

Novel Method for Preparation of Polyurethane Elastomers with Improved Thermal Stability and Electrical Insulating Properties

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ABSTRACT: A novel method was developed for the preparation of polyurethane with enhanced thermal stability and electrical insulation properties via the reaction of epoxy-terminated polyurethane prepolymer (EPU) and poly(amic acid) (PAA). EPUs were synthesized from the reaction of glycidol with NCO-terminated polyurethane prepolymers, which were prepared from the reaction of polycaprolactone-based polyol (CAPA) of different molecular weights and some commercially available diisocyanates including hexamethylene diisocyanate, toluene diisocyanate, and 4,4'-methylene bis(phenyl isocyanate). PAA was prepared from the reaction of equimolar amounts of pyromellitic dianhydride and oxydianiline. The effects of PAA content, the nature of diisocyanate, and the molecular weight of CAPA on the mechanical,

thermal, thermomechanical, and electrical properties of the final networks were investigated. The crosslink density of the samples was determined according to an equilibrium swelling method using the Flory–Rehner equation and was correlated to the structure of the final polymers. Gel content and activation energy of network formation in the absence and the presence of a tertiary amine catalyst were also studied. The results showed considerable improvement in the thermal, electrical, and mechanical properties compared to those of other common polyurethanes. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 103: 1776–1785, 2007

Key words: polyurethanes; polyimides; thermal properties; elastomers

INTRODUCTION

Polyurethanes are one of the most versatile classes of polymeric materials known today. A unique feature of polyurethanes is that a wide variety of structural changes can be produced with different hydroxyl compounds and isocyanates, leading to a wide spectrum of properties and applications.^{1–4} Despite being able to tailor the properties of polyurethanes to specific requirements, their poor thermal stability is a serious disadvantage. For example, mechanical properties (strength, modulus, etc.) cease being acceptable above 80°C–90°C, and thermal degradation occurs above 200°C.⁵

Chemical modification of polyurethane chains by a heterocyclic imide structure has been an accepted method for improving thermal stability. This is because of the unique properties of polyimides, including extremely high thermal stability, high mechanical strength, low leakage current density, and high dielectric breakdown strength.^{6,7} Several synthetic methodologies have been followed to introduce an imide structure into polyurethanes.^{8–20}

The presence of free isocyanate groups is another problem that some commercial polyurethanes may have. One method of overcoming this problem is to block isocyanate groups with an “active hydrogen” compound. These products show little or no reaction at room temperature.²¹ Some disadvantages of employing block isocyanates are their high temperature and the long time required for deblocking, as well as the difficulty of removing the blocking agents during the process.²² Modifying polyurethanes with glycidol and producing epoxy-terminated polyurethane overcome these disadvantages and provide some benefits, such as elimination of the need for deblocking and removal of the blocking agents, enhancement of the reaction rate because of highly active epoxy groups, and introduction of extra hydroxyl groups (generated from the ring-opening reaction of the epoxy groups) in the final product, which increases the mechanical properties through additional hydrogen bonding in the hard segment of the final polyurethane elastomer.

Polymer coatings on metal have come to play an important role in various technological applications, especially as electrical insulators. These coatings are based on various kinds of polyurethane, poly(ester-imide), poly(amide-imide), polyester, poly(vinyl formal), and so forth.^{23–30} Because of special properties

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like superior Q values (reciprocal of dissipation factor),^{31–33} polyurethanes have found a wide range of applications as insulating materials in electrical industry. However, the low service temperature of polyurethane-based electrical insulator coatings is still a challenging problem.

There have been few reports concerning evaluation of electrical properties of imide-modified polyurethane.^{6,34} In this article we present the results of our study of the electrical, thermal, and mechanical properties of new imide-modified polyurethanes prepared by a novel method. Structure–property relationships were also established for the prepared samples.

EXPERIMENTAL

Materials

Polyester polyols CAPA 225, CAPA 210, and CAPA 200, with molecular weights of 2000, 1000, and 535, respectively, from Interox Chemicals, were dried at 85°C in vacuum for 24 h before use. 1,6-Hexamethylene diisocyanate (HDI) and toluene diisocyanate (TDI, mixture of 2,4- and 2,6-isomers) from Merck

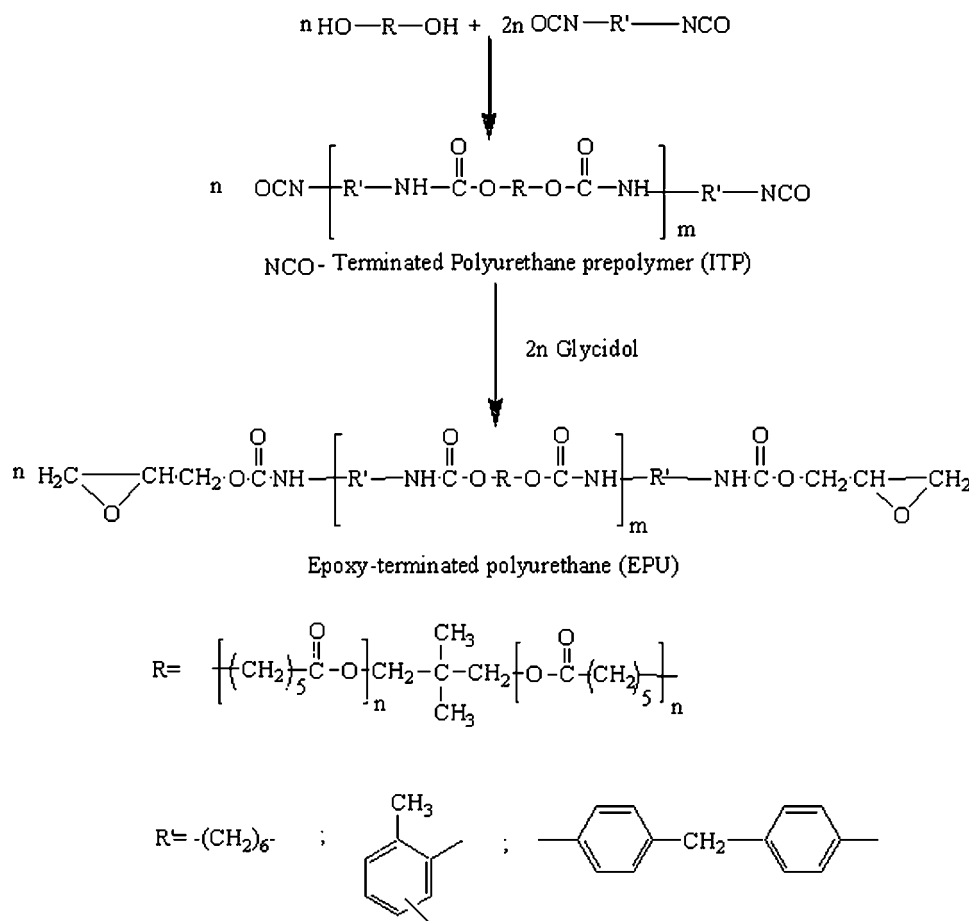
and 4,4'-methylene bis(phenyl isocyanate) (MDI) from Aldrich were vacuum-distilled before use. *N*-methyl pyrrolidone (NMP) was vacuum-distilled from CaH₂ and stored over 4-Å molecular sieves. Pyromellitic dianhydride (PMDA) from Fluka was purified by recrystallization from acetic anhydride. 2,3-Epoxy-1-propanol (glycidol) from Aldrich was vacuum-distilled before use. Other chemicals and solvents were used as received.

Synthesis of epoxy-terminated polyurethane prepolymers

Epoxy-terminated polyurethane prepolymers (EPUs) were prepared as previously described.³⁵

Synthesis of poly(amic acid)

To synthesize poly(amic acid) (PAA), oxydianiline (ODA) (3 mmol, 0.60 g) was poured into NMP (11.3 g) under a nitrogen atmosphere and stirred until a homogeneous solution was obtained. The flask was cooled to 5°C, and PMDA (3 mmol, 0.65 g) was added portionwise. The solution was allowed to warm to room temperature, and stirring was contin-



Scheme 1 Chemical route of synthesis of epoxy-modified polyurethanes (EPUs).

TABLE I
Chemical Composition of Epoxy-Modified Polyurethanes

Code	CAPA (molecular weight)	Diisocyanate	Molar ratio (CAPA : diisocyanate : glycidol)	NCO (%) ITP		Epoxy content (mol epoxy/kg polymer)	
				Theoretical	Experimental	Theoretical	Experimental
EPU1	535	HDI	1 : 2 : 2	9.6	9.2	1.9	1.6
EPU2	1000	HDI	1 : 2 : 2	6.3	6.2	1.3	1.3
EPU3	2000	HDI	1 : 2 : 2	3.6	3.5	0.8	0.8
EPU4	2000	MDI	1 : 2 : 2	3.4	3.3	0.7	0.7
EPU5	2000	TDI	1 : 2 : 2	3.6	3.6	0.8	0.6

ued at room temperature overnight. The solution was refrigerated until use.

Preparation of the blend solution and cast films

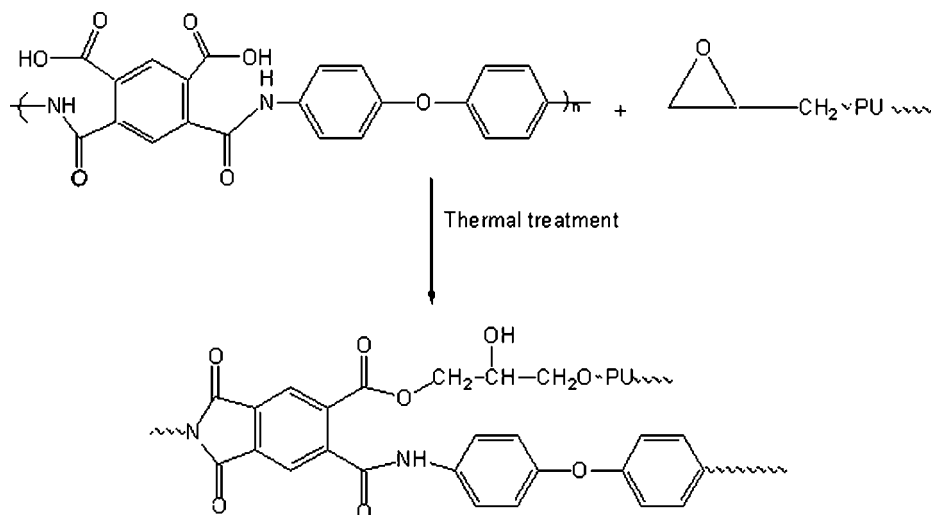
PAA and EPU solutions in NMP were mixed in various weight ratios. The blend solutions were cast on a Teflon mold, and then the solvent was evaporated under vacuum at 60°C for 16 h. The films were thermally treated at 100°C and 200°C, respectively, for 1 h.

Freestanding films were used for subsequent studies.

Testing methods

All measurements were performed by the central laboratory of the Iran Polymer and Petrochemical Institute (Tehran, Iran). FTIR spectra were recorded by a Bruker-IFS 48 instrument. Thermal gravimetric analysis (TGA) was performed on a Stanton STA-780 with a heating rate of 10°C/min in N₂. Tensile properties were measured with a MTS tensile tester model 10/M at a strain rate of 5 cm/min. Measurements were performed at 25°C with a film thickness of about 1 mm and stamped out with an ASTM D 412 die. Dynamic mechanical testing was carried out on a UK Polymer Lab dynamic mechanical thermal

analyzer model MK-II over a temperature range of -100°C-200°C at a heating rate of 10°C/min and a frequency of 1 Hz. The dimensions of the samples were 30 × 10 × 1 mm³. Evaluation of breakdown voltage was performed on a high-voltage kit with an increasing 2 kV/S voltage rate, a spherical electrode 5 cm in diameter, and a frequency of 50 Hz. Dissipation factor and dielectric constant measurements were performed on a Schering bridge device, Tetxag 2801 ZQ. Samples were cut into circular-shaped pieces 5 cm in diameter. The measurements were performed according to ASTM 149-94 and ASTM 150-94 in an air atmosphere at 25°C. For all electrical property measurements, samples with a thickness of 0.5 mm were used. Gel content measurements were performed in a Soxhlet extractor using tetrahydrofuran as the solvent. NCO content of the polyurethane prepolymers was determined according to the procedure detailed in ASTM D 2572, and their epoxy content (moles of epoxy per kilogram of polymer) was determined using a previously described method.³⁶ Density of the polymers was measured according to the method outlined in ASTM D 1817. Solubility (δ), crosslink density (ν_c), and average molecular weight between two crosslinks (M_c) of the polymers were determined accord-



Scheme 2 Synthetic route of imide modified polyurethane (a representative part of final crosslinked product).

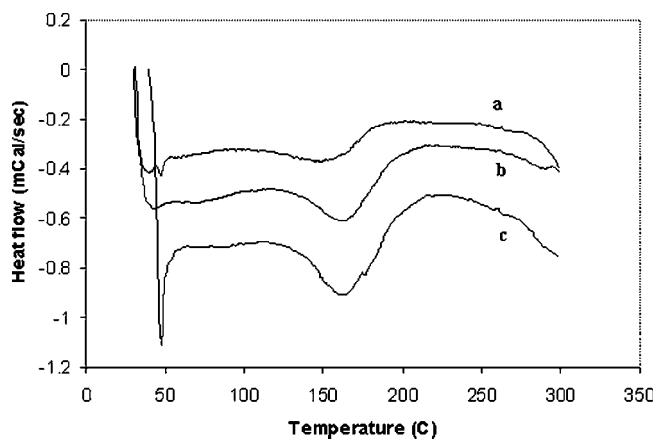


Figure 1 DSC of curing reaction of EPU2/PAA (80 : 20) at different heating rates: (a) 5°C/min, (b) 10°C/min, and (c) 15°C/min.

ing to the equilibrium swelling method based on the Flory–Rehner equation as described previously.³⁷ Curing reactivity was examined by differential scanning calorimetry (Polymer Lab, UK) at heating rates of 5°C/min, 10°C/min, and 15°C/min. Prior to examination EPU, PAA, solvent, and catalyst were mixed fully, and most of the solvent was evaporated at 60°C under vacuum. About 10 mg of the mixture was used for each test.

RESULTS AND DISCUSSION

Synthesis of epoxy-modified polyurethane and poly(amic acid)

Scheme 1 depicts the general method for synthesis of functional polyurethane prepolymers with an average of two epoxy groups (EPU). The reaction of CAPA polyols of different molecular weights with excess diisocyanate resulted in NCO-terminated polyurethane prepolymers, which subsequently were reacted with glycidol to prepare the EPU. The average equivalent weight of the epoxy in an EPU depended on the molecular weight of the prepolymer. Polycaprolactone is a noncrosslinkable segment in the EPU backbone. So increasing the molecular weight of EPU and the subsequent increase in average epoxy equivalent weight reduced the concentra-

tion of epoxy groups in the prepolymers, which were the main crosslinking sites. The different EPU formulations are given in Table I. Spectroscopic assignment of the EPU and evaluation of their epoxy content were reported previously.³⁵

The well-known reaction of PMDA and ODA was used to synthesize the necessary poly(amic acid) oligomer.¹⁸ The spectroscopic data of prepared PAA were in accordance with those reported in the literature.¹⁸

Evaluation of curing reactions by differential scanning calorimetry

PAA was used as the curing agent for the EPU. For the curing reactions solutions of different EPU and PAA in NMP were prepared. Clear and transparent solutions became opaque films after heat treatment, except EPU1-based formulations, which remained transparent.

A crosslinked network was expected to form from the reaction of the COOH groups of PAA and the epoxy groups of EPU upon heat treatment of its components (Scheme 2). This reaction was monitored by differential scanning calorimetry (DSC). As the different formulations designed had similar structures and the reactive groups were the same, DSC analysis was performed only for one formulation. Figure 1 shows the DSC curves for the curing reactions of EPU2 and PAA at different heating rates. The onset temperature of the exothermic reaction of the COOH groups of PAA and the epoxy groups of EPU was in the range of 148°C–166°C depending on the heating rate. To eliminate complex side reactions, which would increase the possibility of ester group formation, and to accelerate the reaction rate, it is customary with organic acid cures of epoxy resins to employ a tertiary amine-based catalyst.³⁸ So DSC analysis was also performed with triethylamine [2% (w/w)] as the catalyst. Table II shows the DSC peak temperatures for the two systems at different heating rates.

A theory by Kissinger³⁹ enables determination of apparent activation energy. This theory shows that the relationship between DSC peak temperature, T_p (K) and heating rate, V_T , is: $\ln V_T = \ln A - E_a/RT_p$

TABLE II
Effect of Added Catalyst on Curing Kinetics

Formulation	Heating rate (°C/min)	DSC peak temperature (°C)	Activation energy (kJ/mol)
EPU2 + PAA (80 : 20 w/w)	5	202	90.63
	10	219	
	15	225	
EPU2 + PAA (80 : 20 w/w) + triethylamine (2 wt % in respect to solid content)	5	185	81.53
	10	200	
	15	210	

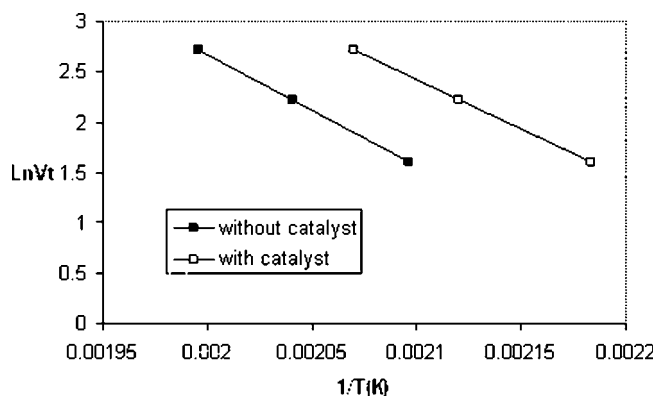


Figure 2 Dependence of $\ln V_t$ on $1/T$ for the curing reactions of the catalyzed and uncatalyzed systems.

where R is the gas constant, A is the frequency factor and E_a is the apparent activation energy. A plot of $\ln V_T$ against $1/T_p$ should be linear, with a slope equal to $-E_a/R$. The data of two systems were plotted, as shown in Figure 2, and good linearity was obtained. The kinetic data derived from these plots are also given in Table II, from which it can be seen that the curing activity of the catalyzed system was

enhanced and that the apparent activation energy of the curing reaction decreased from 90.63 kJ/mol for the uncatalyzed system to 81.53 kJ/mol. This might be related to the formation of a network structure with ester linkages through the reaction of the acid and epoxy groups of the components in the presence of triethylamine.

Spectroscopic characterization

The crosslinked freestanding films obtained from the reaction of EPU and PAA were subjected to ATR-FTIR spectroscopy. A representative example is shown in Figure 3, as well as the FTIR spectra of EPU and PAA for comparison. The FTIR spectra of the cured samples showed a broad peak at about 3330 cm^{-1} because of the NH bonds in the polymer backbone. A asymmetric stretching band of imide carbonyl groups appeared at 1772 cm^{-1} . A strong and broad peak at $1650\text{--}1740\text{ cm}^{-1}$ was related to the symmetrical stretching of the carbonyl groups of the urethane, ester, amide, and imide groups of the polymers. A combination of C–N stretching and N–H out-of-plan bending appeared as a peak at

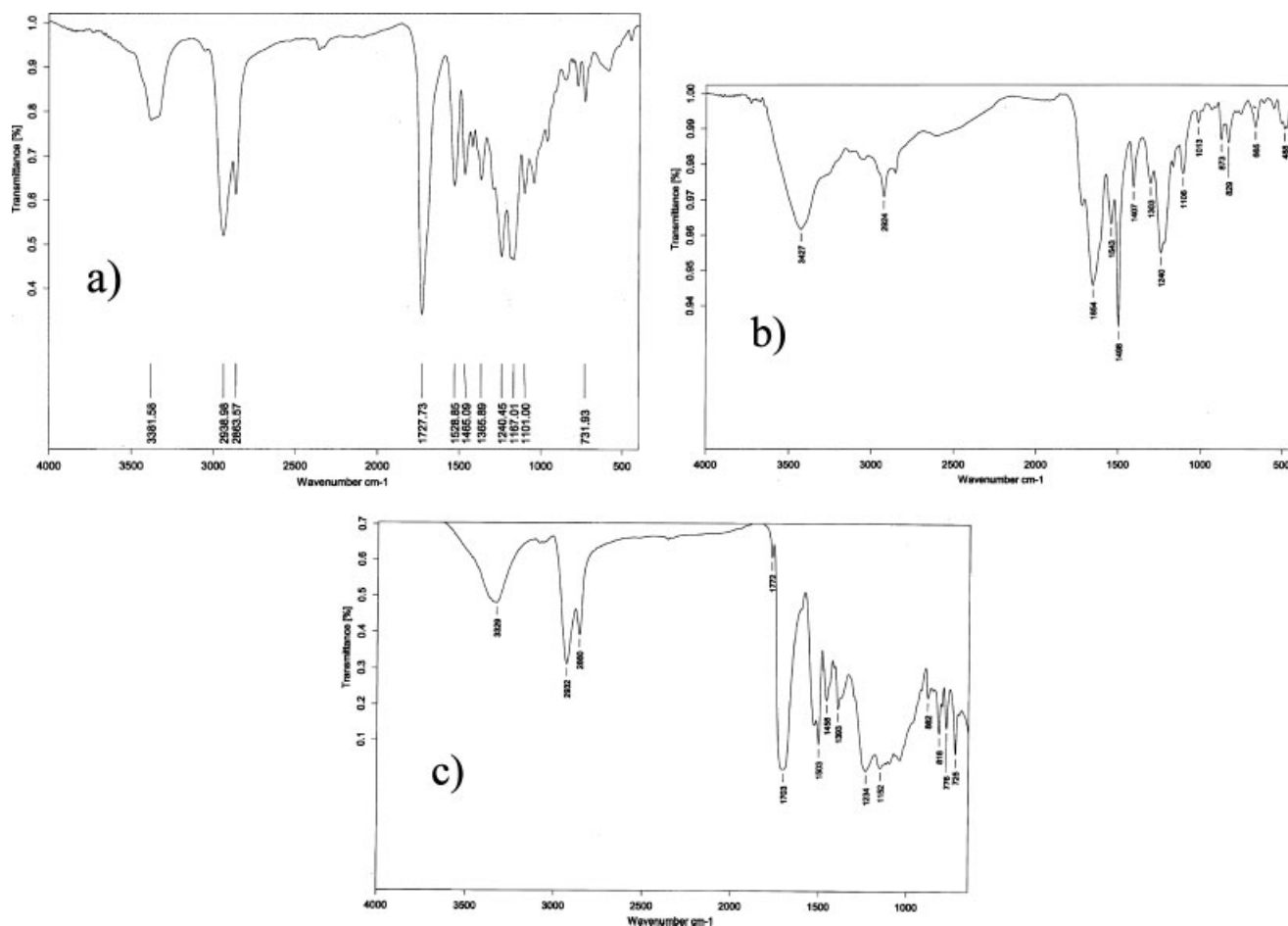


Figure 3 FTIR spectra of (a) EPU1, (b) PAA, and (c) PUI2.

TABLE III
Formulations of Final Cured Films

Code	EPU type	EPU (wt %)	PAA (wt %)	Gel content (%)
PUI1	EPU1	95	5	90
PUI2	EPU1	80	20	87
PUI3	EPU2	95	5	94
PUI4	EPU2	80	20	86
PUI5	EPU3	95	5	93
PUI6	EPU3	80	20	85
PUI7	EPU4	95	5	97
PUI8	EPU5	95	5	92

^a All formulations contained 2% (w/w) triethylamine.

about 1530 cm^{-1} . Imide ring deformation was observed at 725 cm^{-1} . The stretching vibration of the esteric C—O groups appeared as a doublet at 1038 and 1234 cm^{-1} . The presence of peaks because of the imide structure and the considerable reduction in the intensity of the peak of the epoxy groups, at $910\text{--}965\text{ cm}^{-1}$, were the main indications of the formation of a polyurethane insulator (PUI) structure.

Evaluation of network formation

Table III shows the formulations of the different samples obtained from heat treatment of EPUs with different epoxy contents and various weight ratios of PAA (up to 200°C based on DSC data). The upper limit for the amount of PAA in the formulations was the flexibility of the cured films. Although it was desired to increase the amount of PAA that could influence the thermal stability of the final samples, using more than 20 wt % PAA resulted in brittle films. The measured gel content of each sample is also presented in Table III. All the samples had a gel content of more than 85%, an indication of the effectiveness of PAA in the curing of EPUs. However, films with higher amounts of PAA showed a lower gel content. This can be related to the immiscibility of the two components at a higher percentage of PAA.

To gain a better perspective on the curing reaction, the crosslink density and molecular weight between crosslinks were determined for the prepared samples. The maximum position in the plot of the equilibrium degree of swelling (Q) versus solubility parameter of different solvents was used for determination of the solubility parameter of the PUIs. A representative example is shown in Figure 4. M_C and ν_C values were determined according to this plot, other necessary physical data, and related equations (Table IV). The data showed that the ν_C values of the samples were a function of the epoxy content of the EPUs and the weight percent of PAA. As terminal epoxy groups were the sole crosslinking sites, decreasing epoxy content by increasing CAPA polyol

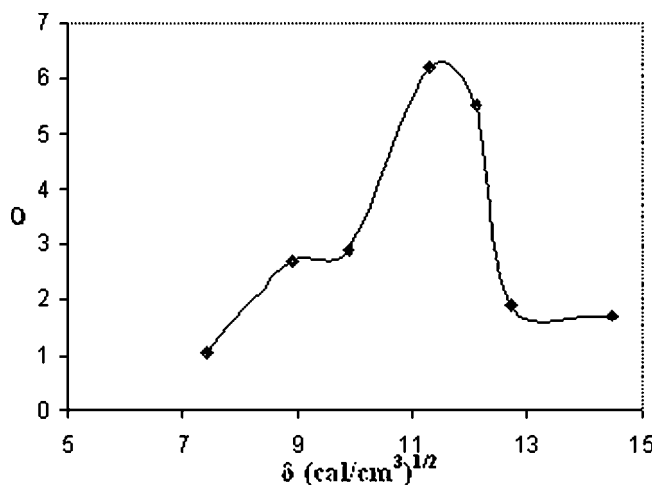


Figure 4 Plot of Q versus δ of different solvents for PUI3.

molecular weight led to a decrease in the ν_C ; consequently, M_C increased as well. Because the molar concentration of the acid groups in PAA was lower than that of the epoxy groups in the EPUs, increasing the amount of PAA caused an increase in the concentration of the available acid groups, resulting in films with a higher crosslink density. The crosslink densities of PUI7 and PUI8 were higher than that of PUI5, which may have been a result of the better compatibility of PAA with EPUs based on aromatic diisocyanate, which can increase the effectiveness of the interactions between COOH and the epoxy groups.

Mechanical properties

Mechanical properties of the PUIs—tensile strength, elongation at break, and modulus—were evaluated from stress–strain curves (Fig. 5 and Table V). The synthesized polymers showed widespread mechanical properties depending on imide content, type of diisocyanate, and the crystallinity that arose from the CAPA polyol with a molecular weight of 2000. Samples PUI1–PUI4, based on CAPA polyol with molecular weights of 535 and 1000, which are com-

TABLE IV
 θ_c and M_c Data of Samples

Code	Density (g/cm^3)	M_c (g/mol)	ν_c (mol/cm^3) $\times 10^3$
PUI1	1.196	390	3.06
PUI2	1.228	384	3.20
PUI3	1.157	747	1.55
PUI4	1.171	695	1.68
PUI5	1.153	1179	0.98
PUI6	1.168	930	1.26
PUI7	1.137	1079	1.05
PUI8	1.145	862	1.33

ν_c , Crosslink density.

M_c , Average molecular weight between two crosslinks.

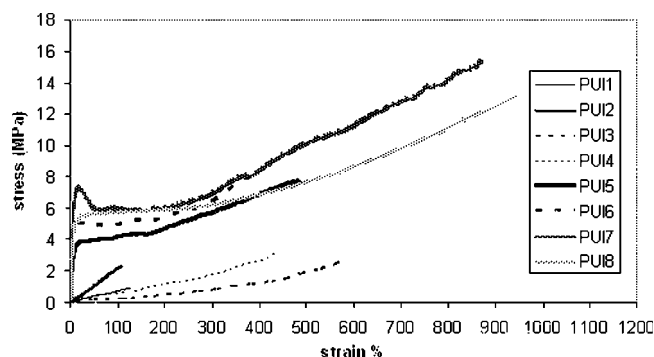


Figure 5 Stress-strain curves for prepared samples.

pletely amorphous, showed the behavior of weak elastomers and display a smooth transition in their stress-strain behavior from the elastic to plastic deformation region. In these samples as the PAA content increased, the modulus and tensile strength increased because of induction of a rigid imide structure in the polymer network as well as enhancement of crosslink density. With the molecular weight of CAPA polyol increased to 2000 (PUI5 and PUI6), the stress-strain behavior of the samples changed, displaying the behavior of semicrystalline elastomers with a pronounced yield point, followed by necking and drawing up to rupture. Also, elongation at break of these samples decreased and initial modulus and tensile strength increased with an increase in PAA content. Using rigid symmetric aromatic diisocyanate (MDI) instead of flexible aliphatic diisocyanate (HDI) in the backbone of the polymer (PUI7) enhanced all the mechanical properties because of more ordered segments and better packing of the polymeric chains. The mechanical properties of the aromatic TDI-based polymer (PUI8) improved compared to those of sample PUI5 as a result of the good compatibility of components and the improved intermolecular interaction. However the stress-strain curve of this sample did not show a pronounced yield point, which is an indication of noncrystalline nature of this elastomer. The asymmetric nature of TDI was apparently responsible for this behavior.

TABLE V
Data on Mechanical Properties

Code	Tensile strength (MPa)	Initial modulus (MPa)	Elongation at break (%)
PUI1	1.0	9.0	127.0
PUI2	2.3	14.3	107.1
PUI3	2.8	2.0	579.0
PUI4	3.2	6.6	437.2
PUI5	7.8	75.6	481.1
PUI6	8.4	125.4	397.0
PUI7	15.3	173.8	869.3
PUI8	13.2	154.2	943.1

TABLE VI
Data from Dynamic Mechanical Analysis

Code	Tan δ^a	T_g^b	T_m
PUI1	0.465	-25	—
PUI2	0.411	-19	—
PUI3	0.433	-35	—
PUI4	0.406	-30	—
PUI5	0.131	-31	38
PUI6	0.122	-30	40
PUI7	0.148	6	45
PUI8	0.599	-23	—

^a Altitude of tan δ peaks.

^b Maxima of tan δ peaks were used for determination of T_g .

Viscoelastic properties

The viscoelastic properties of the prepared polymers were analyzed by DMTA (Table VI). Figures 6 and 7 show the tan δ and storage modulus of the prepared films. Samples PUI1–PUI4 exhibited a one-phase structure, as shown by only one relaxation in the region from -19°C to -35°C , attributed to an α -type transition (T_g). Samples PUI5–PUI7 showed two thermal transitions in the range from -31°C to 6°C , attributed to an α -type transition as well as to another transition in the region of 37°C – 41°C because the melting of the crystalline region arose from CAPA polyol. Also, sample PUI8, based on TDI, showed a one-phase structure. Though generally compatible blends and copolymers have a T_g intermediate between those of the homopolymers, the T_g values of the prepared crosslinked PUIs are characteristic of polyol soft segment. This may be attributed to the microphase separation of the soft (CAPA polyol) and hard (urethane and PAA moieties) segments in the polymers and a high weight percent of soft segments. For samples PUI1–PUI4, which had increased crosslink density and imide content, the α -type transition shifted to a higher temperature. The same trend was observed for the damping of these

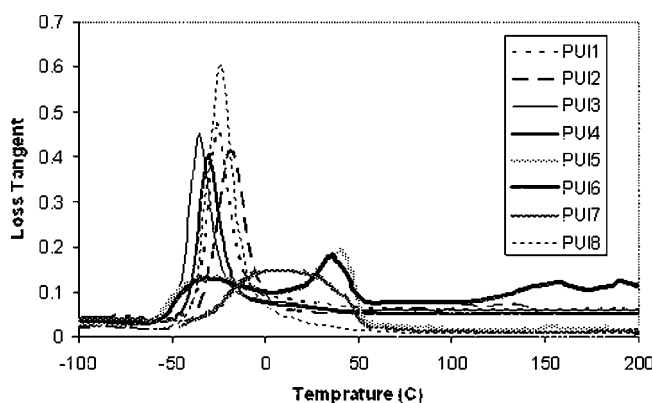


Figure 6 Plot of tan δ versus temperature for prepared samples.

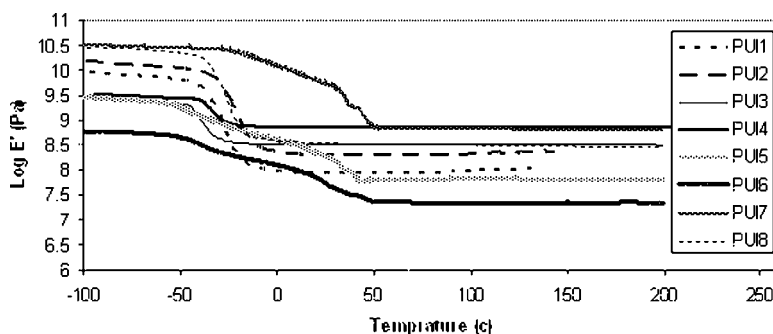


Figure 7 Plot of storage modulus versus temperature for prepared samples.

samples. This was because the viscous flow of the molecular chains was inhibited. Increasing crosslinking and imide content inhibited damping because of the increased chain entanglement, interpinning, and domain cohesion that arose from dipole–dipole interactions of the imide structures. For samples PUI5 and PUI6, which had smaller crosslink densities, the α -type transition shifted to a lower temperature. Samples PUI7 and PUI8, based on aromatic diisocyanate, were observed to behave in two different ways. A more ordered, symmetrical structure of MDI allowed better packing of chains, which led to a considerable increase in the α -type transition and crystalline melting point of the polymer as a result of a decrease in the degree of freedom of the segmental mobility of the polymer backbone. However, the asymmetric structure of TDI reduced the possibility of the chains packing and inhibited the formation of a crystalline domain, so only one transition (α -type transition) was observed.

Thermal stability

The thermal stability of the prepared polymers was evaluated by the TGA method. The results are shown in Table VII and Figure 8. Thermal decomposition of these polymers involved three overlapping

TABLE VII
Data on Thermal Stability and Thermal Behavior

Code	T_0 (°C)	T_{10} (°C)	T_{max} (°C)	CH.Y (%)
PUI1	270	320	360	6
PUI2	275	330	375	12
PUI3	275	330	360	6
PUI4	288	338	373	8
PUI5	287	325	362	4
PUI6	296	340	378	8
PUI7	285	354	420	6
PUI8	280	335	395	4
PU ^a	202	250	315	0

T_0 , initial decomposition temperature.

T_{10} , temperature at 10% weight loss.

T_{max} , maximum decomposition temperature.

CH.Y, weight of remaining polymer at 600°C.

^a Prepared from CAPA225/2TDI/1,4-buthane diol.

steps that were difficult to distinguish from the TGA curves but were more obvious from the DTGA curves. The polymers started to decrease in weight at about 280°C, which was related to simple depolymerization of urethane bonds, and the decrease intensified at about 335°C ($T_{10\%}$). The higher initial thermal stability found in the comparison of conventional polyurethanes may have been a result of the shielding effect of the imide structures. The maximum decomposition temperature of prepared polymers was observed in the range of 360°C–420°C. The decomposition of esteric groups of polyol and aromatic groups from PAA occurred in this region. Table VII shows that $T_{10\%}$ and T_{max} increased as EPU molecular weight increased because of the reduced number of labile urethane bonds and the increased concentration of the more thermally stable esteric bonds. The same trend of enhancement of thermal stability was observed for samples with a greater number of more thermally stable heterocyclic imide structures.

Electrical Properties

To evaluate the electrical properties of prepared PUIs, three main parameters were measured: dissipation factor, dielectric constant, and dielectric breakdown strength. The results are shown in Table VIII. In all electrical applications, it is desirable to

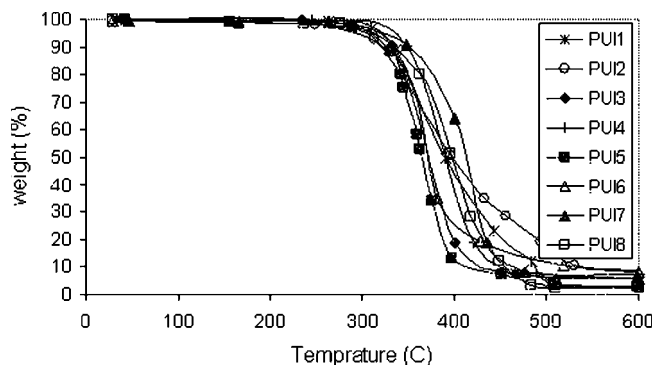


Figure 8 Typical TGA curves for some of the samples.

TABLE VIII
Data on Electrical Properties

Code	Thickness (mm)	Dissipation factor	Dielectric constant	Dielectric breakdown strength (kV/mm)
PUI1	0.5	8.8×10^{-2}	8.5	21.0
PUI2	0.5	7.2×10^{-2}	8.4	22.3
PUI3	0.5	1.3×10^{-3}	7.2	24.6
PUI4	0.5	8.5×10^{-4}	6.6	25.5
PUI5	0.5	9.1×10^{-5}	5.0	30.0
PUI6	0.5	8.3×10^{-5}	4.8	32
PUI7	0.5	1.2×10^{-4}	5.3	33
PUI8	0.5	9.6×10^{-4}	5.2	32

keep electrical loss to a minimum. Electrical loss indicates inefficiency of an insulator. Dissipation factor (DF) is a measure of such electrical inefficiency of an insulating material. In fact, a low DF indicates minimal conversion of electric energy to heat. Table VIII shows the DF values of the prepared polymers were in the range of 8.3×10^{-5} – 8.8×10^{-2} . The DF decreased with an increase in polyol molecular weight because of reduction of the polar character of the whole system. Also by increasing the crosslink density of samples with a higher PAA content, DF was decreased because the reactive polar groups were either neutralized or hindered by crosslinking.^{40,41} The dielectric constant indicates the ability of an insulator to store electrical energy. Electrical signals will lose less of their intensity in a dielectric medium with a low dielectric constant. So it is desirable to have the insulating material have the most minimal capacitance possible. The dielectric constant of the prepared samples was in the range of 5–8.5, which is superior to that of common polyurethane insulators.⁴² Dielectric strength indicates the electrical strength of a material as an insulator. It is an important parameter for selecting an appropriate electrical insulation material to avoid short circuiting, especially in high-temperature operations. The higher the dielectric breakdown strength, the better is the quality of an insulator. The measured values (21–33 kV/mm) indicated the prepared PUI had excellent insulating power. For comparison, electrical porcelains typically have a dielectric breakdown strength of 25 kV/mm, and sol-gel ceramics have a dielectric breakdown strength of 24–22 kV/mm.⁶

The presence of moisture in the polymeric electrical insulator can increase dielectric conductivity and promote the corrosion of metal conductors that leads to device failure. So it is desirable to prepare polymeric insulators with low moisture uptake. The prepared polymers showed low moisture uptake, in the range of 1.1%–2.4%. This may be explained by the use of aliphatic diisocyanate (HDI) and hydrophobic polyol (CAPA) for fabrication of the prepared PUIs. Adhesion of the polymeric insulator to the metallic surface was also very important. Visual inspection of

the polymeric cured films indicated they all had excellent adhesion to aluminum and copper foils.

CONCLUSIONS

The crosslinking reaction of epoxy-terminated polyurethane with poly(amic acid) led to imide-modified polyurethane. The thermal, mechanical, and electrical properties of the prepared polymers changed depending on the chemical structure of the constituents. Samples based on CAPA polyol with a molecular weight of 2000 and MDI as the diisocyanate showed improved thermal stability and mechanical properties, as well as performing the best according to voltage breakdown. However, the sample based on CAPA polyol with a molecular weight of 2000 and HDI as the diisocyanate showed the best performance regarding the dielectric constant and dissipation factor.

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