Interpenetrating Polymer Networks of Bismaleimide and Polyether Polyurethane–Crosslinked Epoxy

J. L. HAN,[1] K. Y. LI[2]

[1] Department of Chemical Engineering, National I-Lan Institute of Agriculture and Technology, I-Lan, Taiwan

[2] Department of Chemical Engineering, National Taiwan University, Taipei, Taiwan

ABSTRACT: In this work, we prepared the interpenetrating polymer networks of bismaleimide and polyether-type polyurethane(polyoxypropylene)–crosslinked epoxy (BMI/PU(PPG)–EP IPNs) by employing the simultaneous bulk polymerization technique. The polyurethane (PU)–crosslinked epoxy was identified via infrared (IR) spectra analysis. Also investigated herein were the mechanical properties, including tensile strength, Izod impact strength, and fracture energy (G_{IC}) of the IPNs with various BMI contents in PU–crosslinked epoxy matrix. In addition, differential scanning calorimetry (DSC) analysis and the thermogravimetric analysis (TGA) were performed to examine the thermal properties of the BMI/PU(PPG)–EP IPNs. In addition, morphology and dynamic mechanical analysis (DMA) of the BMI/PU(PPG)–EP IPNs were also studied. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 2635–2645, 1998

Key words: bismaleimide; polyether polyurethane–crosslinked epoxy; interpenetrating polymer network; morphology; thermal property

INTRODUCTION

Epoxy resins play a vital role in manufacturing composite materials because of their rather high glass transition temperatures, T_g, which results in the stiffness and brittleness by nature of the composites.[1] Therefore, improving the toughness of the epoxy composite has received extensive attention.[1–3] Our previous works,[2,3] have improved the mechanical properties by synthesizing a series of polyurethane (PU)–crosslinked epoxy resins. According to those investigations, the cured PU–crosslinked epoxy by tertiary amine exhibits desirable processibility, excellent mechanical property, high Izod impact strength, and high fracture energy (G_{IC} value). The cured 4,4′-bis-(maleimidiphenylmethane) (BMI) possesses excellent thermal stability[4–6] with no evolution of small molecules during the polymerization.[7–10] However, most cured BMIs have the same limitations as epoxy resins, including poor processibility and brittleness due to high crosslinking density. In addition, several investigations in the recent decade have modified BMI to improve its physical properties and processibility.[11–16]

The BMIs are polymerized through the addition reaction of the double bonds in the maleimide group. However, the polyether-type polyurethane(polyoxypropylene)–crosslinked epoxy [PU(PPG)–crosslinked epoxy] cured by tertiary amine is a ring-opening reaction.[17–19] In this study, we mix the BMI resin with PU(PPG)–crosslinked epoxy resin and then simultaneously polymerize it to form interpenetrating polymer networks (IPNs). The stress–strain property, fracture energy (G_{IC}), and Izod impact

property of the interpenetrating polymer networks of bismaleimide and polyether-type polyurethane(polyoxypropylene)–crosslinked epoxy [BMI/PU(PPG)–EP IPNs] were investigated. In addition, the thermal properties, compatibility, and dynamic mechanical behavior of the BMI/PU(PPG)–EP IPNs are also studied.

EXPERIMENTAL

Table I lists the materials used herein. Polyols (PPG400, PPG1000, and PPG2000) and the diglycidyl ether of bisphenol A epoxy resin (DGEBA) were heated at 60°C and degassed by vacuum overnight prior to use.

Preparation of Polyurethane Prepolymers

Polyurethane prepolymers were prepared by reacting 4,4′-diphenyl methane diisocyanate (MDI) with polyols. MDI (2 equivalent weight) was initially placed in a reaction kettle and heated to a molten state. Polyols of a proper amount (1 equivalent weight) were then poured into the reaction

kettle. The mixture was vigorously agitated by a mechanical stirrer.

The reaction occurred under dry nitrogen (N_2) atmosphere at a temperature of around 68°C. Next, the isocyanate content of the reaction mixture was determined using di-n-butyl amine titration methods.[20] The reaction was curtailed when the isocyanate content reached the theoretical value.

Preparation of Polyurethane-Crosslinked Epoxy

The preparation of PU–crosslinked epoxy was as follows.[2,3] Diglycidyl ether of bisphenol A epoxy (DGEBA) was poured into a reaction kettle. Dried nitrogen gas was blown into the reaction kettle to remove air and moisture in the reaction kettle. A predetermined amount of PU prepolymer (5, 10, and 15 wt %) was then poured into the kettle to mix with the DGEBA. The temperature was maintained at about 68°C. During the reaction, a sample was periodically taken to detect the —NCO group at 2270 cm^{-1} in the infrared (IR) spectra. The reaction was curtailed until the —NCO adsorption peak disappeared. The reaction is according to the following reaction:

Diglycidyl ether of Bisphenol A (DGEBA)

PU-crosslinked epoxy(DGEBA) resin

Table I   Materials

Designation	Description	Source
PPG400	Poly(oxypropylene) glycol, MW = 400	ACRO Chem. Taiwan Co., Taiwan
PPG1000	Poly(oxypropylene) glycol, MW = 1000	ACRO Chem. Taiwan Co., Taiwan
PPG2000	Poly(oxypropylene) glycol, MW = 2000	ACRO Chem. Taiwan Co., Taiwan
MDI	4,4′-diphenyl methane diisocyanate, eq wt = 125	BASF Wyandotte Co., Switzerland
DGEBA	Diglycidyl ether of bisphenol A, EEW = 186, $n \doteq 0.11267$	Dow-Chemical Co., USA
TDMP	2,4,6-tri(dimethyl aminomethyl) phenol	Ciba-Geigy Co., Switzerland
DCPO	Dicumyl peroxide	Merk Co., Germany
BMI	4,4′-bis(maleimide-diphenylmethane)	Merk Co., Germany

where R denotes the urethane-terminated polyether diols (PPG). During this period, the absorption peak of the epoxide group (916 cm^{-1}) did not change.

Preparation of Bismaleimide Monomers

The 4,4′-bismaleimidodiphenyl methane (BMI) monomer was prepared according to the procedure described elsewhere.[15,16] The reaction is according to the following scheme:

maleic anhydride (MA)   diamine

amic acid

Preparation of Interpenetrating Polymer Networks of Bismaleimide and Polyurethane (Polyoxypropylene)–Crosslinked Epoxy

An adequate amount of the above PU(PPG)–crosslinked epoxy and various contents of BMI were placed in a cup, into which 2.9 wt % (based on epoxy weight) of curing agent (TDMP) and 0.5 wt % (based on BMI weight) of initiator (dicume peroxide, DCPO) were added. They were vigorously stirred and drafted for several minutes to eliminate all the bubbles produced during agitation. After removing the bubbles, the reaction

mixture was poured into an aluminum mold and pressed at 120°C for 1 h. The sample was then postcured at 180°C for 2 h. Finally, the sample was removed from the mold and placed in a desiccator, where the relative humidity was maintained at 50% for 3 days before testing.

Testing Methods

Infrared spectra were made using a Hitachi 270-30 infrared spectrophotometer. The sample was directly applied by dabbing onto a KBr pellet

to detect the —NCO group content in the reaction mixture.

The stress–strain properties were determined by using a Tensilon (Mode: TCF-RC, Yashima Works Ltd., Japan) test unit. The test procedure followed ASTM-D638, with a crosshead speed of 1 cm/min.

The Izod impact strength of BMI/PU(PPG)–EP IPNs was measured according to the procedure described in ASTM 256.

The fracture energy (G_{IC}) was measured using compact-tension specimens (CTS).[21] The G_{IC} value can be calculated by the following equation:

$$G_{IC} = \frac{Y^2 \left(\dfrac{a}{w}\right) P^2 a}{EW^2 b^2}$$

where

$$Y(\tfrac{a}{w}) = 29.6 - 186(\tfrac{a}{w}) + 656(\tfrac{a}{w})^2$$

$$- 1017(\tfrac{a}{w})^3 + 639(\tfrac{a}{w})^4$$

Where a, b, and E denote the crack length, thickness, and modulus of the specimen, respectively, and P represents the load on the specimen. A sharp precrack was made with a razor blade before the test. The specimens were tested on a screw-driven Tensilon machine at a crosshead speed of 0.1 cm/min.

Differential scanning calorimetry (DSC) analysis was performed in the temperature range of 50–300°C. The analysis was performed on a DuPont 2200 at a heating rate of 20°C/min.

Thermogravimetric analysis (TGA) was undertaken on a DuPont 9900-951 TGA instrument under nitrogen atmosphere and was measured at a heating rate of 20°C/min. The weight of sample tested was around 8 mg.

Dynamic mechanical analysis (DMA) was performed using DuPont 983 DMA unit with the operating temperature ranging from −100 to 250°C. The size of the samples used was approximately $6 \times 1 \times 0.2$ cm.

Morphological studies were performed by using scanning electron microscopy (SEM). Microphotographs were taken on the surface, which was made by fracturing the specimen in liquid nitrogen and then coating it with gold powder.

Figure 1　IR spectra during reaction of PU(PPG400) prepolymer with DGEBA resin: (—) beginning of the reaction; (- - - -) end of the reaction.

RESULTS AND DISCUSSION

Infrared Spectra

As Figure 1 depicts, the PU(PPG) prepolymer can react with the pendant secondary hydroxyl group of DGEBA. Upon introducing the PU(PPG) prepolymer into the DGEBA, the absorption peak of —NCO group at 2270 cm^{-1} gradually reduced in intensity with reaction time. Finally, the peak disappeared after several hours. This occurrence phenomenon confirms the completion of the crosslinking reaction between the —NCO group of the PU prepolymer and the pendent secondary hydroxyl group of DGEBA.

Stress–Strain

Table II lists the mechanical properties of the IPNs of bismaleimide and polyether-type polyurethane (polyoxypropylene)–crosslinked epoxy (BMI/PU(PPG)–EP IPN) with various BMI contents. Figure 2 displays the tensile strength of the BMI/PU(PPG)–EP IPNs with various BMI content and 15 wt % PU(PPG) prepolymer content. For the shorter segment of polyurethane system, including PU(PPG400) and PU(PPG1000) in the BMI/PU(PPG)–EP IPN, regardless of the amount of PU(PPG) used, the tensile strength of the BMI/PU(PPG)–EP IPN increased while adding BMI content to a maximum value and then gradually decreased with a further increase of BMI content (Table II and Fig. 2). Such behavior is probably attributed to the partial compatibility between the BMI and the PU(PPG)–crosslinked epoxy resin. Restated, the intermolecular interpenetration be-

Table II Mechanical Properties of BMI/PU(PPG)–EP IPN

IPN	BMI Content (%)	Tensile Strength (MPa)	Izod Impact (J/m)	Fracture Energy (KJ/m^2)
BMI/PU(PPG400, 5%)–EP	5	87.5	20.6	18.7
BMI/PU(PPG400, 5%)–EP	10	63.9	18.6	6.7
BMI/PU(PPG400, 5%)–EP	15	67.1	22.3	3.1
BMI/PU(PPG400, 10%)–EP	5	82.0	19.6	7.8
BMI/PU(PPG400, 10%)–EP	10	61.3	10.6	4.8
BMI/PU(PPG400, 10%)–EP	15	27.6	6.7	5.3
BMI/PU(PPG400, 15%)–EP	5	75.9	16.2	5.7
BMI/PU(PPG400, 15%)–EP	10	69.5	10.1	5.2
BMI/PU(PPG400, 15%)–EP	15	64.0	11.8	4.8
BMI/PU(PPG1000, 5%)–EP	5	78.4	47.6	20.0
BMI/PU(PPG1000, 5%)–EP	10	75.6	33.3	14.5
BMI/PU(PPG1000, 5%)–EP	15	56.7	21.4	10.1
BMI/PU(PPG1000, 10%)–EP	5	79.1	38.8	18.6
BMI/PU(PPG1000, 10%)–EP	10	79.3	27.3	16.8
BMI/PU(PPG1000, 10%)–EP	15	72.5	22.9	16.6
BMI/PU(PPG1000, 15%)–EP	5	72.8	32.4	9.8
BMI/PU(PPG1000, 15%)–EP	10	67.8	21.6	7.8
BMI/PU(PPG1000, 15%)–EP	15	66.3	20.1	5.8
BMI/PU(PPG2000, 5%)–EP	5	39.9	23.3	19.0
BMI/PU(PPG2000, 5%)–EP	10	41.0	19.4	6.3
BMI/PU(PPG2000, 5%)–EP	15	36.7	22.1	7.5
BMI/PU(PPG2000, 10%)–EP	5	50.4	33.3	10.9
BMI/PU(PPG2000, 10%)–EP	10	51.4	22.1	11.0
BMI/PU(PPG2000, 10%)–EP	15	62.6	19.7	9.4
BMI/PU(PPG2000, 15%)–EP	5	39.9	20.3	7.0
BMI/PU(PPG2000, 15%)–EP	10	41.0	12.8	3.0
BMI/PU(PPG2000, 15%)–EP	15	36.7	10.2	6.2

tween the BMI and PU(PPG)–crosslinked epoxy resin is obviously formed (that is, the IPN effect). The more intermolecular physical entanglements that form between the BMI and PU(PPG)–crosslinked epoxy networks imply a more significant IPN effect. In addition, the crosslinking density also increased with the IPN formation. Therefore, the tensile strength of BMI/PU(PPG)–EP IPN increased with an increasing BMI content. However, owing to rigid and brittle features of the cured BMI, the rigid and brittle behaviors were observed with a further increase of BMI content in the BMI/ PU(PPG)–EP IPN system. The tensile strength of BMI/PU(PPG)–EP IPN was then gradually decreased. Therefore, the tensile strength of the BMI/PU(PPG)–EP IPN increased up to a maximum value at the BMI content ranging from 5 to 10 wt %. According to Figure 2, the shorter chain length of PU(PPG) in the BMI/ PU(PPG)–EP IPN, which contributed more sig-

Figure 2 Dependence of tensile strength of BMI/ PU(PPG, 15 wt %)–EP IPN on various BMI contents and 15 wt % PU(PPG): (○) BMI/PU(PPG400)–EP IPN; (□) BMI/PU(PPG1000)–EP IPN; (△) BMI/ PU(PPG2000)–EP IPN.

Figure 3 Dependence of Izod impact strength of BMI/PU(PPG, 15 wt %)–EP IPN on various BMI contents and 15 wt % PU(PPG): (○) BMI/PU(PPG400)–EP IPN; (□) BMI/PU(PPG1000)–EP IPN; (△) BMI/PU(PPG2000)–EP IPN.

nificantly to the IPN effect, leads to a tensile strength at a lower BMI content, as shown in Figure 2.

In contrast, for the longer soft segment of PU(PPG) system [that is, PU(PPG2000)] introduced into the BMI/PU(PPG)–EP IPN, the tensile strength of BMI/PU(PPG)–EP IPN decreased with an increasing BMI content for any amounts of PU(PPG2000) introduced (Table II and Fig. 2). In our previous studies,[2,3] the longer chain length of the polyether-type PU [for example, PU(PPG2000), PU(PTMO2000)] introduced into the PU–crosslinked epoxy structure exhibited the phase separation from the epoxy and formed PU rubber particles dispersed in the matrix. This occurrence was attributed to the fact that the BMI introduced was preliminarily dissolved in the PU(PPG2000) domains. The BMI network was nearly formed in the PU(PPG2000) rubber particle domains. Therefore, the intermolecular interpenetration of the BMI network with the crosslinked epoxy network could barely to be observed in the BMI/PU(PPG2000)–EP IPN. Moreover, the tensile strength of BMI/PU-(PPG2000)–EP IPN decreased with an increasing BMI content.

Izod Impact Strength

Table II and Figure 3 summarize the Izod impact strength of the BMI/PU(PPG)–EP IPN with various BMI contents and 15 wt % PU(PPG) prepoly-

mer content. According to those results, the Izod impact strength of the most BMI/PU(PPG, 15 wt %)–EP IPN sample decreased with an increasing BMI content. This behavior is attributed to the rigid and brittle nature of the cured BMI. Thus, the ductility of the BMI/PU(PPG)–EP IPN matrix was not enhanced. In our previous study,[22] we demonstrated that the Izod impact property, which is referred to as a high shear rate fracturing, depends on the toughness of the overall matrix. In this case, the BMI is not as compatible with the epoxy-dominated matrix. Therefore, the Izod impact strength of the most BMI/PU(PPG)–EP IPN samples did not significantly improve. However, when the BMI content is as low as 5 wt %, the BMI/PU(PPG1000, 15 wt %)–EP IPN possesses an excellent Izod impact strength (Fig. 3). This characteristic is attributed to the cured BMI networks at this low level are more compatible and uniformly interpenetrated into the PU(PPG1000)–crosslinked epoxy networks, ultimately resulting in a toughening of the overall matrix toughness. Therefore, the BMI/PU(PPG1000, 15 wt %)–EP IPN at 5 wt % BMI content has excellent Izod impact strength. In addition, the Izod impact strength of the BMI/PU(PPG)–EP IPN with the molecular weight of the soft polyether polyol at 1000 exceeds that of the others (that is, BMI/PU(PPG400, 15 wt %)–EP IPN and BMI/PU(PPG2000, 15 wt %)–EP IPN). This finding reveals that the soft-segment PU(PPG1000) is an adequate chain length for enhancing the Izod impact property (that is, high-shear-rate fracturing) of the BMI/PU(PPG)–EP IPN.

Fracture Energy, G_{IC}

Table II lists the fracture energy (G_{IC} values) of the BMI/PU(PPG)–EP IPN with various BMI contents. As shown in Figure 4, no effect of molecular weight of PU prepolymer is observed. The fracture energy (G_{IC} value) of BMI/PU(PPG, 15 wt %)–EP IPN does not significantly differ with an increase in BMI content. Moreover, this figure reveals that the G_{IC} value of BMI/PU(PPG, 15 wt %)–EP IPN based on the soft-segment PU(PPG) of the molecular weight 1000 (that is, BMI/PU(PPG1000)–EP IPN) is higher than the other 2 IPNs when the PU(PPG) prepolymer content was 15 wt % (that is, BMI/PU(PPG400, 15 wt %)–EP IPN and BMI/PU(PPG2000, 15 wt %)–EP IPN). This toughening behavior corresponds to the result from Izod impact test mentioned above. Con-

Figure 4 Dependence of fracture energy of BMI/PU(PPG, 15 wt %)–EP IPN on various BMI contents and 15 wt % PU(PPG): (○) BMI/PU(PPG400)–EP IPN; (□) BMI/PU(PPG1000)–EP IPN; (△) BMI/PU(PPG2000)–EP IPN.

sequently, the BMI/PU(PPG1000, 15 wt %)–EP IPN exhibited excellent fracture toughness in both the high-shear-rate fracturing and low-shear-rate fracturing tests.

Thermal Properties

Figure 5 depicts the dynamic DSC analysis for the BMI/PU(PPG)–EP IPNs with various BMI contents and 5 wt % PU(PPG1000) prepolymer content. According to this figure, the BMI/PU(PPG1000, 5 wt %)–EP IPN contained a single glass transition temperature (T_g) at 75°C. In ad-

Figure 5 DSC for BMI/PU(PPG1000, 5 wt %)–EP IPN on various BMI contents: (—) 5 wt % BMI; (— · —) 10 wt % BMI; (— · · —) 15 wt % BMI.

Figure 6 Thermogravimetric analysis of BMI/PU(PPG2000, 10 wt %)–EP IPN on various BMI contents: (—) 0 wt % BMI; (— · —) 5 wt % BMI; (— · · —) 10 wt % BMI; (— · · · —) 15 wt % BMI.

dition, the T_g of pure BMI network cannot be found in this study owing to the fact that its T_g exceeds 380°C[23] and the BMI/PU(PPG)–EP IPN is initially decomposed at 350°C. (see TGA diagram, Fig. 6) Therefore, the T_g of BMI network cannot be identified in the BMI/PU(PPG)–EP IPN. Although the cured BMI has a high T_g at or above 380°C, the T_g of BMI/PU(PPG)–EP IPN does not shift to a high temperature with an increasing BMI content. This inactivity can be attributed to the low compatibility between the BMI and the PU(PPG)–crosslinked epoxy resin.

Figure 6 presents the TGA of the BMI/PU(PPG)–EP IPNs with various BMI contents and 10 wt % PU(PPG2000) prepolymer content. This figure indicates that the BMI/PU(PPG2000, 10 wt %)–EP IPN has 2 decomposition temperatures (T_d): a high decomposition temperature (T_{dh}) at 600°C and a low decomposition temperature (T_{dl}) at 350°C. The neat BMI/PU(PPG2000, 10 wt %)–crosslinked epoxy without BMI in the system exhibits a single decomposition temperature (T_d) at 310°C. Therefore, T_{dh} denotes the decomposition temperature of the BMI network in the BMI/PU(PPG2000, 10 wt %)–EP IPN system, and the T_{dl} should be of the BMI/PU(PPG2000, 10 wt %)–crosslinked epoxy dominating network in the BMI/PU(PPG2000, 10 wt %)–EP IPN.

Dynamic Mechanical Analysis

Figures 7–9 illustrate the damping factor, tanδ, of dynamic mechanical analysis for the IPNs of

Figure 7 Temperature dependence of damping factor (tanδ) of BMI/PU(PPG400, 10 wt %)–EP IPN at various BMI contents: (—) 5 wt % BMI; (— · —) 10 wt % BMI; (· · · · · ·) 15 wt % BMI.

bismaleimide and polyether-type polyurethane–crosslinked epoxy (BMI/PU(PPG)–EP IPN). The BMI/PU(PPG)–EP IPNs demonstrated 2 distinct transition temperatures (T_g). The lower temperature transition (T_l) is is approximately at −40 to

Figure 8 Temperature dependence of damping factor (tanδ) of BMI/PU(PPG1000, 10 wt %)–EP IPN at various BMI contents: (—) 5 wt % BMI; (— · —) 10 wt % BMI; (· · · · · ·) 15 wt % BMI.

Figure 9 Temperature dependence of damping factor (tanδ) of BMI/PU(PPG2000, 10 wt %)–EP IPN at various BMI contents: (—) 5 wt % BMI; (— · —) 10 wt % BMI; (· · · · · ·) 15 wt % BMI.

−60°C (see Table III), and the higher one (T_h) is around 110–140°C (see Table III). However, the hard segment motion of the IPN network resulted in the high transition temperature (T_h). Regarding the transition temperature of the BMI network in the BMI/PU(PPG)–EP IPN, it was not found from the DMA because the cured BMI network had a high crosslinking density and a high glass transition temperature (T_g).[23] The T_h of BMI/PU(PPG)–EP IPNs comprised polyether-type PU based on PPG400, PPG1000, or PPG2000, as shown in Figures 7–9. According to those figures, regardless of the molecular weight of the PU(PPG) in the BMI/PU(PPG)–EP IPNs, the T_h peak broadened and decreased in intensity with an increasing BMI content. This phenomenon is attributed to the fact that the cured BMI network has the characteristics of rigidity, high crosslinking density, and high storage modulus E'. Therefore, the BMI/PU(PPG)–EP IPN decreases the damping factor, tanδ (equal to E''/E', where E' is the storage modulus and E'' is the loss modulus), the value of the high transition temperature region (T_h) when the BMI is added. Another similar effect of the molecular weight of the PU(PPG) on the T_h shift was found in the BMI/PU(PPG)–EP IPN. The T_h of the BMI/PU-(PPG)–EP IPN did not shift significantly with an increasing BMI content. This was owing to the poor compatibility between the BMI network and

Table III   Glass Transition Temperatures of BMI/PU(PPG)–EP Interpenetrating Polymer Networks

IPN	BMI Content (%)	T_l [a] (°C)	T_h [a] (°C)
BMI/PU(PPG400, 5%)–EP	5	−41	128
BMI/PU(PPG400, 5%)–EP	10	−39	129
BMI/PU(PPG400, 5%)–EP	15	−37	131
BMI/PU(PPG400, 10%)–EP	5	−37	129
BMI/PU(PPG400, 10%)–EP	10	−42	126
BMI/PU(PPG400, 10%)–EP	15	−47	134
BMI/PU(PPG400, 15%)–EP	5	−42	119
BMI/PU(PPG400, 15%)–EP	10	−42	130
BM//PU(PPG400, 15%)–EP	15	−48	138
BMI/PU(PPG1000, 5%)–EP	5	−52	112
BMI/PU(PPG1000, 5%)–EP	10	−56	116
BMI/PU(PPG1000, 5%)–EP	15	−61	115
BMI/PU(PPG1000, 10%)–EP	5	−51	120
BMI/PU(PPG1000, 10%)–EP	10	−48	127
BMI/PU(PPG1000, 10%)–EP	15	−50	125
BMI/PU(PPG1000, 15%)–EP	5	−44	124
BMI/PU(PPG1000, 15%)–EP	10	−45	127
BMI/PU(PPG1000, 15%)–EP	15	−44	128
BMI/PU(PPG2000, 5%)–EP	5	−40	122
BMI/PU(PPG2000, 5%)–EP	10	−39	127
BMI/PU(PPG2000, 5%)–EP	15	−50	125
BMI/PU(PPG2000, 10%)–EP	5	−43	129
BMI/PU(PPG2000, 10%)–EP	10	−44	128
BMI/PU(PPG2000, 10%)–EP	15	−43	129
BMI/PU(PPG2000, 15%)–EP	5	−46	135
BMI/PU(PPG2000, 15%)–EP	10	−40	133
BMI/PU(PPG2000, 15%)–EP	15	−42	132

[a] Measured by DMA.

PU(PPG)–crosslinked epoxy network (see Fig. 10). Those results correlate with the mechanical behavior of the BMI/PU(PPG)–EP IPN. Table III lists the transition temperatures of the BMI/PU(PPG)–EP IPN. However, T_l did not significantly change despite of the molecular length of polyols in PU(PPG) in BMI/PU(PPG)–EP IPN. In addition, the T_h of the BMI/PU(PPG)–EP IPN did not significantly change as the BMI was added (Table III).

Compatibility and Morphology

Morphology studies were performed using the SEM for the BMI/PU(PPG)–EP IPNs with PU based on polyether-type PU [that is, PU(PPG)]. As Figure 10 indicates, a heterogeneous morphology of the fracture surface is observed for the BMI/PU(PPG)–EP IPN with the epoxy crosslinked with 15 wt % PU(PPG1000) prepolymer.

This finding reveals that the compatibility between the BMI-dissolved PPG domains and the epoxy matrix is not as significant. In addition, as the BMI content increased, the smooth fracture surface was observed due to the brittle nature of the BMI (Fig. 10).

CONCLUSION

According to the results presented herein, the tensile strength of the BMI/PU(PPG)–EP IPN with longer PU(PPG) segment [that is, PU-(PPG2000)] decreased with an increasing BMI content. The Izod impact strength decreased with an increasing BMI content for the majority of BMI/PU(PPG, 15 wt %)–EP IPNs. The fracture energy, G_{IC}, value of the BMI/PU(PPG, 15 wt %)–EP IPNs has no significant difference with an

Figure 10   Microphotographs of BMI/PU(PPG1000, 15 wt %)–EP IPN at various BMI contents: (a) 5 wt % BMI; (b) 10 wt % BMI; (c) 15 wt % BMI.

increase of BMI content. From the thermal analysis, the BMI/PU(PPG1000)–EP IPN presented a single glass transition temperature (based on the DSC test), T_g, without shifting to a high temperature as the BMI content increased. In addition, the following 2 decomposition temperatures were observed from the data: 1 high decomposition temperature at 600°C, and the other low

decomposition temperature at 350°C. The BMI network interpenetrating with the polyether-type polyurethane(polyoxypropylene)–crosslinked epoxy network to BMI/PU(PPG)–EP IPN contains a heterogeneous phase morphology. The damping factor (tanδ) of T_h peak always decreased with an increasing BMI content for the BMI/PU(PPG)–EP IPN. In the less compatible BMI/PU(PPG)–EP IPN system with the PU based on the polyether-type PPG polyols, it was no significant shift with an increasing BMI content.

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