

Effects of Polyurethane Soft Segment and Crosslink Density on the Morphology and Mechanical Properties of Polyurethane/Poly(allyl diglycol carbonate) Simultaneous Interpenetrating Polymer Networks

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ABSTRACT: Tough, optically clear simultaneous interpenetrating polymer networks (SINs) of polyurethane (PU) and poly(allyl diglycol carbonate) (ADC) at different compositions were synthesized. The effects of the molecular weight of PU soft segment on the morphology, mechanical properties, and thermal transition behavior of the SINs at two levels of crosslinking agent were studied. The miscibility of PU/ADC SINs, studied by TEM and DMA, was greatly influenced by the SIN composition and the molecular weight of poly(ϵ -caprolactone) diol (PCL) as the PU soft segment. A single-phase morphology at a PU concentration of 10% changed to a very fine microheterogeneous morphology as the molecular weight of PCL changed from 530 to 1250. The two-phase morphology of the PU10/ADC90 SIN based on higher PCL molecular weight (PCL 1250) was also confirmed by DMA, which displayed a sharp peak for the ADC-rich phase and a small shoulder for the PU-rich phase

transition in the $\tan \delta$ plot. The SINs at 20–30% PU composition exhibited co-continuous phase morphology in the transmission electron micrographs, in which the phase regions grew larger as the PCL molecular weight increased from 530 to 1250. All the SIN samples possessed excellent optical transparency except two samples with 30% PU composition based on PCL 1250, which showed a hazy appearance. The tensile strength, modulus, and toughness of the SINs decreased by increasing the molecular weight of PCL from 530 to 1250, whereas the elongations at break remained nearly unchanged. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 89: 1583–1595, 2003

Key words: simultaneous interpenetrating polymer networks; poly(allyl diglycol carbonate); polyurethanes; morphology; mechanical properties

INTRODUCTION

The chemical and physical combination of two or more structurally dissimilar polymer provides a convenient route for the modification of properties to meet specific needs. Among these materials, interpenetrating polymer networks (IPNs), which are intimate mixtures of two or more polymer networks held together by permanent entanglements, represent a unique new class of blends.^{1–3} The combination of varied chemical types of polymeric networks in different compositions, often resulting in controlled different morphologies, has produced IPNs with properties that show synergism between the two components. Plastic materials may attain improved toughness with an elastomeric component as the minor phase and a reinforced elastomer may result if phase proportions are reversed.⁴

Among the several methods of making IPNs, simultaneous interpenetrating networks (SINs), where both monomers are mixed and polymerized by independent and noninterfering routes, have advantages over other methods such as ease of synthesis and the ability to control their morphology by several factors.^{5,6} Because the morphology of multiphase materials significantly affects their properties, formation of SINs is a new approach for combining the polymer components to achieve desired properties.

The degree of intermixing of SINs is mainly affected by the thermodynamic miscibility of the components,^{7–9} the relative rate of network formation,^{5,10,11} the composition,^{12–14} the degree of crosslinking, and the mobility of the polymer chains at the time of phase separation.^{13–15} However, complete miscibility is not essential to achieve complete phase mixing because the permanent entanglements and crosslinking associated with simultaneous polymerization restrict phase separation.^{17,18}

Maximum restriction of phase separation should result when the polymer begins to gel near or before phase separation of the two components. If phase separation precedes gelation, the domains are rela-

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TABLE I
Materials Used in Preparation of SINs

Material description	Source	Designation
Isophorone diisocyanate	Aldrich	IPDI
1,4-Butanediol	Aldrich	BDO
Trimethylolpropane	Aldrich	TMP
Poly(caprolactone) diol, MW 530	Aldrich	PCL530
Poly(caprolactone) diol, MW 1250	Aldrich	PCL1250
Allyl diglycol carbonate	Sola, Inc.	ADC
Benzoyl peroxide (ADC initiator)	Interox	BP

tively large and the crosslinking stabilizes the phase-separated morphology.¹⁸

The combination of elastomeric and glassy networks in interpenetrating polymeric networks often allows IPNs to range in properties from filler-reinforced elastomer to rubber-toughened plastics, depending on which component becomes the continuous phase.⁵ Some attempts have been made to improve the fracture toughness of glassy polymers through formation of a variety of IPNs.^{11–13,19–22}

The effects of SIN composition, kinetics of phase separation, and crosslink density of the PU network on the final morphology and mechanical properties of the novel SINs composed of poly(allyl diglycol carbonate) (ADC) resin and various polyurethane (PU) networks were presented previously.^{11–13} In the present study the effect of polyol molecular weight in the PU component on the SINs' properties at two levels of crosslinking agents is investigated. Morphology of the SINs is studied by means of transmission electron microscopy (TEM) and dynamic mechanical analysis (DMA). The results of stress–strain properties and single-edge-notch tensile strength are employed to characterize the mechanical properties of the PU/ADC SINs.

EXPERIMENTAL

Materials

The raw materials used for these series of SINs are listed in Table I. They were used without further purification.

Synthesis

Casting of ADC homopolymer

Dried benzoyl peroxide powder as radical initiator was dissolved (3 wt %) in ADC monomer with vigorous stirring at 50°C. The solution was degassed and then poured into a glass mold and polymerized as flat sheets (3 mm thick) in a temperature-programmed oven. The temperature was slowly increased from 50 to 85°C over a period of 22 h. The ADC samples were

then removed from the mold and annealed at 105°C for 2 h.

Casting of SINs

All the hydroxyl components were first vacuum dried. Then the reagents including isophorone diisocyanate, poly(caprolactone)diol, trimethylolpropane (TMP), butanediol (BDO), and the ADC monomer containing the initiator were thoroughly mixed together at 50°C for about 30 min. A clear solution was obtained. The mixture was then degassed for about 30 min at room temperature, after which it was introduced into the glass mold and polymerized using the same method as for the ADC homopolymer. The SINs were annealed at 105°C for 2 h. The completion of each component reaction was verified by the disappearance of the NCO peak at 2270 cm⁻¹ and the allyl peak at 3092 cm⁻¹ using FTIR spectroscopy. The NCO/OH ratio was kept constant at a level of 1.03 for all SIN samples. To investigate the effect of molecular weight of the PU soft segment in the SINs at different compositions, six samples containing 10–30% PU with a TMP/BDO ratio of 2/3 (corresponding to 0.8 equivalent TMP) and the polyol molecular weight of 530 and 1250 were made. The crosslink density of the samples was also changed with increasing the TMP level from 0.8 to 1.6 equivalents.

Mechanical properties

The stress–strain properties were determined on an Instron model 1115 universal testing machine. The specimens had dimensions of 120 × 12 × 3 mm³. The test procedure was carried out at a crosshead speed of 1 mm min⁻¹ at 22°C using flat-forced grips at an intergrip distance of 60 mm. Fracture toughness was determined by the single-edge-notch tension method. Molded sheets were cut into specimens having dimen-

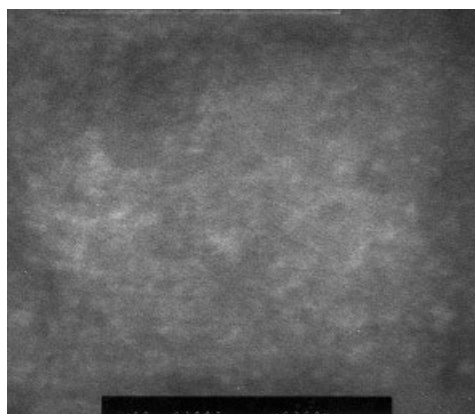


Figure 1 A TEM micrograph of the ADC90/PU10 SIN based on PCL 1250 and TMP = 0.8 equivalents (original magnification ×80,000).

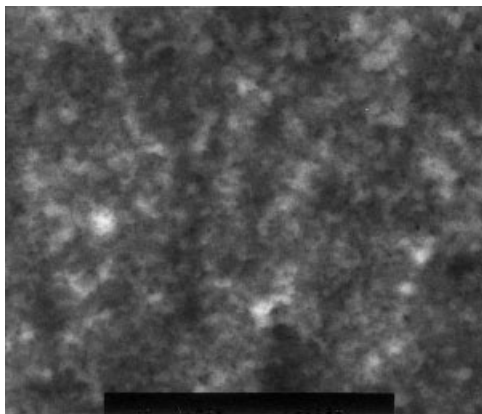


Figure 2 A TEM micrograph of the ADC80/PU20 SIN based on PCL 1250 and TMP = 0.8 equivalent (original magnification $\times 30,000$).

sions of $120 \times 15 \times 3.4 \text{ mm}^3$. The fracture toughness, or stress intensity at the onset of crack propagation K_{Ic} , was measured at a crosshead speed of 1 mm min^{-1} at 22°C according to the following equation²³.

$$Y = K_{Ic}BW/Pa^{1/2} = 1.99 - 0.41(a/W) + 18.70(a/W)^2 - 38.48(a/W)^3 + 53.85(a/W)^4$$

where a is the crack length, W is the width of the specimen, B is the thickness of the specimen, and P is the applied load.

Thermal-transition behavior

The thermal-transition behavior was determined using a DuPont dynamic mechanical analyzer (model 983; Boston, MA) in resonance mode. The $\tan \delta$ maximum was taken as the glass-transition temperature (T_g).

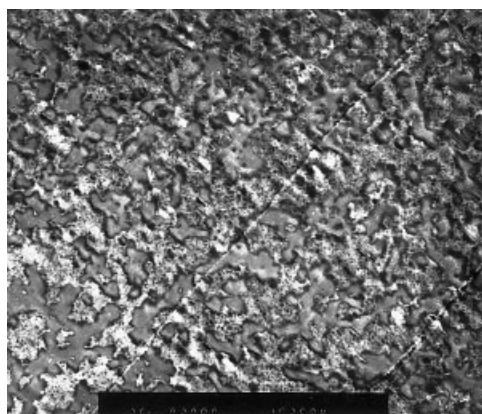


Figure 3 A TEM micrograph of the ADC70/PU30 SIN based on PCL 1250 and TMP = 0.8 equivalents (original magnification $\times 25,000$).



Figure 4 A TEM micrograph of the ADC90/PU10 SIN based on PCL 530 and TMP = 0.8 equivalents (original magnification $\times 100,000$).

TEM observation

A small piece of each SIN sample was stained in 2% aqueous osmium tetroxide vapor for 1 week. The samples were trimmed and then thin-sectioned (80 nm) using a Reichert-Jung Ultra-cut E ultramicrotome (Austria). The electron micrographs were observed in an H-7000 Hitachi (Tokyo, Japan) electron microscope. The polyurethane phase stained with osmium tetroxide and appears as the darker area in the TEM micrographs.

RESULTS AND DISCUSSION

Morphology

Figures 1–3 show the TEM micrographs of the PU/ADC SINs at 10–30% PU content. The molecular weight of the PU soft segment was 1250 (PCL 1250) and the TMP level was constant at 0.8 equivalent. A two-phase morphology is observed, indicating microphase separation. Moreover, the domains are not well defined, indicating dual-phase continuity. Similar

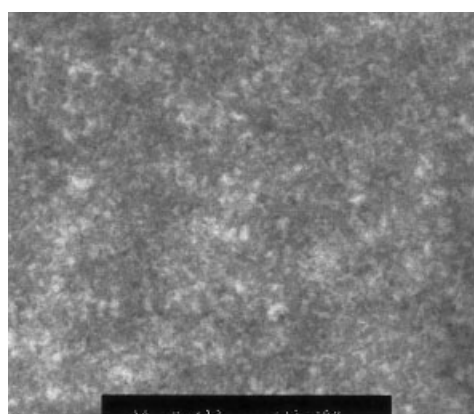


Figure 5 A TEM micrograph of the ADC80/PU20 SIN based on PCL 530 and TMP = 0.8 equivalents (original magnification $\times 30,000$).

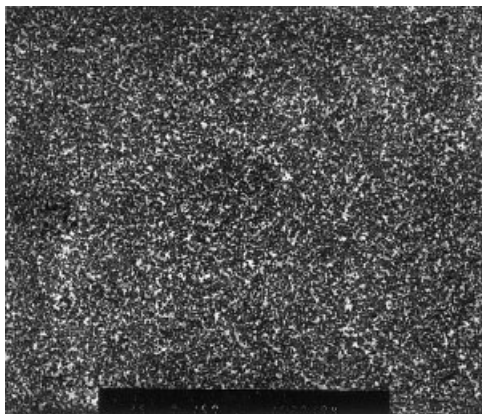


Figure 6 A TEM micrograph of the ADC70/PU30 SIN based on PCL 530 and TMP = 0.8 equivalents (original magnification $\times 2,500$).

co-continuous morphology but with finer structure is observed in the TEM micrographs of the SINs based on the lower molecular weight polyol (PCL 530) at 10–30% PU content (Figs. 4–6). Thus an increase in the molecular weight of polyol decreases the miscibility of the ADC and PU networks. Formation of a larger microstructure with increasing the molecular weight of the soft segment is believed to be attributed to a looser PU network that produces less interlocking between the two networks. It is of interest that, even at the low PU concentration of 10%, increasing the PCL molecular weight affects the morphology of the SIN. As the molecular weight of the PCL increased from 530 to 1250, a single-phase morphology changed to heterogeneous morphology with very fine phase structure. The two-phase morphology of the PU10/ADC90 SIN based on PCL 1250 was also confirmed by DMA studies, which displayed a sharp peak for the ADC-rich phase and a small shoulder corresponding to the PU-rich phase in the $\tan \delta$ plot (Fig. 12). Increasing the level of PU crosslinking agent (TMP) had only

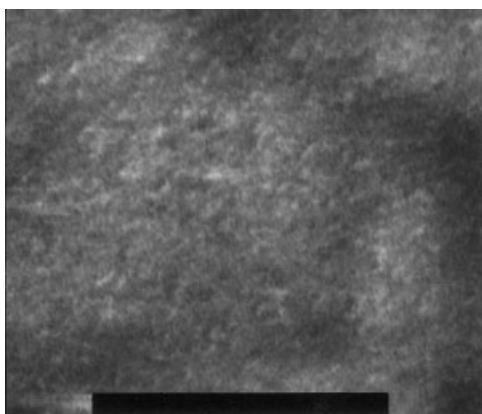


Figure 7 A TEM micrograph of the ADC90/PU10 SIN based on PCL 1250 and TMP = 1.6 equivalents. (original magnification $\times 80,000$).

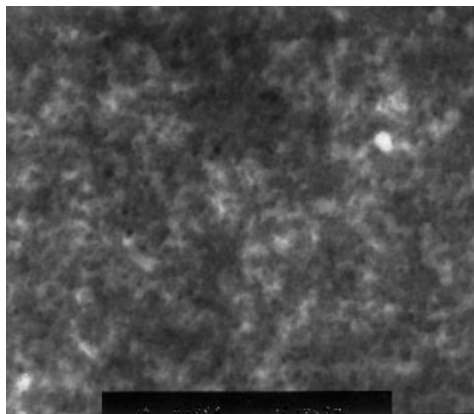


Figure 8 A TEM micrograph of the ADC80/PU20 SIN based on PCL 1250 and TMP = 1.6 equivalents (original magnification $\times 30,000$).

a slight effect on the morphology of the SINs based on PCL 1250 (Figs. 7–9). The TEM micrograph of the PCL 1250-based SIN at PU content of 10% shows a somewhat lower degree of heterogeneity at 0.8 equivalent TMP (Fig. 1) compared to 1.6 equivalents TMP (Fig. 7). This result is consistent with the earlier report¹³ for the SINs based on PCL 530, which showed coarser morphologies with increasing the TMP levels. The latter was believed to be a result of the faster formation of the PU network and thus a larger difference between gelation times of PU and ADC networks that resulted in a greater phase separation.

Dynamic mechanical analysis

Figure 10 shows the DMA thermogram of the pure polyurethane. The DMA thermograms of PU/ADC SINs containing 0–30% PU based on PCL 530 and a TMP level of 0.8 equivalent are shown in Figure 11. With 10% PU the SIN has a single glass transition around 115°C, indicating single-phase morphology

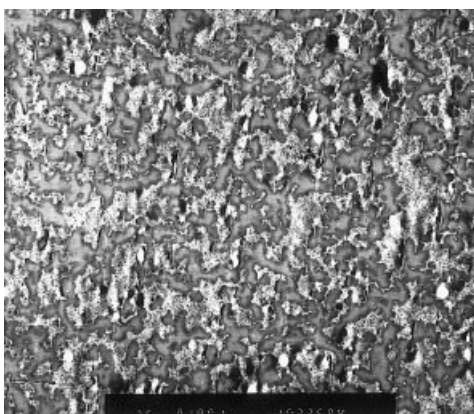


Figure 9 A TEM micrograph of the ADC70/PU30 SIN based on PCL 1250 and TMP = 1.6 equivalents (original magnification $\times 2,500$).

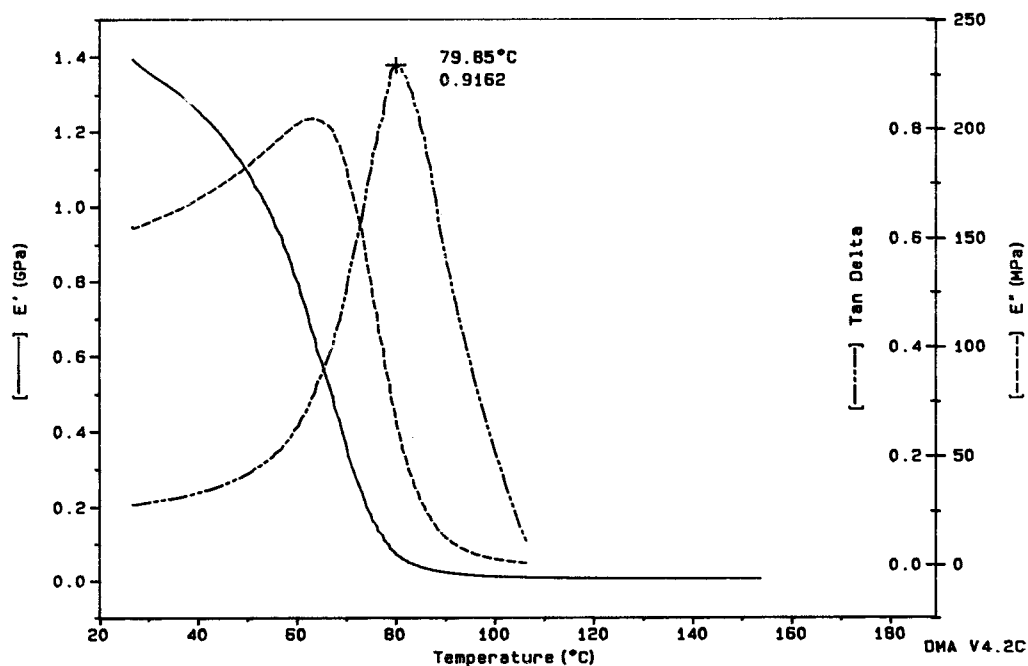


Figure 10 DMA thermograms of pure PU based on PCL 530 and TMP = 0.8 equivalents.

and complete miscibility. This is consistent with the TEM micrograph in Figure 4. As the PU content increased to 30%, the DMA thermogram showed two broad peaks with maxima at about 116 and 70°C, indicative of a two-phase structure and consistent with the TEM micrographs in Figures 5 and 6. At the same TMP level, increasing the molecular weight of polyol decreased the extent of intermixing between

the two SIN components. This result may be deduced from DMA thermograms of the SINs based on PCL 1250 (Fig. 12), in which the PU-rich phase in samples containing 10–20% PU appears as a weak shoulder, in contrast with the SINs based on PCL 530 (Fig. 11) that show a single T_g at 10% PU and a broad transition at 20%. At 30% PU composition the shoulder becomes more prominent as a result of increasing the PU con-

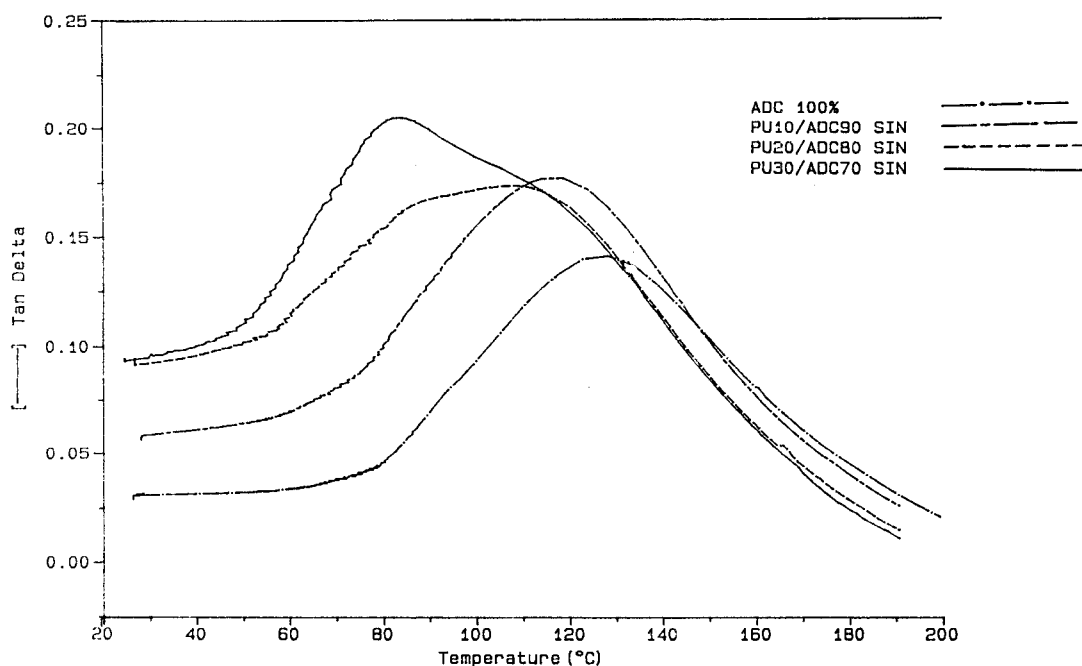


Figure 11 Variation of tan δ plots with temperature for the ADC/PU SINs at different compositions; PU based on PCL 530 and TMP = 0.8 equivalents.

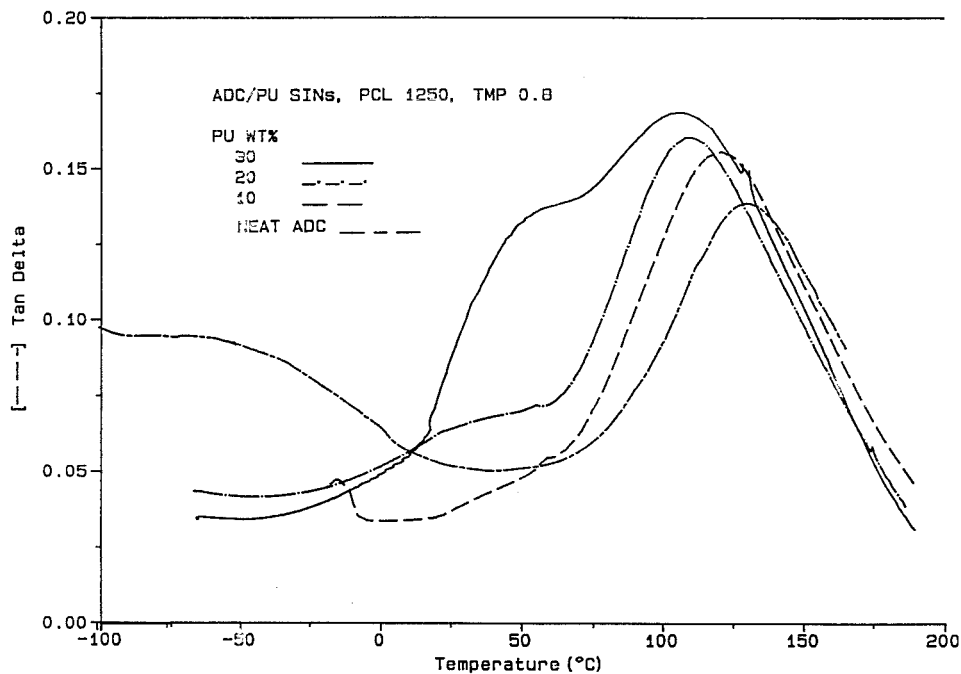


Figure 12 Variation of $\tan \delta$ plot with temperature for the ADC/PU SINs at different compositions; PU based on PCL 1250 and TMP = 0.8 equivalents.

centration and higher degree of heterogeneity, as shown in the TEM micrograph (Fig. 3). Figures 13–15 display the effect of increasing the TMP level on the phase behavior of the ADC/PU SINs based on PCL 1250. The lower shift in the ADC glass transition at a higher TMP level implies lower miscibility between the two components. With an increase in the TMP concentration the PU gels faster, so the gelation time

difference between the two constituents increases and the miscibility in the SINs decreases. This result was also confirmed by TEM micrographs, particularly at SINs with 10% PU content, which exhibited a higher degree of heterogeneity at higher PU crosslink density. It should be noted that increasing the PU glass transition in these DMAs is an indication of an increase in its crosslink density.

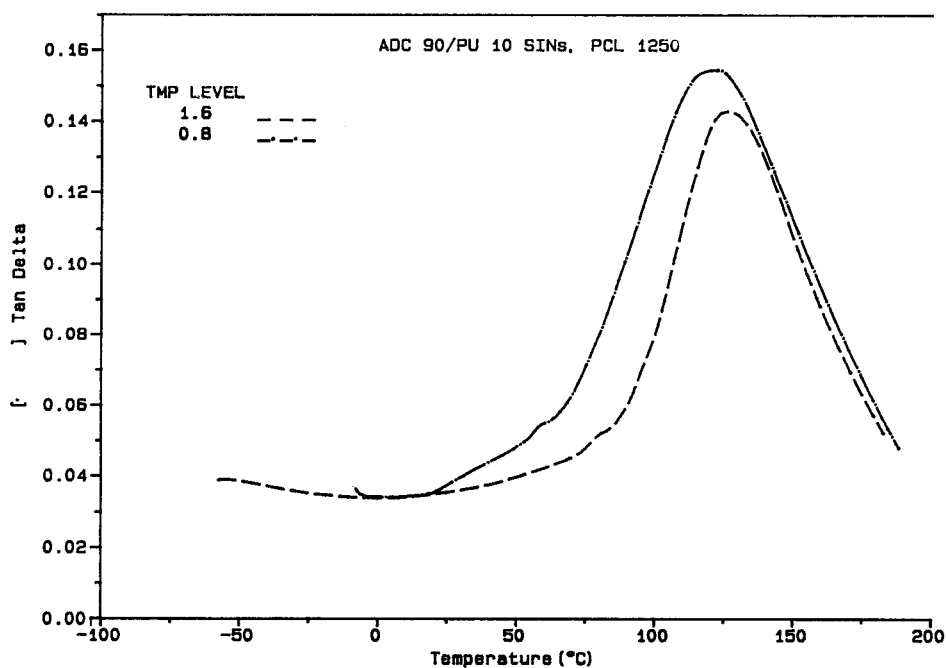


Figure 13 Temperature dependency of $\tan \delta$ for ADC90/PU10 SIN; PU based on PCL 1250 varying TMP contents.

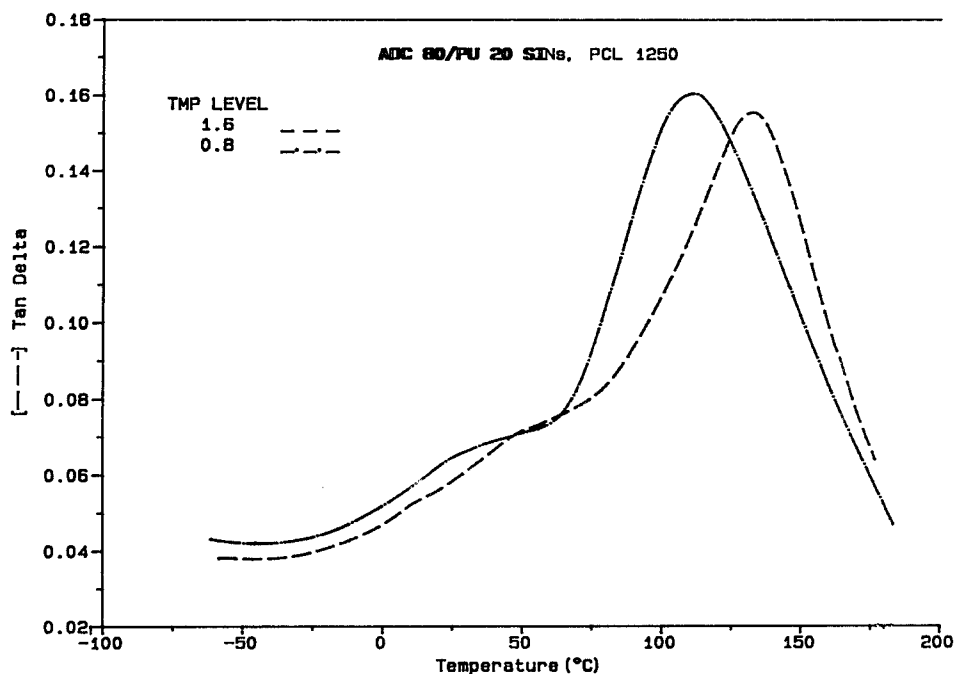


Figure 14 Temperature dependency of $\tan \delta$ for ADC80/PU20 SIN; PU based on PCL 1250 varying TMP contents.

Optical transparency of the SINs

The transparency of the PU/ADC SINs is directly linked to the phase domain sizes and to the difference of the refractive index between the two phases. All the PU/ADC SINs studied in this investigation possessed excellent optical transparency, except the samples with 30% PU composition based on PCL 1250. The large phase domain size of these SINs is most likely

responsible for their hazy appearance. However, when the PU phase was based on PCL 530, the transparency was not influenced by phase domain size and the SINs had optical transparency even at large domain sizes, which is an indication of a very close refractive index match between the two SIN constituents in this PU formulation. The light transmission of some of the SINs at 650 nm is shown in Table II. A

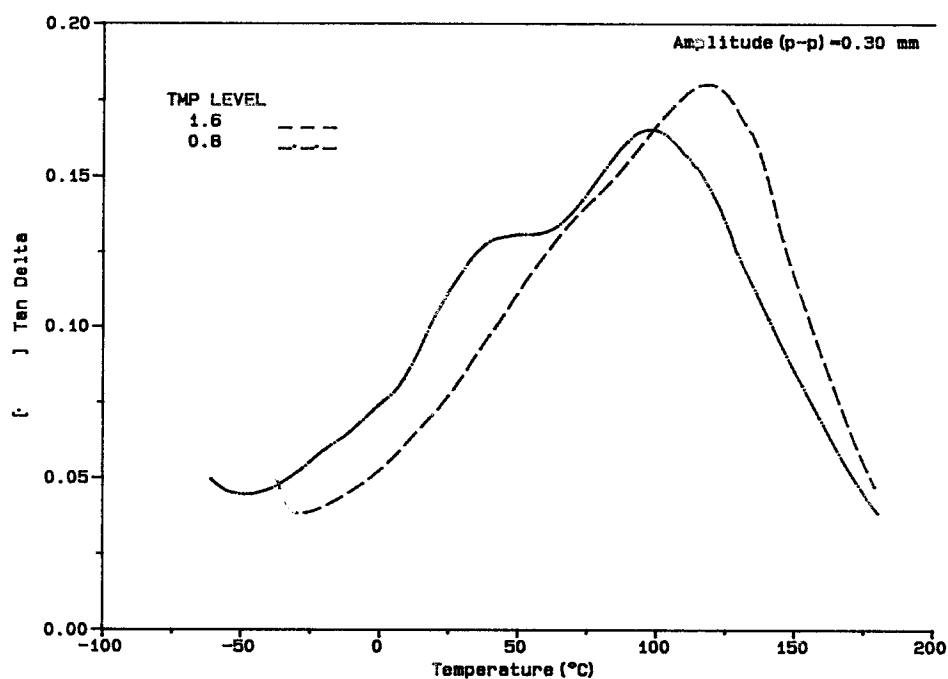


Figure 15 Temperature dependency of $\tan \delta$ for ADC70/PU30 SIN; PU based on PCL 1250 varying TMP contents.

TABLE II
Light Transmission of the SINs at 650 nm

PU%	TMP level (equivalents)	PCL molecular weight	Light transmission at 650 nm (%)
0 (neat ADC)	—	—	90.4
10	0.8	1250	90.1
20	0.8	530	89.7
20	1.6	530	90.3
20	1.6	1250	90.3
30	0.8	530	92.9
30	1.6	1250	16.7

typical light-transmission spectrum in the range of 350–750 nm is shown for the ADC70/PU30 SIN (Fig. 16).

Mechanical properties

As the PU content of ADC/PU SINs based on PCL 530 increased from 0 to 30%, the fracture toughness as measured by the stress intensity factor K_{Ic} increased from 0.6 to 1.2 MPa · m^{1/2} (Fig. 17) and the elongation at break increased from 6 to 23.5% (Fig. 18). This increase in fracture toughness and elongation was accompanied by a decrease in modulus from 1020 to 580 MPa (Fig. 19). Imparting more flexibility in the PU network by increasing the molecular weight of the polyol from 530 to 1250 decreased the modulus, tensile strength, and toughness of the SINs, particularly at 20 and 30% PU composition, whereas the elongation at break remained constant (Figs. 17–20). These results indicate that the mechanical properties of each constituent network in the SIN systems with co-continuous morphology strongly influence the mechanical properties of the SIN. Furthermore, interfacial intermixing between the two networks is a very important factor in formation of SINs with improved mechanical properties. Thus it can be inferred that the lower intermixing of the two components as a result of

larger morphology, accompanied by a PU network with poorer mechanical properties, is responsible for the formation of samples with lower mechanical properties in the SINs based on PCL 1250 compared to those in the SINs based on PCL 530.

Increasing the PU crosslink density by raising the TMP level significantly improved the mechanical properties of the SIN samples of lower polyol molecular weight of 530, particularly at a composition of 30% PU. As the TMP level was increased, the modulus of ADC70/PU30 SIN increased to 950 MPa (Fig. 21), marginally less than the ADC resin itself, and the tensile strength increased to 35 MPa (Fig. 22). Concomitant with this increase in modulus, the stress intensity factor K_{Ic} also increased to a maximum value of 1.4 MPa · m^{1/2}, decreasing slightly as the crosslink density further increased (Fig. 23). As the crosslink density of the PU network is increased, the phase regions of each network grow in size because of the higher gelation time difference between the two networks, although the morphology still remains co-continuous. The high fracture resistance of these SINs indicates strong interfacial bonding of the two networks at all TMP levels. It may thus be inferred that each component contributes to the total toughness of the SIN system. This is consistent with the enhancement of fracture toughness of the SINs with the increase in strength of the PU phase as the TMP content, and hence the crosslink density, increases. At very high PU crosslink densities, the fracture toughness slightly decreases, possibly because of a decrease in the toughness of the PU phase. Alternatively, it may be attributed to the very large size of the domains, which have a relatively small interfacial area and thus less interlocking between the two phases.

Increasing the crosslinking agent (TMP) did not affect the modulus, tensile strength, elongation at break, and K_{Ic} of the SINs based on PCL 1250, as given in Table III. At 10–20% PU content, these ADC/PU SINs showed the expected behavior, that is, the elongation

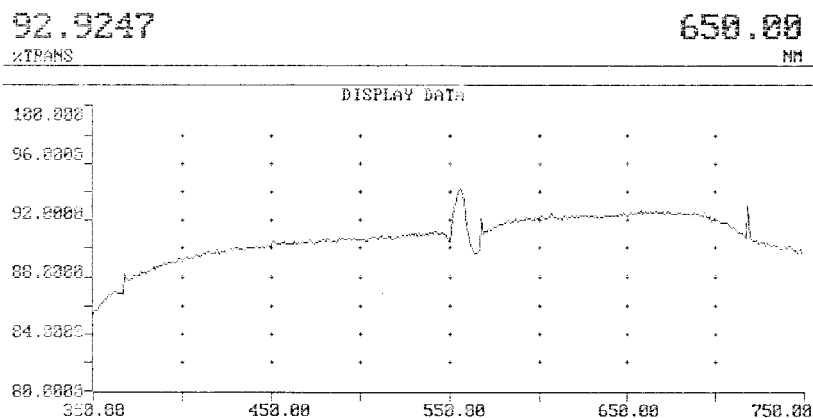


Figure 16 A typical light transmission spectrum in the range of 350–750 nm for the ADC70/PU30 SIN; PU based on PCL 530 and TMP contents of 0.8 equivalents.

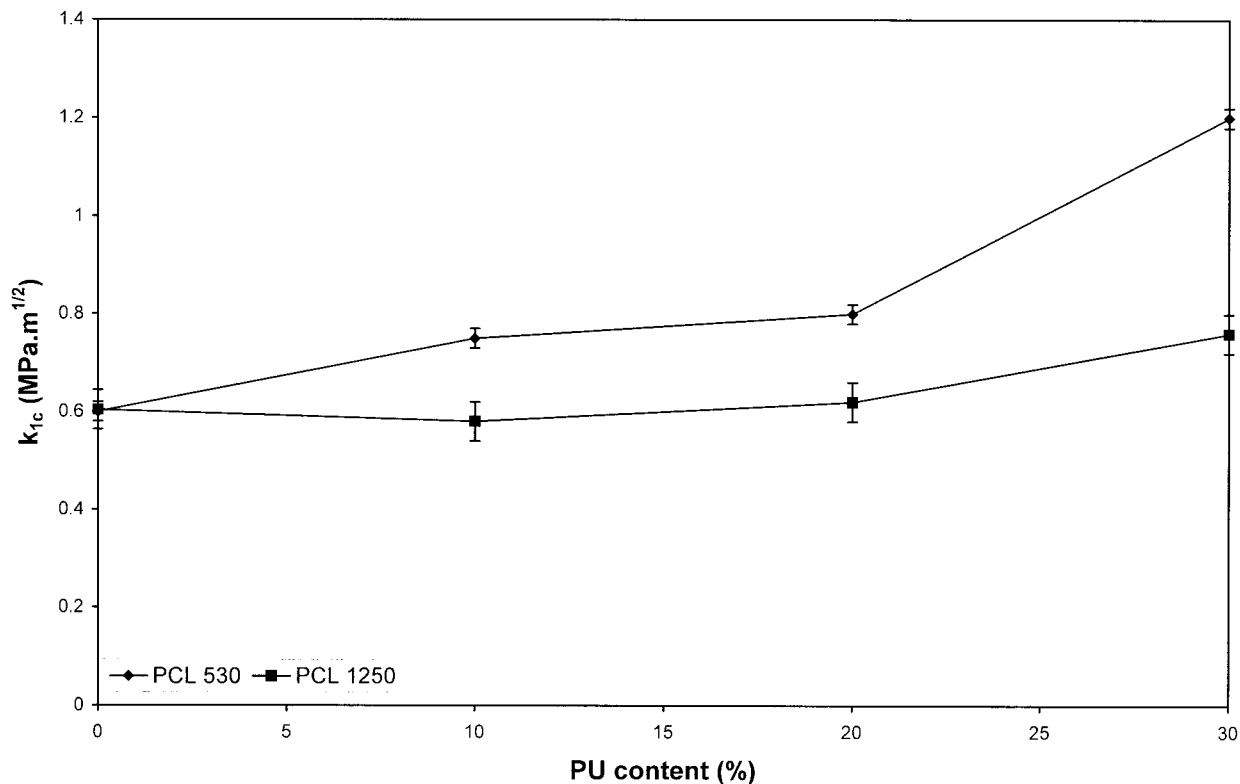


Figure 17 Variation of the fracture toughness (K_{1c}) of the ADC/PU SINs with composition at different PCL molecular weights; TMP = 0.8 equivalents.

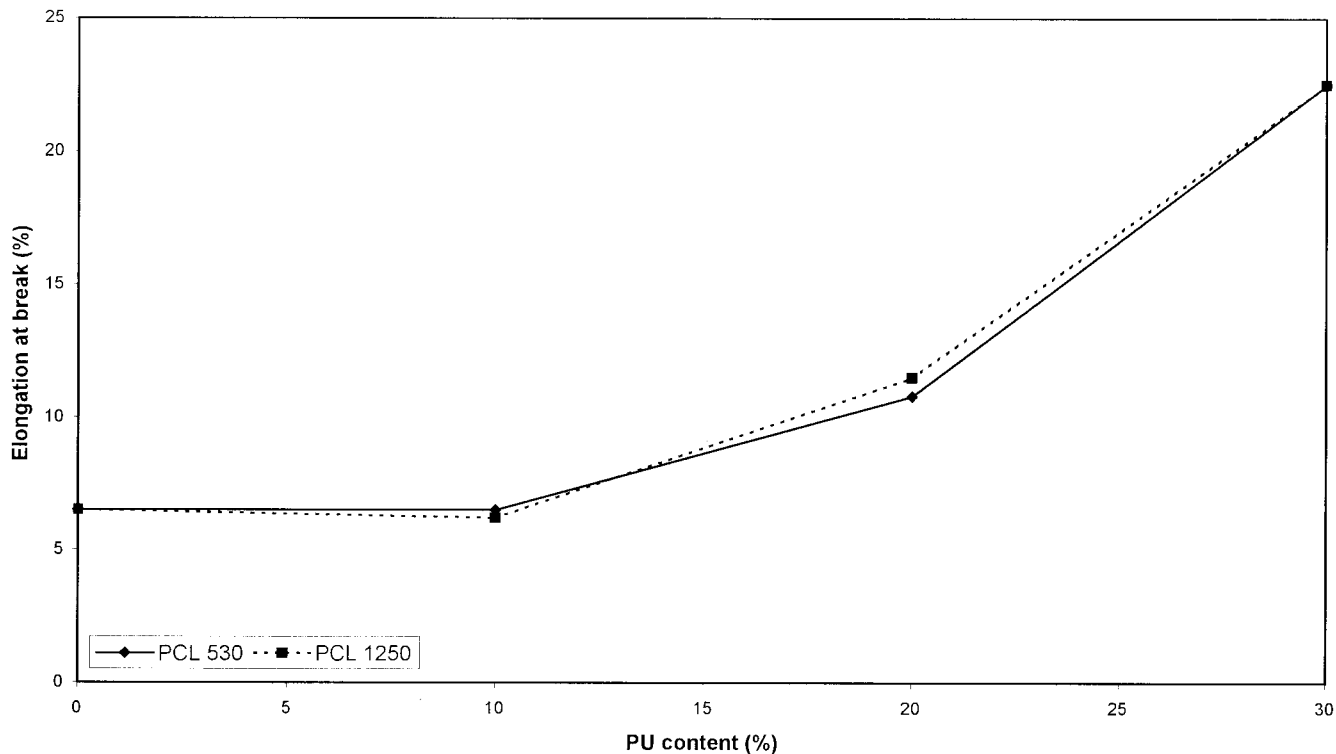


Figure 18 Variation of elongation at break of the ADC/PU SINs with composition at different PCL molecular weights; TMP = 0.8 equivalents.

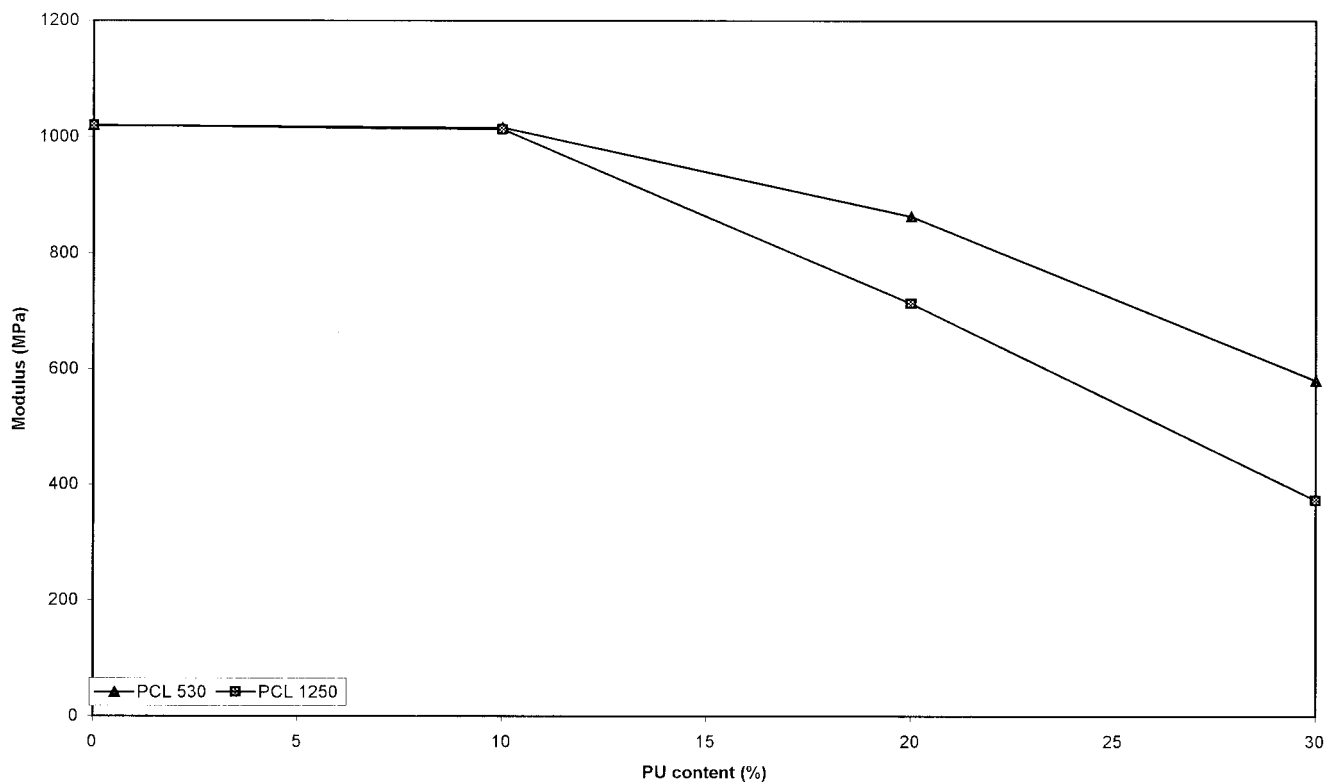


Figure 19 Variation of modulus of the ADC/PU SINs with composition at different PCL molecular weights; TMP = 0.8 equivalents.

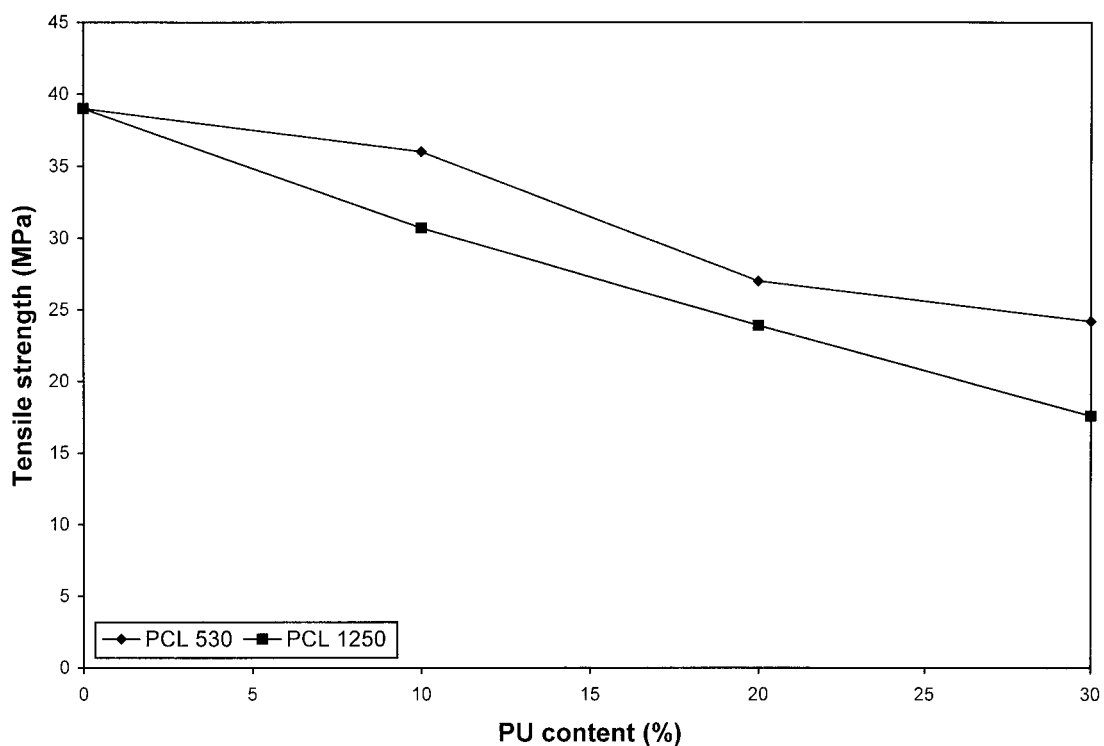


Figure 20 Variation of the tensile strength of the ADC/PU SINs with composition at different PCL molecular weights; TMP = 0.8 equivalents.

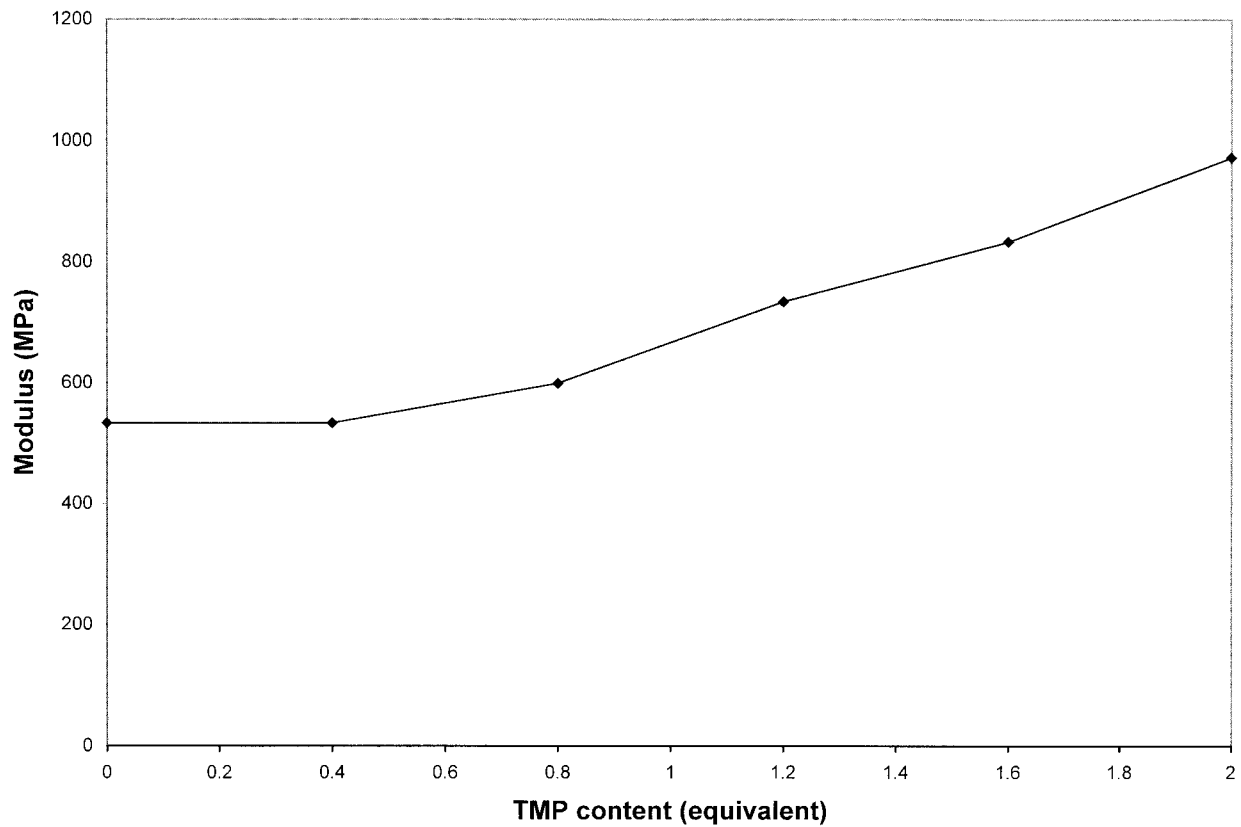


Figure 21 Variation of the modulus of ADC70/PU30 SINs; PU based on PCL530 at different TMP contents.

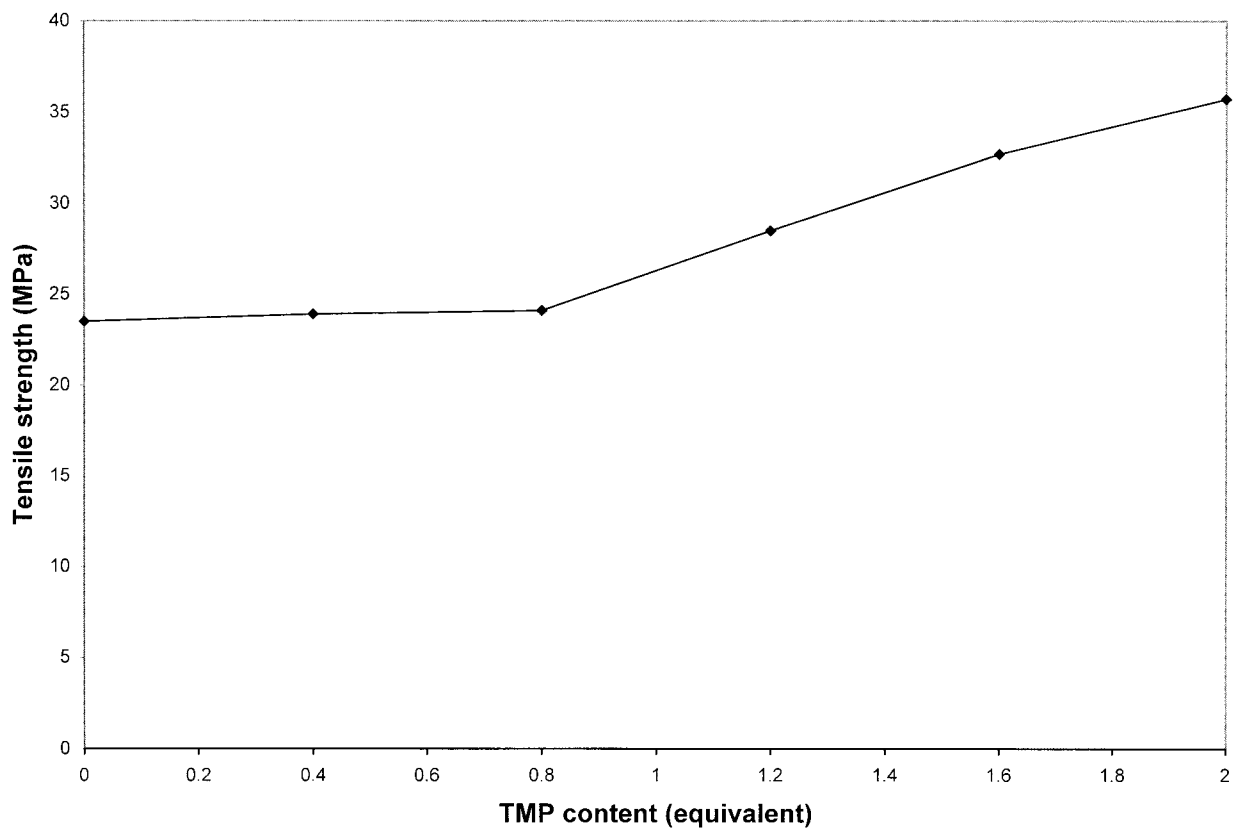


Figure 22 Variation of the tensile strength of ADC70/PU30 SINs; PU based on PCL530 at different TMP contents.

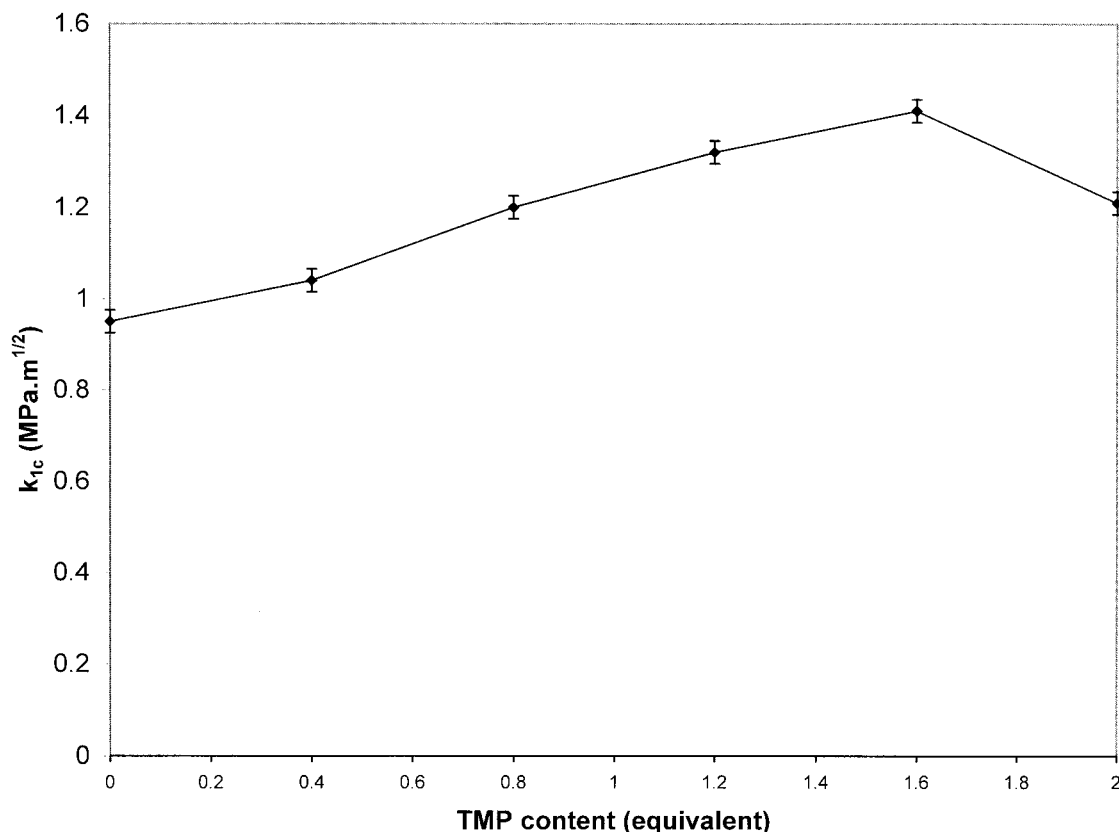


Figure 23 Variation of the fracture toughness (K_{1c}) of ADC70/PU30 SINs; PU based on PCL530 at different TMP contents.

decreased and modulus increased slightly with increasing the crosslink density of PU. The slight decrease in tensile strength and modulus of the ADC70/PU30 SINs with increasing the crosslink density of PU may be associated with the higher degree of phase separation and consequently less interlocking at this composition. These results confirm the foregoing statement that contribution of the PU network to the properties of the ADC/PU SINs is strongly dependent on the strength of the PU network itself and the extent of interlocking between the two networks. With higher molecular weight polyol a polyurethane network with relatively poorer mechanical properties is formed, which is not able to contribute effectively to the properties of SINs.

CONCLUSIONS

Tough, optically clear simultaneous interpenetrating polymer networks (SINs) based on poly(allyl diglycol carbonate) and polyurethane were synthesized. Increasing the PU content in these ADC/PU SINs decreased miscibility between the two constituents. At a fixed SIN composition an increase in the molecular weight of the soft segment in the PU decreased the miscibility between two component polymers, that is, larger phase regions were obtained as the polyol molecular weight was increased from 530 to 1250. However, increasing the PU crosslinking agent at a fixed composition decreased the miscibility between the two constituents, particularly when the lower polyol

TABLE III
Effects of TMP Contents on the Mechanical Properties of the ADC/PU SINs Based on PCL1250

PU (wt %)	TMP level (equivalents)	Tensile strength (MPa)	Elongation (%)	K_{1c} (MPa · m ^{1/2})	Modulus (MPa)
10	0.8	30.7	6.2	0.6 ± 0.03	1013.2
10	1.6	29.5	4	0.62 ± 0.02	1170.5
20	0.8	23.9	11.5	0.62 ± 0.04	713.5
20	1.6	24	10.9	0.64 ± 0.03	759.4
30	0.8	17.6	22.5	0.76 ± 0.04	373.1
30	1.6	15.8	20.5	0.77 ± 0.03	308

molecular weight (PCL 530) was used in the PU formulation.

The morphology of the SINs changed from complete miscibility, through a phase-separated structure, to a structure that exhibited co-continuous phase morphology. A lower polyol molecular weight in PU increased the interlocking and interpenetration between the two networks, especially at the phase boundaries, thus promoting greater miscibility and finer microstructure. The excellent optical transparency of these SINs was an indication of a very close refractive index match between the two components.

The mechanical properties of the SINs were strongly dependent on the morphology of the SINs. It was found that in co-continuous network morphology, when strong interfacial bonding exists, each component contributes to the total toughness of the SIN system. Increasing the PU strength by increasing its crosslink density increased the mechanical properties of the SINs, particularly at 30% PU composition. At this composition, the fracture toughness K_{Ic} increased from 0.6 to a maximum value of $1.4 \text{ MPa} \cdot \text{m}^{1/2}$ for the lower molecular weight polyol (PCL 530) with an optimum level of TMP. The modulus of the latter sample also increased from 550 to 950 MPa with an increase in the TMP level, only slightly less than that for the ADC resins. However, use of PCL 1250 in the PU network formed weaker structures, which led to the formation of SINs in which a minor increase in fracture toughness accompanied a significant decrease in the modulus and tensile strength at 30% PU composition.

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