The Epoxy–Polycarbonate Blends Cured with Aliphatic Amine—II. Thermal and Mechanical Properties Mechanisms

MIAW-LING LIN,[1] KUO-HUI CHANG,[1] FENG-CHIH CHANG,[1] MING-SHIU LI,[2] CHEN-CHI M. MA[2]

[1] Institute of Applied Chemistry, National Chiao Tung University, 1001 Ta Hsueh Road, Hsinchu, Taiwan 30050, Republic of China

[2] Institute of Chemical Engineering, National Tsing Hua University, Hsinchu, 30043, Taiwan, Republic of China

ABSTRACT: The transamidation reaction converts the carbonate and amine into N-aliphatic aromatic carbamate and urea, causing the equivalent of oxirane and amine nonstoichiometric in the primary cure stage. After postcure, the substitution reaction takes place and results in a more tightly crosslinked structure. Such a higher crosslinking density is responsible for higher T_g, tensile strength, and tensile modulus in the postcure stage than that in the primary and secondary cure stages. This trend is more pronounced in those PC–epoxy blends containing higher molecular weight aliphatic amine or a higher content of PC. This phenomenon is due to the difference in the fraction of amino group of aliphatic amine consumed in the transamidation. PC–epoxy/aliphatic amine blends show minor improvement in the high strain rate Izod impact tests, while toughness improvement for some blends is substantial at low strain rate tensile tests. © 1997 John Wiley & Sons, Inc. J Polym Sci B: Polym Phys **35:** 2183–2191, 1997
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INTRODUCTION

Thermal and mechanical properties of various PC–epoxy/aliphatic amine blend systems have been investigated previously.[1–3] Mera and Umetani[1] used the amine derived from isophoronediamine (IPDA) and bis(4-amino-3-methylcyclohexyl)methane to cure the PC–epoxy blends and found that the flexural modulus of the blend is increased with the increase of the PC content. However, the presence of PC causes reduction of the heat-deflection temperatures (HDT) of the blends. On the contrary, Chen et al.[2] studied PC–epoxy blends cured by diethylenetriamine (DETA) and showed that the presence of 10 wt % of PC leads to T_g reduction but shows no clear

trend of the resultant Young's moduli (E) of the postcured samples. Rong and Zeng[3] utilized tetraethylenepentamine (TEPA) to cure PC–epoxy blends and resulted in slight variations on T_gs.

Essentially none of these earlier reports discussed the influence of the chemical reactions involved during curing on the resultant thermal and mechanical properties of the cured PC–epoxy/aliphatic amine blends. Our previous studies of the PC–epoxy/aliphatic amine blends[4,5] showed that the aliphatic amine can react readily with carbonate of PC at a much higher rate than the normal curing reaction at ambient condition to produce carbamate, and the remaining amine then proceeds the normal curing reaction with oxirane. In other words, the PC long chains present in the original PC–epoxy/aliphatic amine blend actually have been instantly scissored by the aliphatic amine. These newly found chemical aspects in the PC–epoxy/aliphatic amine blend system require

revision of many previous explanations of the resultant properties. For example, Chen et al.[2] explained the reduction of T_g with PC presence as the result of a dilution effect based on earlier reports.[6,7] We interpreted the observed T_g variations of the blends depending on several factors, including degree of crosslinking, plasticization, chain ends, and flexibilizers in the cured PC–epoxy/aliphatic amine blends.[4]

In this article, various PC–epoxy blends are cured by various molecular weights of polyoxypropylene diamines (Jeffamine D230, D400, and D2000). The chemical structures of a blend at various thermal histories are closely related with their morphological, thermal, and mechanical properties.

EXPERIMENTAL

Materials

The materials utilized were described in Part I.[5] The polycarbonate and liquid epoxy resin obtained from Dow Chemical Company are Calibre 301-15 and DER 331, respectively. The polyoxypropylene diamines with different molecular weights used as hardeners from Huntsman Chemical Co. are D230, D400 and D2000.

Procedures and Instrumentations

The procedures of preparing the blends were also described in Part I.[5] Blend compositions, together with their corresponding codes, are listed in Table I. Stoichiomatic equivalent of epoxy to amine was employed in all the blends.

Curing Procedures

The typical curing procedures were carried out by mixing the PC–epoxy blend with polyoxypropylene diamine (POPDA) at room temperature for 3 min, followed by primary curing at 80°C for 2 h, secondary curing at 150°C for 2 h, and postcuring at 180°C for 2 h.

Infrared Spectroscopy

One drop of the mixture was placed between two sodium chloride plates and are then mounted on a sample holder in the IR instrument. Infrared spectra were obtained on a Perkin–Elmer 842 Infrared Spectrometer with a resolution of 2.4 cm^{-1}. The observed spectrum was separated into many

Table I. The Compositions and Codes of the PC–Epoxy/POPDA Blends

| Code	Epoxy (g)	Amine (g) D230/D400/D2000	PC (g)	Epo. + Am. (wt %)	PC (wt %)
A00	100	30/—/—	0.00	100	0
A03	100	30/—/—	4.02	97	3
A06	100	30/—/—	8.30	94	6
A09	100	30/—/—	12.86	91	9
A12	100	30/—/—	17.73	88	12
B00	100	—/50/—	0.00	100	0
B03	100	—/50/—	4.64	97	3
B06	100	—/50/—	9.57	94	6
B09	100	—/50/—	14.84	91	9
B12	100	—/50/—	20.45	88	12
C00	100	—/44/12.5	0.00	100	0
C03	100	—/44/12.5	4.84	97	3
C06	100	—/44/12.5	9.99	94	6
C09	100	—/44/12.5	15.48	91	9
C12	100	—/44/12.5	21.34	88	12
D00	100	—/38/25	0.00	100	0
D03	100	—/38/25	5.04	97	3
D06	100	—/38/25	10.40	94	6
D09	100	—/38/25	16.12	91	9
D12	100	—/38/25	22.23	88	12
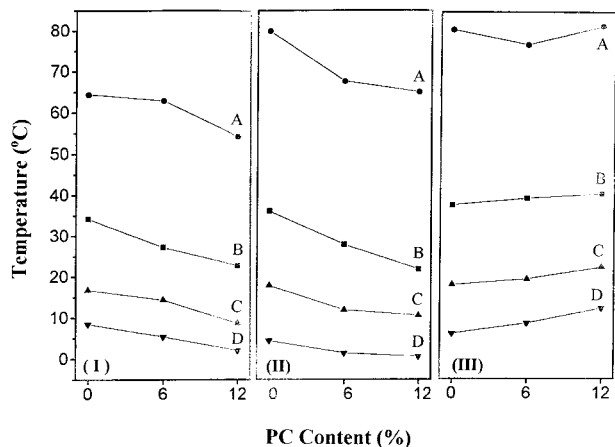
Figure 1. T_g of the PC–epoxy/POPDA blend cured after (I) primary cure stage, (II) secondary cure stage, and (III) postcure stage: (A) Code A series, (B) Code B series, (C) Code C series, and (D) Code D series.

components by a least-squares curve-fitting technique.

Differential Scanning Calorimeter

The glass transition temperatures were determined by a Differential Scanning Calorimeter (DSC), a Du Pont 2100, using a heating rate of 10°C/min in the dynamic scan.

Mechanical Property Testing

Tensile properties were measured by an Instron Universal Testing Machine Model 4201 using a crosshead speed of 10 mm/min as described in ASTM D-638. The area under the stress–strain curve was defined as tensile toughness. Notched Izod impact strengths were measured according to ASTM D-256 method.

RESULTS AND DISCUSSION

T_gs of the PC–Epoxy/POPDA Blend

Products from various stages of curing were examined by DSC to determine their T_gs after different stages of curing, and the results are summarized in Figure 1. T_gs of all the blends investigated are increased by following sequential stages of curing, from primary, secondary, and postcuring. The general trend clearly shows that the T_g of the cured blend at any stage of curing decreases with increasing of the amine molecular weight due to

lower crosslinking density, as would be expected. Additionally, T_gs of the PC–epoxy/POPDA blends after primary and secondary curing decrease with increasing of the PC content, while T_gs of the corresponding postcured products show no clear trend. These phenomena can be attributed to different chemical reactions involved during curing of these blends containing different amounts of PC.

Figure 2 shows the dynamic DSC scans of the neat epoxy/POPDA B00 system (see Table I) in the temperature range of 0–300°C where the exotherm (curve A) represents the normal curing reaction. This small exotherm remains in the sample after the primary curing (curve B), an indication of an incomplete normal curing reaction. This exothermic peak disappears completely in curves C and D. Comparing curves A to D of Figure 2 implies that this neat B00 system has already completed the normal curing reaction after the secondary stage of curing (150°C for 2 h). This result can also be confirmed again by infrared analyses. Curve A of Figure 3 represents the oxirane fraction of the B00 blend after various stages of curing determined by IR where the oxirane con-
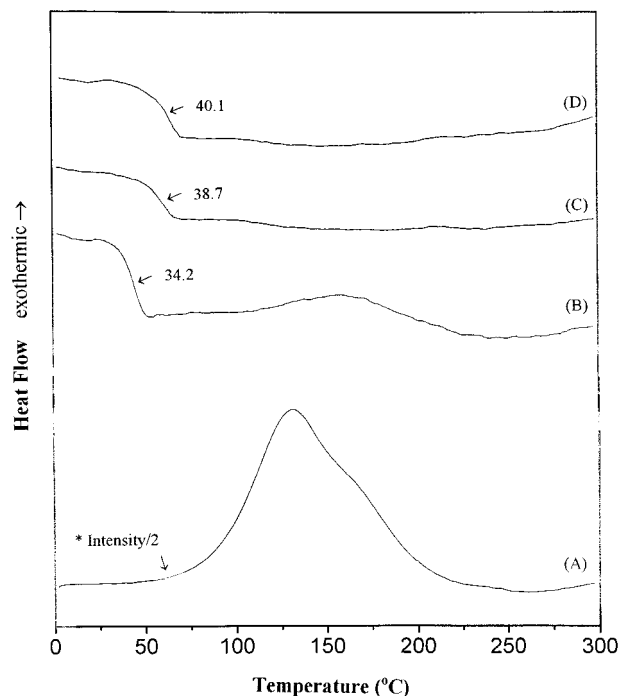

Figure 2. Dynamic DSC runs of the neat B00 blend (Epoxy/D400 = 100/50) in the temperature range between 30 and 300°C: (A) initial, (B) after 80°C 2 h, (C) 150°C 2 h, and (D) 180°C 2 h.
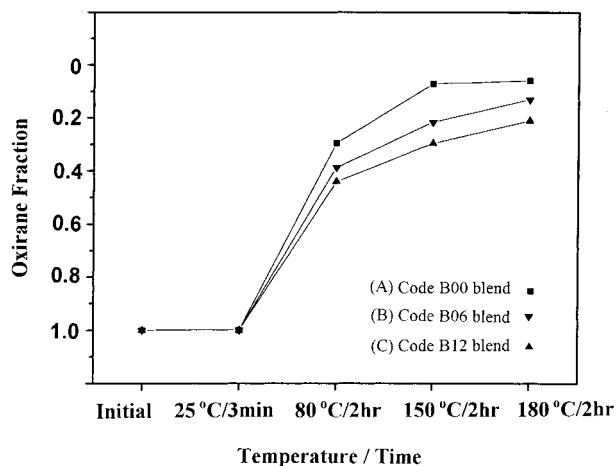
Figure 3. Plots of oxirane conversion vs. various curing procedure. (A) Code B00 blend, (B) Code B06 blend, and (C) Code B12 blend.

tent of the blend approaches zero after the secondary curing (150°C for 2 h).

Figure 4 presents the dynamic DSC scans of the PC–epoxy/POPDA blend (B06, PC 6 wt %) where curve A shows two distinct exothermic peaks. The first and larger peak represents the
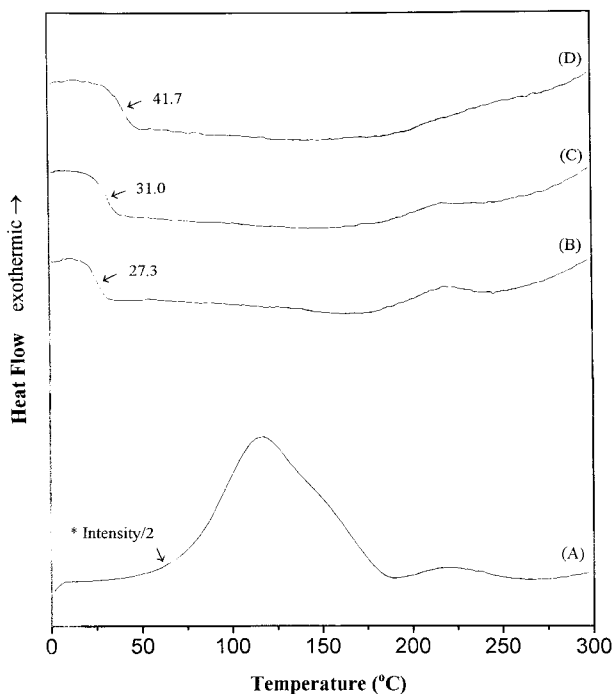

Figure 4. Dynamic DSC runs of Code B06 blend in the temperature range between 30 and 300°C: (A) initial, (B) after 80°C 2 h, (C) 150°C 2 h, and (D) 180°C 2 h.

Table II. Characteristic IR Bands of Carbonyl Groups

Band (cm^{-1})	Assignment
1780	C=O stretch of Ar—O—CO—O—Ar
1740	C=O stretch of R—NH—CO—O—Ar
1660	C=O stretch of R—NH—CO—NH—R
1720	C=O stretch of R—NH—CO—O—R
1810	C=O stretch of cyclic carbonate

heat generated from the normal cure reaction between amine and epoxy, while the second and smaller peak probably come from the later substitution reaction. The first and the larger exothermic peak disappears totally in curves B, C and D. Additionally, Figure 4 shows the intensity of the smaller exothermic peak decreasing at high temperature and disappearing gradually from curve B to curve D. Another characteristic revealed in this figure is that the T_g of B06 blend (PC 6 wt %) increases more substantially between secondary and postcuring than that of the neat B00 system (also see Fig. 1). For example, T_gs of the B06 blend (PC 6 wt %) after secondary and postcuring are 31.0 and 41.7°C, while the corresponding T_gs of the B00 system are 38.7 and 40.1°C [Fig. 1(II) and (III)]. This phenomenon can be interpreted in terms of different chemical structures of the carbonyl group between the neat blend and this PC-containing blend. Various types of carbonyl groups are produced during various stages of curing that have been identified by infrared spectroscopy previously[5] as shown in Table II. The characteristic IR peak changes through various stages of curing are separated into many components by a least-squares curve-fitting technique and summarized in Figure 5. Figure 5 shows approximate fractions of various carbonyl groups at various stages of curing including aromatic carbonate (curve A), N-aliphatic aromatic carbamate (curve B), urea (curve C), N-aliphatic aliphatic carbamate (curve D), and cyclic carbonate (curve E) of the B06 blend. Immediately after mixing (25°C, 3 min), approximately 80% of the original aromatic carbonate has been converted into N-aliphatic aromatic carbamate (curve B). After the primary curing (80°C, 2 h), over 90% of the aromatic carbonate has disappeared and the urea becomes the major component (70%, curve C). The N-aliphatic aromatic carbamate exists as a chain end while the urea acts as a chain extender in the epoxy
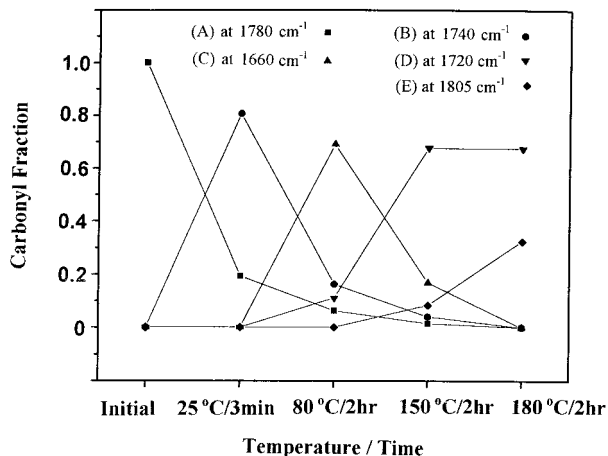
Figure 5. Plots of IR absorption fraction in the carbonyl region of the B06 blend at various curing procedure. (A) at 1780 cm^{-1}, (B) at 1740 cm^{-1}, (C) at 1660 cm^{-1}, (D) at 1720 cm^{-1}, and (E) at 1805 cm^{-1}.

network structure.[4,5] This structure should reduce the crosslinking density of the cured resin network and, therefore lowers the T_g of the blend. Curve D of Figure 5 shows that N-aliphatic aliphatic carbamate dominates the carbonyl fraction ($>60\%$) after postcuring. When the N-aliphatic aliphatic carbamate is formed, this structure can be considered as a crosslinker of the network.[4] Additionally, the substitution reaction of the urea by reacting with hydroxyl group would release amine that can also proceed the normal curing with the remaining oxirane.[5] In other words, the crosslinking density of the blend can be further increased at a higher temperature during postcuring and results in higher T_g of this blending system. However, comparing this B06 product with the product of the B00 neat system, the B06 blend (PC 6 wt %) is less than stoichiometric (epoxy to amine) than that of B00 system. Curve B of Figure 3 also provides additional evidence that relatively more unreacted oxirane is left from the B06 blend than that of the B00 blend. This result indicates that the transamidation between PC and amine tends to cause lower crosslinking density and, therefore, lowers the T_g of the B06 blend (PC 6 wt %) than the corresponding B00 blend.

Figure 6 gives the dynamic DSC scans of the B12 blend (PC 12 wt %) where curve A also shows two distinct exothermic peaks. Curves B, C, and D again show the disappearance of the first exotherm, while the glass transition temperature increases following primary, secondary, and postcuring. As revealed in this figure, the T_g of the
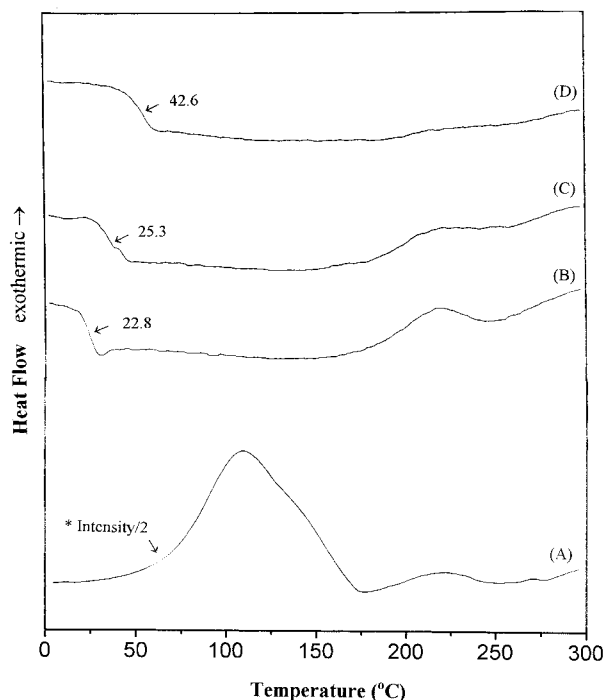

Figure 6. Dynamic DSC runs of Code B12 blend in the temperature range between 30 and 300°C: (A) initial, (B) after 80°C 2 h, (C) 150°C 2 h and (D) 180°C 2 h.

B12 blend (PC 12 wt %) exhibits a similar trend as the B06 blend (PC 6 wt %). Fractions of various carbonyl groups of this B12 blend at various stages of curing are shown in Figure 7. The diagrams of carbonyl fraction (Fig. 7) and oxirane
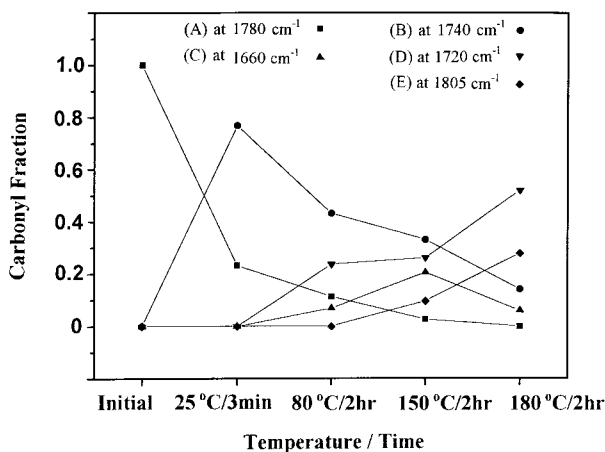

Figure 7. Plots of IR absorption fraction in the carbonyl region of Code B12 blend at various curing procedure. (A) at 1780 cm^{-1}, (B) at 1740 cm^{-1}, (C) at 1660 cm^{-1}, (D) at 1720 cm^{-1}, and (E) at 1805 cm^{-1}.
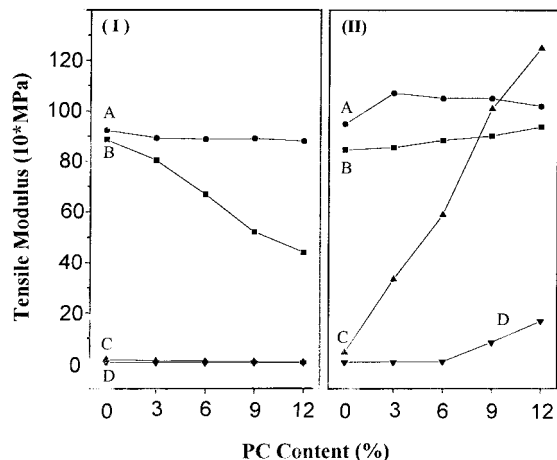
Figure 8. Tensile modulus of the PC–epoxy/POPDA blend cured after (I) secondary cure stage and (II) post-cure stage: (A) Code A series, (B) Code B series, (C) Code C series, and (D) Code D series.

fraction (curve C of Fig. 3), indicate the T_g of the B12 blend (PC 12 wt %) increasing after post-curing.

Essentially, all the measured T_gs of the PC–epoxy/POPDA blends decrease with PC content increasing after primary and secondary curing stages. However, such trend is reversed after postcuring as shown in Figure 1(I)–(III). T_g of a cured PC–epoxy/POPDA blend depends on many factors including crosslinking, copolymer, and plasticizer effect. The transamidation reaction between PC and amine tends to reduce the cross-linking density of the blend, as mentioned above. Additionally, the transamidation reaction also releases Bisphenol A and/or PC-oligomers that can act as plasticizers for the cured network. However, the copolymer effect implies that the new carbamate crosslinking within the network is not structurally and chemically identical to the normally cured neat epoxy network. Therefore, the copolymer effect on a network can either raise or reduce the T_g of the blend.[8] Comprising these effects, the observed T_g of the PC–epoxy/POPDA blend after postcuring is not very consistent, as shown in Figure 1(III).

Tensile Modulus of the Cured PC–Epoxy/POPDA Blends

Tables III–VI tabulate the tensile properties and impact strengths after secondary and postcuring. Figures 8 summarizes these tensile moduli. Glass

transition, crosslinking density, and testing conditions (temperature and rate) are the important intrinsic and extrinsic factors affecting the measured modulus of any polymeric material. Certain blends have their T_gs close to testing temperature (15–20°C) that show the greatest modulus variation, as would be expected (compare Fig. 1 and Fig. 8).

After secondary and postcuring, those neat blends (% PC = 0) containing lower molecular weight of POPDA (A00 and B00) are at a glassy state with high moduli, while others (C00 and D00) are at a rubbery state with extremely low moduli (Tables III–VI and Fig. 8). Chain length (in terms of molecular weight) of the POPDA dictates the crosslinking density, T_g, and thus the modulus of the cured blend. After secondary stage curing, the blend's modulus decreases with the increase of the PC content [Tables III–VI and Fig. 8(I)] due to reduced crosslinking density caused by the transamidation reaction. As mentioned earlier, the carbonate group of PC can readily react with amino group of POPDA and consumes a fraction of the originally stoichiomatric amine curing agent to yield N-aliphatic aromatic carbamate, urea, PC oligomer, and Bisphenol-A(BPA) monomer. The reacted amino group of POPDA is unable to react with oxirane during primary and secondary curing and, therefore, causes the normal curing reaction less than stoichiometric and thus lower crosslinking density. Additionally, those produced N-aliphatic aromatic carbamate, urea, and PC oligomer and BPA after the secondary stage of curing are existed as chain ends or free small molecules that can act as plasticizers of the network to reduce network T_g.

In contrast to the trend observed after secondary curing, modulus of the postcured PC–epoxy/POPDA blend tends to increase with the increase of the PC content [Tables III–VI and Fig. 8(II)]. Substitution reactions occurring during postcuring are responsible for the observed phenomenon. As mentioned earlier, N-aliphatic aromatic carbamate and urea yielded from the transamidation reactions are able to react with the hydroxyl groups of the cured epoxy network and become part of the network structure. In other words, these substitution reactions tend to transform the original network into a more tightly crosslinked structure. Therefore, modulus of the postcured PC–epoxy/POPDA blend is increased with the increase of PC content.

Table III.   The Tensile Properties, Impact Strength, and T_g of the Code A Series Blending Systems

PC Content (wt %)	Secondary Cure Stage					Postcure Stage				
	0	3	6	9	12	0	3	6	9	12
Stress at max. load (MPa)	48.8	46.8	46.2	45.7	40.7	65.5	69.2	60.8	52.5	45.3
Young's modulus (MPa)	923	892	887	889	879	948	1069	1048	1048	1017
Elongation (%)	10.1	8.7	6.5	5.2	3.3	5.8	5.5	5.4	4.9	4.3
Toughness (J/m²)	4.97	4.35	2.45	1.59	0.62	1.38	1.37	1.31	1.25	0.95
Notched izod impact strength (J/M)	5.94	3.78	2.64	2.47	2.12	7.62	7.27	6.34	6.10	5.84
T_g (°C)	80.2	—	68.5	—	66.1	80.8	—	77.2	—	81.6

Tensile Elongation and Tensile Toughness

Tensile elongation and toughness of A-series blends (containing the lowest MW of POPDA) decrease substantially with the increase of the PC content after the secondary stage of curing despite a decreasing trend on modulus and crosslinking density (Table III). If the network crosslinking density is achieved through the use of less than stoichiometric equivalent of the curing agent, this network structure should be more flexible and should have resulted in higher tensile elongation and toughness. However, the less than stoichiometric equivalent of POPDA of these PC–epoxy/POPDA blends is caused by the transamidation reaction to generate additional chain ends of *N*-aliphatic aromatic carbamate and urea. Free chain ends in a network may provide weak points to nucleate voids under stress, and thus decrease tensile elongation and toughness. The expected toughness gain by the lower crosslinking network is offset by the creation of more chain ends. Modulus of the blend actually increases with the in-

crease of PC content after postcuring (Table III). The observed reduction on tensile elongation and toughness following the increase of PC content after postcuring is relatively less significant than that after secondary curing (Table III). Substitution reactions occurring during postcuring tend to increase crosslinking density and decrease the number of chain ends within the network structure.

On the contrary, tensile elongation and toughness increase with the increase of PC content after the secondary stage of curing for the B-series blends (Table IV). This phenomenon can be interpreted as these blends shifting from the glassy state to the rubbery state by increasing the PC content (Fig. 1). Higher tensile elongation and toughness near its T_g or rubbery state than in the glassy state can be expected from any polymeric material. After postcuring, all B-series blends are in a glassy state and both elongation and toughness decrease with the increase of PC content, as would be expected. Similar to the A-series, the

Table IV.   The Tensile Properties, Impact Strength, and T_g of the Code B Series Blending Systems

PC Content (wt %)	Secondary Cure Stage					Postcure Stage				
	0	3	6	9	12	0	3	6	9	12
Stress at max. load (MPa)	50.7	39.2	27.0	17.4	15.5	35.9	48.5	44.8	41.7	34.2
Young's modulus (MPa)	840	792	668	518	438	845	855	882	900	936
Elongation (%)	7.6	13.1	46.1	121.7	143.8	5.9	5.8	5.4	4.7	3.9
Toughness (J/m²)	2.31	3.44	7.01	15.7	20.0	1.50	1.47	1.25	1.01	0.68
Notched izod impact strength (J/M)	3.04	3.50	3.82	3.97	3.37	3.87	3.17	2.97	2.77	2.17
T_g (°C)	38.7	—	31.0	—	25.3	40.1	—	41.7	—	42.6

Table V.   The Tensile Properties, Impact Strength, and T_g of the Code C Series Blending Systems

PC Content (wt %)	Secondary Cure Stage					Postcure Stage				
	0	3	6	9	12	0	3	6	9	12
Stress at max. load (MPa)	10.1	8.7	6.5	5.9	3.4	10.9	11.0	11.3	20.0	22.0
Young's modulus (MPa)	15.1	8.8	7.6	6.4	3.3	44.6	332	587	1006	1247
Elongation (%)	125	122	105	98.0	92.0	105	55.0	34.7	29.0	5.6
Toughness (J/m²)	7.5	6.4	5.1	4.9	3.1	6.8	4.4	4.2	3.3	0.9
Notched izod impact strength (J/M)	—[a]	—[a]	—[a]	—[a]	—[a]	6.22	5.52	4.70	4.03	4.01
T_g (°C)	21.5	—	15.9	—	14.7	21.7	—	22.9	—	25.7

[a] Nonbreak.

blend possessing a higher PC content has greater crosslinking density (and modulus) and, therefore, has lower tensile elongation and toughness, as shown in Table IV.

For the C-series blends with a relatively higher MW of POPDA (Table V), all blends are in the rubbery state after the secondary stage of curing and possess high elongations. Again, the number of chain ends plays the decisive role in dictating the resulted tensile elongation and toughness. The blend containing more PC results in the lower tensile elongation and toughness due to a greater number of chain ends within the network. After postcuring, this C-series blends transform gradually from the rubbery state into the glassy state with the increase of the PC content [Fig. 1 and Fig. 8(II)] and, therefore, the resultant tensile elongation and toughness decrease quite drastically.

For D-series blends containing the highest POPDA molecular weight (Table VI), the trend after secondary curing is similar to that of C-series blends (Table V). After postcuring, those blends containing higher PC (9 and 12%) are close to their respective T_gs and, therefore, have higher modulus and lower elongation and toughness.

Impact strength has the same trend as tensile elongation and toughness as shown in Tables III–VI. It is worth mentioning that the impact strengths from those blends (in the rubbery state) remain extremely low even though they are in the rubbery state, and the corresponding tensile elongation and toughness are very high.

CONCLUSION

Glass transition temperature and mechanical properties of the PC–epoxy blend cured by ali-

Table VI.   The Tensile Properties, Impact Strength, and T_g of the Code D Series Blending Systems

PC Content (wt %)	Secondary Cure Stage					Postcure Stage				
	0	3	6	9	12	0	3	6	9	12
Stress at max. load (MPa)	2.2	1.6	1.0	0.8	0.7	3.4	3.6	3.9	5.4	9.8
Young's modulus (MPa)	2.6	2.0	1.2	1.0	0.9	4.0	4.1	6.5	82.1	167.3
Elongation (%)	127	136	139	142	147	122	128	117	111	86.5
Toughness (J/m²)	1.5	1.2	0.8	0.7	0.6	2.4	2.7	2.8	3.1	7.3
Notched izod impact strength (J/M)	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]	—[a]
T_g (°C)	8.9	—	6.0	—	5.2	10.5	—	12.8	—	16.2

[a] Nonbreak.

phatic amine are influenced by the following factors: (1) the PC content, (2) the molecular weight of aliphatic amine, and (3) the stage of curing. During the primary cure stage, the transamidation reaction converts nearly all the carbonate and fraction of amine into N-aliphatic aromatic carbamate and urea. The transamidation reaction causes the equivalent ratio of oxirane to amine less than stoichiometric and results in lower crosslinking density, modulus, and T_g of the blend. The substitution reaction between N-aliphatic aromatic carbamate and urea occurs during postcuring and yields the N-aliphatic aliphatic carbamate, leading to more a tightly crosslinked network. Higher crosslinking density results in higher T_g, tensile strength, and tensile modulus of the postcured product than that of the products after primary and secondary curing. The impact toughness of the PC–epoxy/aliphatic amine blend does not increase significantly with the presence of PC. The tensile toughness of certain blends have extremely higher tensile toughness because their T_gs are close or below room temperature.

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