. Their storage modulus in the rubber plateau (275°C) slightly decreased with the increase of PPENK content, maybe owing to decreasing the crosslink density of materials.

The network structure of the blends was also analyzed based on α-relaxation peak of the loss factor (tan δ). Figure 7 presented that all the blends identified only a single tan δ peak, indicating no detectable phase separation in the blends. The height and half width of the α-relaxation peak were calculated for each blend sample in order to observe trends in the crosslink density and network homogeneity of the blends. The position, height, and width at half height of the α-relaxation peaks derived from
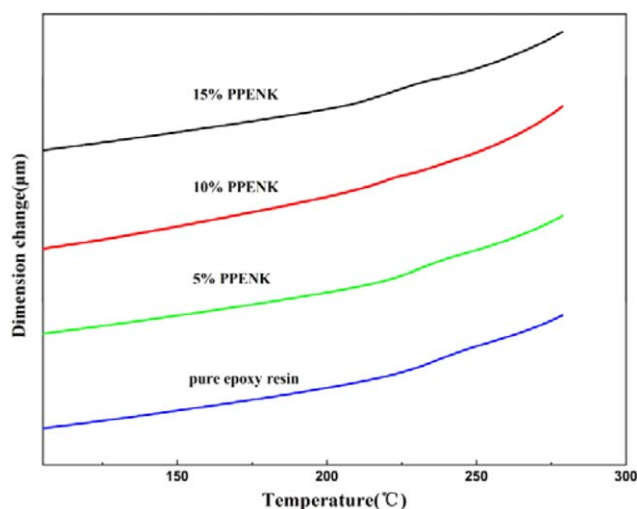

Figure 11. Dimension change as a function of temperature for the epoxy-PPENK blends. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table III. The Data of CTE from the TMA Curves for PPENK-modified TGDDM/F-51

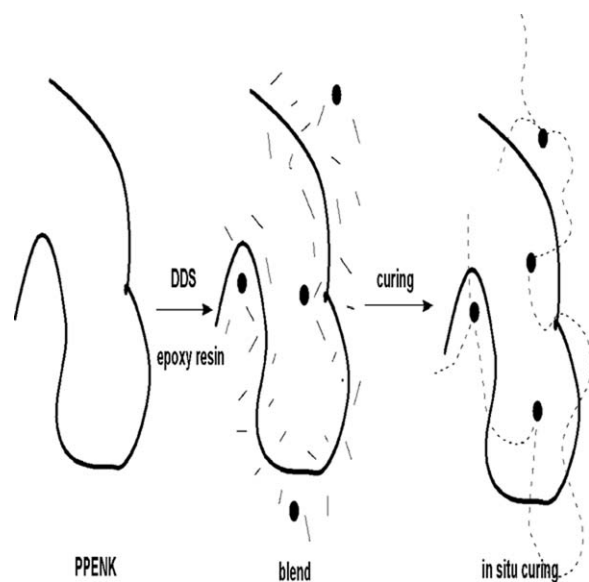| phr-PPENK	100–200°C, CTE[μm (m °C^{-1}) $^{-1}$]	250–275°C, CTE[μm (m °C^{-1}) $^{-1}$]
0	68.1 ± 1.5	162.7 ± 1.9
5	68.0 ± 0.8	178.8 ± 2.1
10	68.2 ± 1.2	191.0 ± 1.7
15	68.0 ± 1.4	192.5 ± 1.9

Figure 12. TGA and DTG curves of cured blends.

the tan δ curves for varying content of PPENK in the PPENK-modified TGDDM/F-51 blends was summarized in Table II. The height of the α-relaxation peak increased as the PPENK content in the blends was increased, suggesting that the cross-link density of the cured blends was decreased by the addition of PPENK into the system. It was worth mentioning that the T_g value of cured blends almost unchanged by virtue of excellent heat resistance of the toughening agent PPENK with 280°C of T_g value.

The hydrothermal properties of the blends were also investigated. The cured TGDDM/F-51/PPENK specimens were boiled for 48 h in the boiling deioned water. This method is usually used to evaluate hygrothemal property of composites in aerospace industry. The boiled specimens, also called wet samples, were taken out from the hot water and quickly dried by face tissue, followed by detecting DMA. The resulting E' and tan δ curves of the wet samples were shown in Figures 8 and 9, respectively. As seen in Figure 8, all of the wet samples' storage modulus slightly decreased. It is worth noting that the E' of neat epoxy resin decreased obviously from 150°C, while for the toughening blends, this temperature was updated to nearly 195°C. From Figure 9, obvious shoulder peaks were observed in the wet TGDDM/F-51/PPENK blends, while they exhibited only a single tan δ peak before boiled. The results implied that a large number of physical crosslinks formed by intermolecular and intramolecular hydrogen bonding indeed existed in the TGDDM/F-51/PPENK blends. In the process of boiling in the deioned water, these hydrogen bondings were destroyed by water molecular, leading to the occurrence of phase separation in molecular level. The shoulder peaks were corresponding to the epoxy-rich phase, and main peaks were corresponding to the PPENK-rich phase. This phenomenon supported the above DSC and FT-IR results. It maybe speculated that the development of an epoxy resin network at the presence of PPENK may follow a special mechanism like polycarbonate/epoxy blends.[11]

The reaction of epoxide and amine of harder DDS might take place along the PPENK chain, namely via an *in situ* curing mechanism (Figure 10). Semi-interpenetrating polymer networks (semi-IPNs) with molecular-level entanglement of segments were likely formed. Therefore, the hydrogen bonding gave good compatibility between PPENK and TGDDM epoxy resin. Beyond that, these physical interacting could also maintain the T_g and stiffness of the blends, despite the crosslink density decreased due to adding PPENK.

Thermal Properties

Thermomechanical analysis (TMA) was used to evaluate the thermal expansion coefficients (CTEs) of the blend epoxy resins. It has been known that the mismatch of the CTEs between polymer matrix and fiber will cause internal stress in the resulting composite.[30] The resulting CTEs in both the glassy and the rubbery state were recorded in Figure 11 and the corresponding values were collected in Table III. For thermosetting, its CTEs will usually enlarge if its crosslink density decreased. However, from Table III we could see that similar CTEs values in our blends were obtained in the glassy state and an increase of 30 μm (m °C^{-1})$^{-1}$ for the addition of 10 wt % PPENK into TGDDM epoxy resin compared to neat epoxy resin was observed in the rubbery state. It could be reasonably explained that the physical crosslinks caused by hydrogen bonding maintained the stiffness of the blends in the glassy state, while in the rubbery state these physical crosslinks were destroyed by heating to lead to increase their CTEs.

Thermal gravity analysis (TGA) measurements can serve as a useful indicator for material thermostability. Figure 12 shows TGA and DTG curves of the cured blends. The similar shape of the TGA curves of our toughening samples to the neat TGDDM epoxy resin signified that their degradation mechanism might be the same.

Table IV lists the TGA analysis of the cured blends. The temperatures for 5% weight loss ($T_{5\%}$) of the cured resins were all more than 350°C. Especially, the blends with 10 phr PPENK displayed the highest $T_{5\%}$ and the temperature for the maximum weight loss (T_{max}). It could be also found that Char yield at 800°C increased with increasing the addition of PPENK into the epoxy resins, due to excellent thermoxidative properties of the toughening agent PPENK used in our experiment. These results demonstrated that the incorporation of PPENK into the TGDDM/F-51/DDS system could improve the thermal stability of the resultant cured epoxy resins.

The limiting oxygen index (LOI) was calculated by the Formula (1).

Table IV. The Thermal Stability Data of the Cured PPENK-modified TGDDM/F-51

phr-PPENK	$T_{5\%}$(°C)	T_{max}(°C)	Char yield (%)	LOI
0	356	414	23.1	26.8
5	353	412	28.1	28.7
10	363	419	28.9	29.0
15	356	408	30.5	29.6

Table V. Mechanical Parameter of the Cured PPENK-modified TGDDM/F-51

phr-PPENK	Impact strength (kJ m^{-2})	K_{IC} (MPa m$^{-1/2}$)	Flexural strength (MPa)	Flexural modulus (GPa)
0	5.37 ± 0.7	0.67 ± 0.04	83.9 ± 12.1	3.58 ± 0.18
5	10.58 ± 1.8	0.93 ± 0.04	104.9 ± 9.7	3.47 ± 0.15
10	11.44 ± 1.6	1.28 ± 0.08	105.7 ± 8.6	3.37 ± 0.16
15	6.26 ± 1.5	0.96 ± 0.12	73.1 ± 6.6	3.58 ± 0.08

$$LOI = 17.5 + 0.4CR \qquad (1)$$

The Formula (1) was found by Van Krevelen in 1974. It described the correlation between LOI and char residue (CR) for halogen-free polymer only. In this equation, CR stands for the char yield on pyrolysis. The computation was shown in Table III. The LOI values increased with increasing PPENK content. Generally speaking, the materials can be classified as self-extinguishing materials if LOI > 27%. Thus the flame resistance of the PPENK modified TGDDM/F-51/DDS system was better than that of the unmodified materials.

Mechanical Properties

The data of flexural properties, impact strength and critical stress-intensity factor (K_{IC}) of the cured blends were summarized in Table V. The results showed that the K_{IC} value and impact strength for TGDDM/F-51/DDS/10-phr PPENK were all up to the highest values, about 91 and 116% of increase in K_{IC} and impact strength, respectively, compared to neat epoxy resin, reflecting that the toughness of the materials was obviously improved. It was worth noting that the flexural strength of blends was also updated to more than 20 Mpa, because of essential high-strength of PPENK and the physical crosslinks between PPENK and TGDDM epoxy resin, although flexural modulus of the blends slightly decreased.

Morphology of Fracture Surface

Scanning electron microscopy (SEM) was often used to observe the phase morphology of the polymer blends.[31] The SEM photographs of the fracture surfaces of the cured blends were shown in Figure 13. As shown in Figure 13, all the fractural
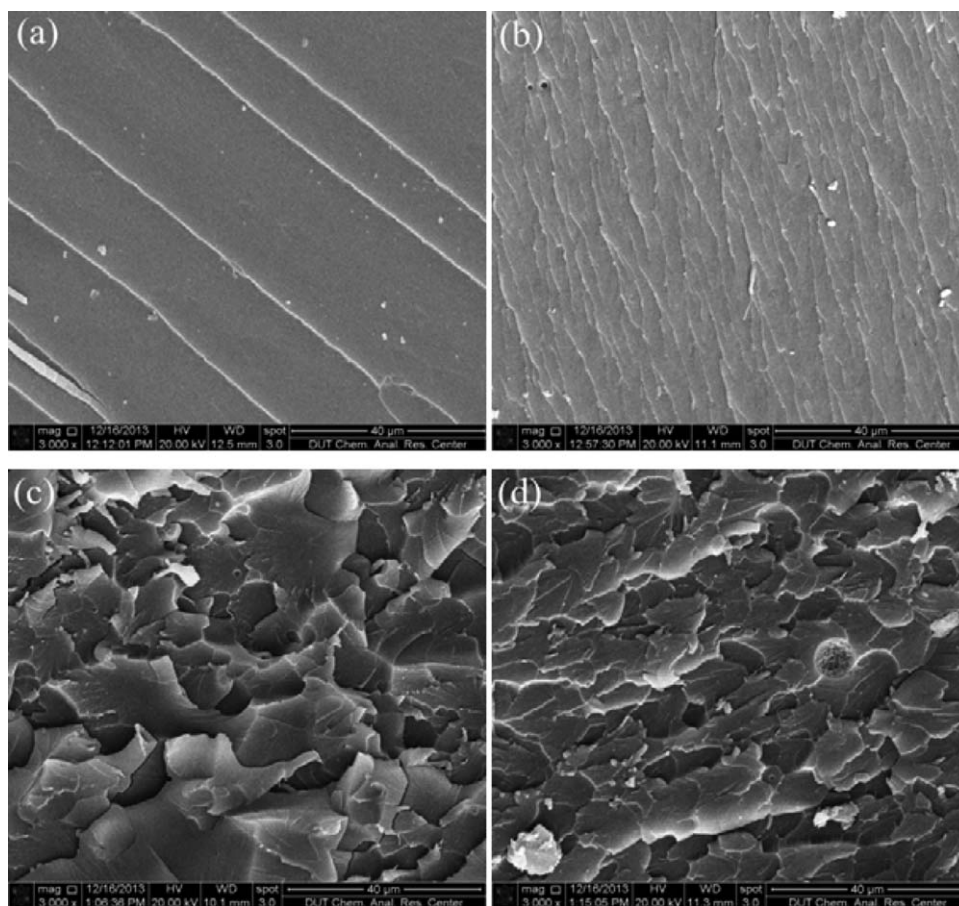

Figure 13. SEM micrographs of the fractured surfaces of failed specimens from impact test for epoxies modified with 0% (a); 5% (b); 10% (c); and 15% (d) PPENK (Magnification: ×3000).

surfaces did not display phase separation, indicating the compatibility of PPENK and TGDDM epoxy components. It is widely recognized that an important factor for improving the fracture toughness in a thermoplastic modified epoxy system is adequate interfacial adhesion. For the neat TGDDM epoxy resin, there was not plastic formation emerged on the fracture plane, and the fracture plane was smooth and glassy [Figure 13(a)]. This was a typical characteristic of brittle fracture. The crack propagated uninterruptedly, consequently there was no energy dissipation mechanism occurring here. The fracture surfaces of the epoxy resin modified by PPENK were different from that of the neat epoxy resin. The fracture surfaces of the modified epoxy blends were rough and ridge pattern. And highly deformed state could be observed. Therefore, one important factor for the increase in the fracture toughness was the plastic deformation of the matrix. Furthermore, the crack deflection and bifurcation at the crack tip region could also be observed, resulting absorbed more energy for propagation of the crack, thereby increasing the toughness.

CONCLUSIONS

In the present work, a novel high-performance engineering thermoplastic PPENK has been used to modify TGDDM/F-51/DDS epoxy system. PPENK/epoxy blends displayed unusual phase behavior, depending on intermolecular interaction. The reaction-induced phase separation had not been found in the PPENK/epoxy curing process. All the cured blends identified only a single tan δ peak. But if the cured specimens were boiled 48 h in the boiling water, obvious shoulder peaks were observed in the tan δ curves. The results implied that a large number of physical cross-links formed by intermolecular and intramolecular hydrogen bonding indeed existed in the TGDDM/F-51/PPENK blends. The DSC and FT-IR results also proved this hypothesis. The formation of these hydrogen bonds enhanced the miscibility of the blends. So that, any phase separation had not been detected by DMA and SEM. These physical interactions could maintain the T_g and stiffness of the blends. These results were desirable properties when used as coatings. The T_g, CTEs values, and $T_{5\%}$ of blends had not been decreased as usual. Compared with the neat epoxy resin, the critical stress intensity factor values reached the maximum at 10-phr PPENK, as well as the impact strength.

ACKNOWLEDGMENTS

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