

High-Performance Semi-Interpenetrating Polymer Networks Based on Acetylene-Terminated Sulfone.

II. Phase Separation and Morphology

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SYNOPSIS

The dynamic mechanical behavior, phase separation, and morphology of high-performance semi-interpenetrating polymer network (semi-IPN) obtained from acetylene-terminated sulfone (ATS-C) and high-performance thermoplastics have been studied by torsional braid analysis (TBA) and scanning electron microscopy (SEM). All the ATS-C/thermoplastic blends studied are compatible before curing. The addition of ATS-C results in a dramatic reduction in the glass transition temperature (T_g) of the thermoplastic. As the reaction of cure proceeds, the initially compatible blend passes through a stage of partial compatibility to achieve a fully incompatible semi-IPN. SEM observation of the fractured-etched surface reveals the formation of a co-continuous two-phase structure in the semi-IPNs. The connected globule and network morphology of the cured ATS-C phase are dispersed in a matrix of the thermoplastic phase. The size of dispersed particles decreases with increasing T_g of the thermoplastic. The mechanism of phase separation is discussed.

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INTRODUCTION

In the last few years, the concept of the semi-interpenetrating polymer network (semi-IPN) has been successfully applied to develop tough and microcrack-resistant high-temperature matrix resins for use in advanced carbon and glass-reinforced composites. The materials that serve as matrix for fiber-reinforced composites should possess high fracture toughness, high modulus, and good high-temperature properties. Several works have reported a semi-IPN approach to achieve the objective.¹⁻⁶ High-performance semi-IPNs can be prepared from a thermostable high-modulus, high glass transition temperature thermoplastic, and easy-to-process but brittle thermosetting polymer. Such high-performance semi-IPNs could be processed like a thermoset and possess good toughness like a thermoplastic and show notable improvements in fracture toughness and microcracking resistance of fiber-

reinforced composites over the unmodified thermosetting composites. Synergy in processability has also been realized.^{7,8} The cross-linking component acts as a plasticizer for the linear thermoplastic and the linear thermoplastic adds toughness to the brittle thermoset. The fracture toughness and microcracking resistance of brittle thermoset composites can be improved without compromising the easy processing and outstanding elevated temperature mechanical properties. These results demonstrate that the semi-IPN approach provides an effective way to toughen a very brittle thermosetting polymer of high T_g and have encouraged scientists to explore further the concept to develop a wider range of high-performance semi-IPNs.

Using the concept of semi-IPN, we developed a series of new semi-IPNs based on an acetylene-terminated sulfone (ATS-C) oligomer and high-performance thermoplastic (TP) blends. In a previous article,⁹ we described the synthesis, cure characteristics, and thermal properties of ATS-C/thermoplastic blends and their semi-IPNs that were prepared from a cross-linkable acetylene-terminated sulfone (ATS-C) oligomer and thermoplastic bis-

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phenol A polysulfone (PSF), a poly(ether sulfone) bearing a phthalidylidene group (PES-C), and a poly(ether ketone) bearing a phthalidylidene group (PEK-C), respectively. The results showed the addition of the thermoplastics had no effect on the mechanism of the ATS-C cure reaction and thermal behavior of cured systems. The glass transition temperature (T_g) and heat distortion temperature (HDT) of thermoplastics were remarkably decreased with adding the ATS-C oligomer, and the higher T_g and HDT of the thermoplastic, the more obvious the reduction of T_g and HDT. In the case of fully cured semi-IPNs, the T_g and HDT values of the thermoplastic phase were higher than or equal to the T_g and HDT of neat thermoplastic. But the thermal stabilities of semi-IPNs decreased as the content of the ATS-C oligomer increased. In this article, the phase separation, morphology, and dynamic mechanical behavior of ATS-C blends and their semi-IPNs have been investigated by torsional braid analysis and scanning electron microscopy.

EXPERIMENTAL

Materials

The bisphenol A polysulfone (PSF) was obtained from Shuguang Chemical Factory, Shanghai. The poly(ether sulfone) bearing a phthalidylidene group (PES-C), the poly(ether ketone) bearing a phthalidylidene group (PEK-C), and the acetylene-terminated sulfone (ATS-C) were provided by Changchun Institute of Applied Chemistry, Academic Sinica.

Resin Preparation

The thermoplastics and ATS-C were dissolved at 10 wt % of the total polymer in 1,2-dichloroethane. The samples of torsional braid analysis (TBA) were prepared by impregnating the glass braid with the above solutions, followed by drying at 100–120°C for 5 h. Cured samples are obtained by heating from room temperature (RT) to dynamic cure temperature (T_{max}) at 3°C/min (dynamic cure sample) or isothermal curing at given temperature (T_{iso}) for a certain time (isothermal cure sample) in the TBA apparatus, then cooling to RT. A 30% resin solution was used for the preparation of the uncured semi-IPN neat resin. The solution was cast onto a glass plate. The cast film was further dried in an oven at 100–120°C for 5 h to obtain the molding sample. The molding samples for ATS-C and the uncured

semi-IPN resin were then placed in a cold-matched metal die. This was then inserted into a press preheated to 320°C. A thermometer was attached to the die to determine the thermal history. When the die temperature reached 240°C, 20 MPa pressure was applied. The temperature was continuously raised to 320°C at a rate of 5°C/min and was cured at 320°C under 40 MPa pressure for 0.5 h. For the purpose of comparing the properties, the neat thermoplastic was also prepared. The die temperatures of the molded PSF, PEK-C, and PES-C samples are 320, 340, and 360°C, respectively, followed by cooling to $T_g + 50^\circ\text{C}$, and 60 MPa pressure was applied. When the die cooled to room temperature, the heat-pressured samples were removed from the press.

Resin Characterization

The dynamic mechanical behavior and the T_g 's of the uncured and cured specimens were measured by TBA by coating a glass braid with each resin solution. The TBA spectra were recorded at a heating rate of 3°C/min using a Model NB-1 torsional braid analyzer (Changchun Nonmetal Materials Test Instrument Factory). The damping (Δ) and relative rigidity ($1/p^2$) curves are obtained as a function of temperature. The transition temperature is determined by the temperature of damping peak. A scanning electron microscope was used to examine the phase morphology and fracture surfaces. To study the phase morphology, the molded specimen was fractured at room temperature. The fractured specimen was immersed in 1,2-dichloroethane at room temperature for 38 h and then dried at 100–120°C for 5 h. The weight loss due to the solvent extraction was determined. Then, the surface of the etched sample was observed under an SEM (Hitachi 520).

RESULTS AND DISCUSSION

Compatibility of Uncured Blends

The experimentally determined or calculated values of the solubility parameter have been used with some degree of success in the studying of polymer–polymer and polymer–solvent miscibility.^{10,11} In this study, the solubility parameter values for ATS-C and the thermoplastics were determined by calculation to ensure a common basis for comparisons between them. The method used was that proposed by Fedors.¹² Table I shows the calculated solubility parameter values for ATS-C and the thermoplastics used in this study. On this basis, the uncured ATS-

Table I Calculated Solubility Parameters, Molar Volumes, and Molecular Weight (MW) of Repeat Unit for ATS-C and Thermoplastics

Compound	MW of Repeat Unit	Molar Volumes (cm ³ /mol)	Solubility Parameters	
			(cal/cm ³) ^{1/2}	(J/cm ³) ^{1/2}
ATS-C	450	365	10.7	21.9
PSF	442	364	10.3	21.1
PES	236	157	11.3	23.1
PES-C	532	377	11.5	23.5
PEK-C	496	372	11.7	23.8

C blends with PSF, PEK-C, and PES-C with solubility parameter values between 21.1 and 23.8 (J/cm³)^{1/2} would be expected to be compatible, and this is found to be the case in practice.^{3,13}

Figure 1 shows the curves (damping [Δ] and relative rigidity [$1/p^2$] vs. temperature) of TBA for uncured ATS-C and PSF, PEK-C, and PES-C, respectively. The thermoplastics give two damping peaks: a primary peak corresponding to the glass transition of the thermoplastic, accompanied by decreasing the relative rigidity, and a higher temperature peak corresponding to the liquid-liquid transition of the thermoplastic.^{14,15} For ATS-C, the glass transition temperature (T_g) is 100°C. Above 180°C, the thermally induced reaction of the ethynyl group

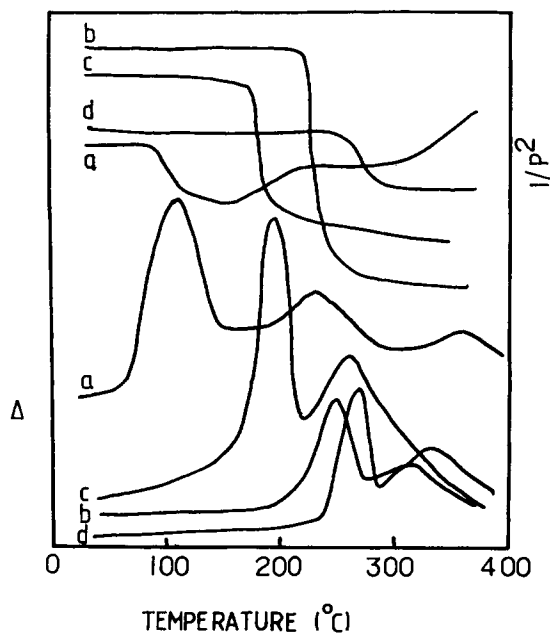


Figure 1 TBA of uncured ATS-C and PSF, PEK-C, and PES-C thermoplastics: (a) ATS-C; (b) PEK-C; (c) PSF; (d) PES-C.

occurs, accompanied by increasing the relative rigidity. A reaction peak of the ethynyl group at 250°C is observed. A damping peak at about 360°C is attributed to the high-temperature thermooxidative cross-linking reaction of cured ATS-C resin.¹⁶ The TBA curves of uncured ATS-C/PEK-C blends containing various ATS-C contents are shown in Figure 2. At all compositions, a single glass transition temperature was observed. The T_g is between the T_g of both components and decreases with increasing ATS-C content. These results indicate that uncured blends are compatible systems. The glass transition temperatures are summarized in Table II. Comparisons are made with values of T_g predicted from the Fox equation¹⁷:

$$1/T_g = W_1/T_{g1} + W_2/T_{g2} \quad (1)$$

and the Fried equation¹⁸:

$$\begin{aligned} \ln(T_g/T_{g1}) \\ = W_2 \ln(T_{g2}/T_{g1}) / [W_1(T_{g2}/T_{g1}) + W_2] \quad (2) \end{aligned}$$

The observed T_g values are lower than the calculated values. A similar observation has been observed for the blends plasticized with other reactive plasticizers.^{19,20} It can be seen from Table II that addition of ATS-C has substantially lowered the T_g of the thermoplastics, indicating that the temperature requirement of thermoplastics for processing can be improved. However, the addition of thermoplastics has no effect on the temperature of the ATS-C cure reaction and cure mechanism. These are also confirmed by the DSC results.^{9,21}

Phase Separation in Semi-IPNs

Although uncured ATS-C/thermoplastic blends are compatible, phase separation between the linear thermoplastic and the cross-linked ATS-C occurs

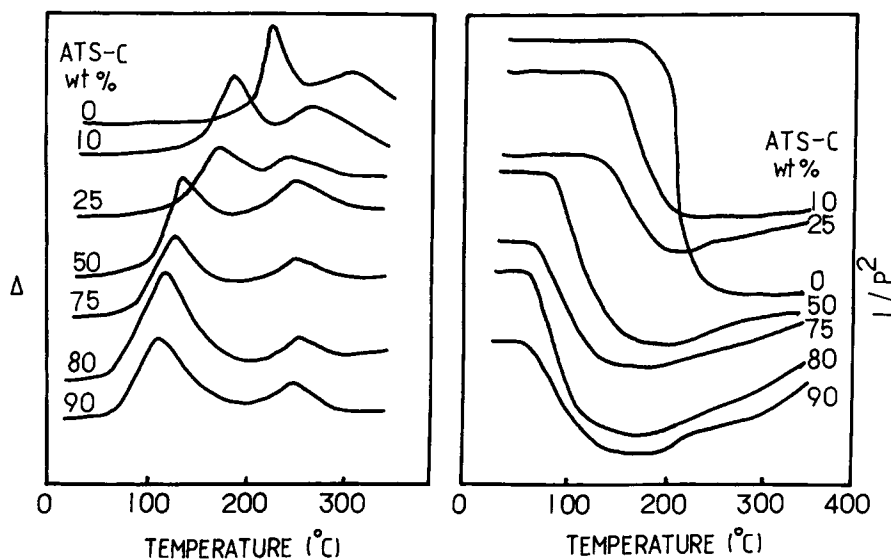


Figure 2 TBA of uncured ATS-C/PEK-C = 50/50 blend.

during the dynamic or isothermal cure, owing to a rise in molecular weight of cured ATS-C. The extent of the phase separation and the formation of partial compatible or incompatible systems depends on the cure conditions (Table III). Figures 3 and 4 show the damping (Δ) and relative rigidity ($1/p^2$) curves of TBA for ATS-C/PEK-C = 50/50 blends cured dynamically and isothermally. The glass transition temperatures of cured blends are summarized in Tables IV and V. It can be seen that cured ATS-C/thermoplastic blends are compatible systems, as the temperature of dynamical cure $T_{\max} < 240^\circ\text{C}$ (with a heating rate of $3^\circ\text{C}/\text{min}$) or the temperature of isothermal cure $T_{\text{iso}} < 210^\circ\text{C}$ (for 1 h). Only one T_g can be observed and partial compatible systems are obtained as $T_{\max} = 240\text{--}300^\circ\text{C}$ or $T_{\text{iso}} = 210\text{--}270^\circ\text{C}$. The present semi-IPN exhibited two apparent glass transition temperatures. The lower temperature

peak is attributed to the glass transition of the thermoplastic phase and the broadened damping peak with higher temperature is the glass transition of the cured ATS-C phase. In this case, the T_g of the thermoplastic phase in semi-IPN is lower than the T_g of the corresponding neat thermoplastic, and it is found that the higher the T_g of the thermoplastic the greater is the difference between the T_g of the thermoplastic phase and the T_g of the neat thermoplastic. These results indicate that the cured ATS-C phase still plasticizes the thermoplastics and decreases the T_g of the thermoplastics.

DSC results also show that for the cured ATS-C and its semi-IPNs cured at $T_{\max} < 260^\circ\text{C}$ (heating rate $10^\circ\text{C}/\text{min}$) a residual exotherm is observed on rescanning of the cured samples because of the presence of the remaining reactive groups.⁹ However, the curing of blends caused phase separation to form

Table II T_g of Uncured ATS-C/PEK-C Blends

ATS-C/PEK-C	Exp. T_g ($^\circ\text{C}$)	Fox T_g ($^\circ\text{C}$)	Fried T_g ($^\circ\text{C}$)	Tr ($^\circ\text{C}$)
0/100	225	225	225	—
10/90	185	209	206	260
25/75	175	187	183	250
50/50	140	154	149	250
70/30	130	130	127	250
80/20	120	120	118	250
90/10	110	110	109	250
100/0	100	100	100	250

Tr, the temperature of the damping peak for the ATS-C cure reaction.

Table III Compatibility of ATS-C/PEK-C Semi-IPNs

Dynamical Cure T_{\max} (°C)	Isothermal Cure T_{iso} (°C)	T_g	Compatibility
< 240	< 210	Single T_g	Compatibility
240-300	210-270	Two T_g $T_{g\text{ATP phase}} < T_{g\text{ATP}}$	Partial compatibility
300-360	270-300	Two T_g $T_{g\text{ATP phase}} = T_{g\text{ATP}}$	Incompatibility
> 360	> 300	Single T_g $T_{g\text{ATP phase}} > T_{g\text{ATP}}$	Incompatibility

two-phase systems exhibiting two T_g 's. With increasing the cure temperature, the degree of cure and phase separation increase. As $T_{\max} = 300\text{--}360^\circ\text{C}$ or $T_{\text{iso}} = 270\text{--}300^\circ\text{C}$, an incompatible semi-IPN can be obtained. The T_g of the thermoplastic phase in the semi-IPN network is equal to the T_g of the corresponding neat thermoplastics. This suggested that very little physical interpretation of the components occurred.¹ In this case, T_{\max} has no effect on the T_g of the thermoplastic phase. When cured at $T_{\max} > 360^\circ\text{C}$ or $T_{\text{iso}} > 300^\circ\text{C}$, the T_g of the thermoplastic phase is higher than the T_g of the neat thermoplastic and the T_g of the cured ATS-C phase is above 360°C and increases with increasing the cure temperature due to the increasing of the degree of cure.

It should be pointed out that the T_g of the two phases also shifts to high temperature with increasing cure time. The T_g of the two phases for ATS-

C/PEK-C blends cured at 300°C for different times are summarized in Table VI. It can be seen that with increasing cure time the T_g of the PEK-C phase slightly increases and is higher than that of the neat thermoplastic because the highly cross-linked ATS-C phase restricts the motion of the thermoplastic macromolecular segments. The ATS-C/PEK-C weight ratio also affects the T_g of the PEK-C phase (Table VII). The T_g of the thermoplastic resin phase slightly increases with increasing ATS-C content.

Although thermoplastics in semi-IPNs are different, the resulting semi-IPNs have similar dynamic mechanical behavior. In all the cases, the experiment clearly demonstrates that the T_g 's of cured ATS-C continuously shift toward higher temperature with increasing cure temperature due to the increase in molecular weight of the cured ATS-C. However, at the dynamic cure temperature, T_{\max}

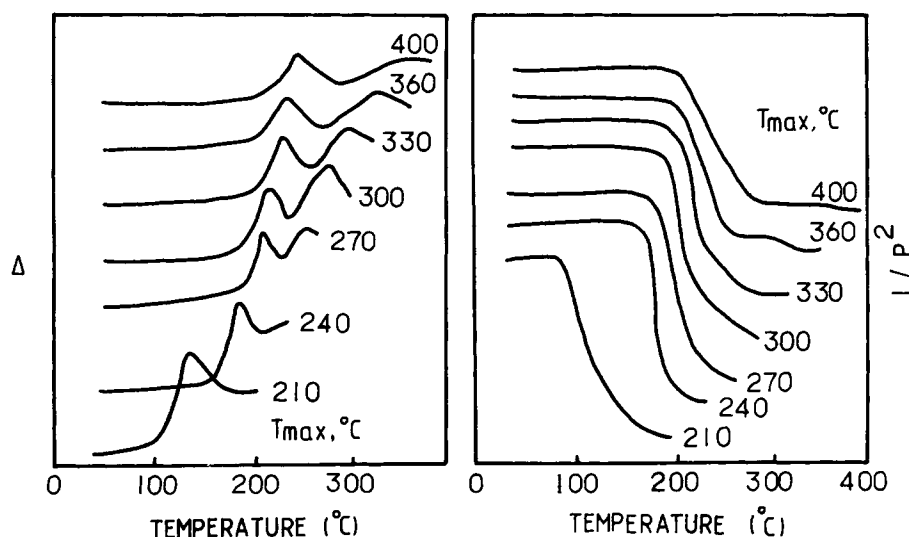


Figure 3 TBA of ATS-C/PEK-C = 50/50 blends cured dynamically from room temperature to T_{\max} repeatedly.

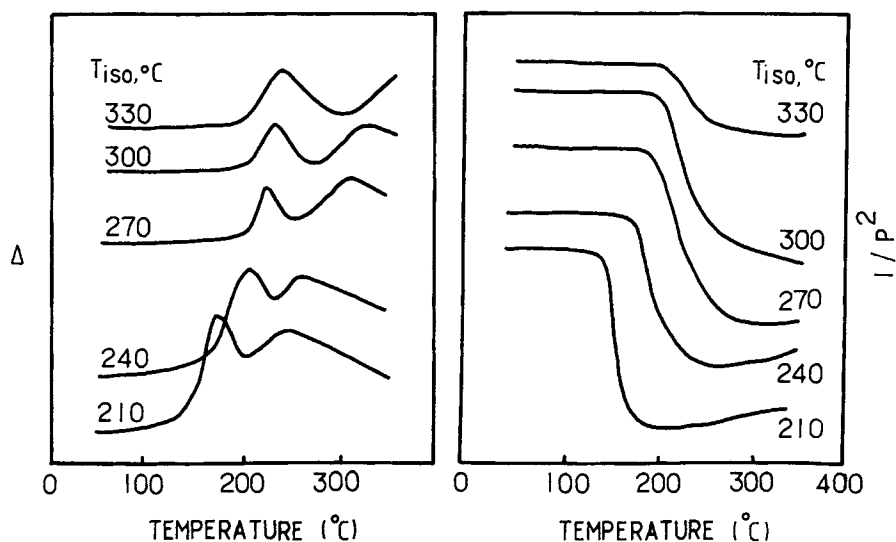


Figure 4 TBA of ATS-C/PEK-C = 50/50 blends cured at different temperatures (T_{iso}) for 1 h.

$< 360^{\circ}\text{C}$, the T_g of cured ATS-C $< T_{max}$; while cured at $T_{max} > 360^{\circ}\text{C}$, the T_g of cured ATS-C $> T_{max}$. These results indicate that a high-temperature reaction occurs at about 360°C . No T_g can be found for fully cured ATS-C in semi-IPNs cured at $T_{max} > 400^{\circ}\text{C}$. For semi-IPNs cured at $T_{max} < 300^{\circ}\text{C}$, the T_g of the thermoplastic phase increases with increasing T_{max} . However, the resulting cured ATS-C still plasticizes thermoplastics and the T_g of the thermoplastic phase in the semi-IPN is lower than the T_g of the corresponding neat thermoplastic because of the presence of the residual ethynyl groups or the formation of the lower molecular weight of cured ATS-C in semi-IPN. When T_{max} is between 300 and 360°C , the T_g of the thermoplastic phase is equal to the T_g of the corresponding neat thermoplastic. T_{max} has no effect on the T_g of the thermoplastic. But the T_g of the thermoplastic phase in the

semi-IPN cured at $T_{max} > 360^{\circ}\text{C}$ further increases. In this case, the T_g of the thermoplastic phase in the semi-IPN is higher than the T_g of the corresponding neat thermoplastics. One reason for this increase is that the thermooxidative cross-linking of thermoplastic resin occurs at high temperature. Second, the highly cross-linked ATS-C phase restricts the motion of the thermoplastic resin segments due to the entanglement of the polymer chains at the interface between the two phases and the formation of the semi-IPN.

Two-phase Morphology in Semi-IPNs

The results of dynamic mechanical analyses of ATS-C/thermoplastic blends indicate that the dynamic or isothermal cure induces phase separation. SEM examinations clearly indicate a two-phase morphology in the semi-IPN. Figure 5 shows the SEM

Table IV T_g of ATS-C/PEK-C = 50/50 Blends Cured Dynamically from Room Temperature to T_{max} Repeatedly

	T_{max} ($^{\circ}\text{C}$)						
	Uncured	210	240	270	300	330	360
T_{g1} ($^{\circ}\text{C}$)	140	180	210	220	230	235	240
T_{g2} ($^{\circ}\text{C}$)			250	275	295	330	> 360

T_{g1} , T_g of the PEK-C phase; T_{g2} , T_g of the cured ATS-C phase.

Table V T_g of ATS-C/PEK-C = 50/50 Blends Cured at T_{iso} for 1 H

	T_{iso} ($^{\circ}\text{C}$)				
	210	240	270	300	330
T_{g1} ($^{\circ}\text{C}$)	185	210	220	230	235
T_{g2} ($^{\circ}\text{C}$)	250	255	305	320	> 360

T_{g1} , T_g of the PEK-C phase; T_{g2} , T_g of the cured ATS-C phase.

Table VI T_g of ATS-C/PEK-C = 50/50 Blends Cured at 300°C for Different Times

	Cure Time (h)				
	1	2	4	8	16
T_{g1} , (°C)	230	235	240	245	245
T_{g2} , (°C)	320	> 360	> 360	> 360	> 360

T_{g1} , T_g of the PEK-C phase; T_{g2} , T_g of the cured ATS-C phase.

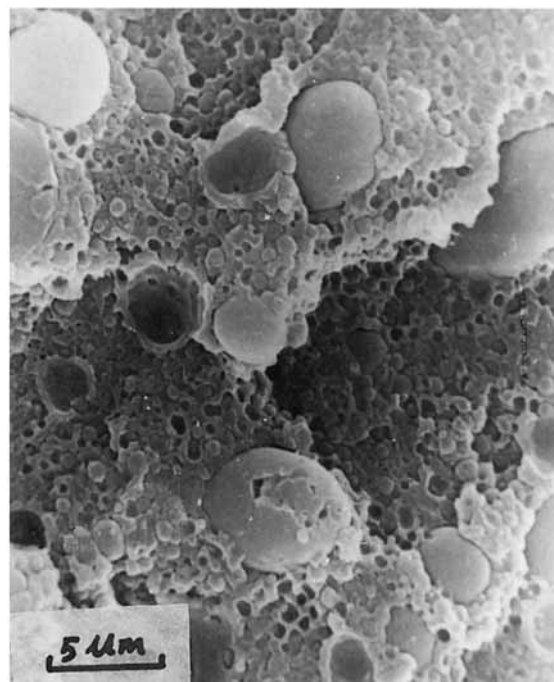
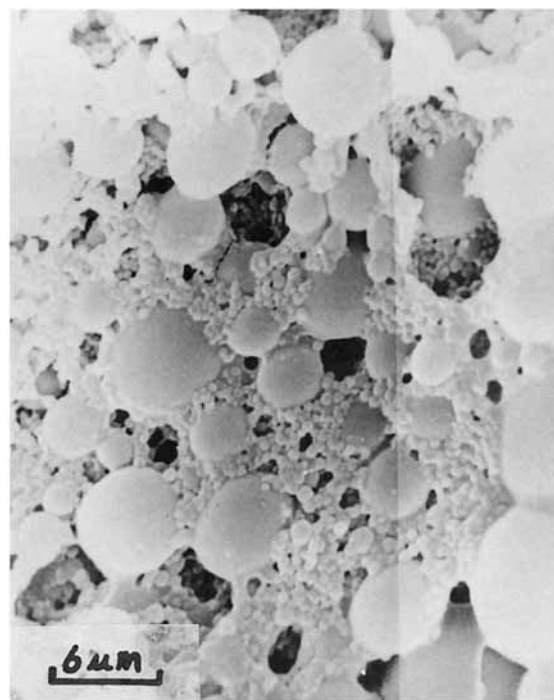
morphology of fractured surfaces of the ATS-C/PSF semi-IPN. It is clear from this photograph that the dispersed phase is present in the form of a bimodal particle distribution morphology (small and large particles). Large particles vary in size between 3 and 5 μm diameter; small particles are of sizes less than 1 μm and they are spherical. The two-phase morphology is similar to the one observed in the cured systems for thermoplastic-toughened epoxy or BMI resin.^{22,23}

In Figure 6 are shown scanning electron micrographs of the fractured-etched surface of the ATS-C/PSF semi-IPN. Because PSF, PEK-C, and PES-C are easily dissolved in dichloroethane, the remaining material is a cross-linked ATS-C-rich phase. Fine particles of a few micrometers diameter can be observed and particles seem to be connected to each other. The connected particle structure implies a two-phase morphology of interconnected spherical domains of the ATS-C-rich phase dispersed in a matrix of the thermoplastic phase to obtain a co-continuous two-phase structure. A similar morphology was obtained in the cured systems for thermoplastic-toughened epoxy resin and the mechanism of the development of a co-continuous two-phase structure has been discussed.²⁴ In addition,

Table VII T_g of ATS-C/PEK-C Semi-IPN Cured Dynamically from Room Temperature to 360°C

ATS-C/PEK-C	T_{g1} , (°C)	T_{g2} , (°C)
0/100	225	
10/90	235	> 360
25/75	235	> 360
50/50	235	> 360
80/20	240	> 360
90/10	—	> 360
100/0		> 360

T_{g1} , T_g of the PEK-C phase; T_{g2} , T_g of the cured ATS-C phase.

**Figure 5** Two-phase morphology of ATS-C/thermoplastic semi-IPNs. Cured conditions: room temperature (RT) to 250°C for 0.5 h to 330°C to RT.**Figure 6** The connected particles structure of ATS-C/thermoplastic semi-IPNs. Cured conditions: room temperature (RT) to 250°C for 0.5 h to 330°C to RT.

tion, network morphology is also observed on the fractured-etched surface in the cured ATS-C/thermoplastic semi-IPNs (Fig. 7).

The development of the connected-globule and network morphology may be as follows: As the cure reaction proceeds, the homogeneous mixture starts to phase-separate by the spinodal decomposition²⁴⁻²⁶ induced by the increase in the molecular weight of ATS-C phase, resulting in the development of a continuous two-phase structure. When phase separation proceeds, phase connectivity will be interrupted by the increase in interfacial tension, resulting in a dispersed droplet-type morphology. However, if a network has been established in the ATS-C phase, complete interruption cannot be realized and it eventually results in the connected-globule structure or network morphology. If once the interruption has taken place, the dispersed droplets grow in size without changing their location. Because the cured ATS-C particles are dispersed in a mixture of the thermoplastic phase with low mobility (high T_g), then, by further growth, the droplets finally contact each other to yield the connected-globule structure.

The major factors controlling the morphology of semi-IPNs include the chemical compatibility and the composition of both components, the cross-

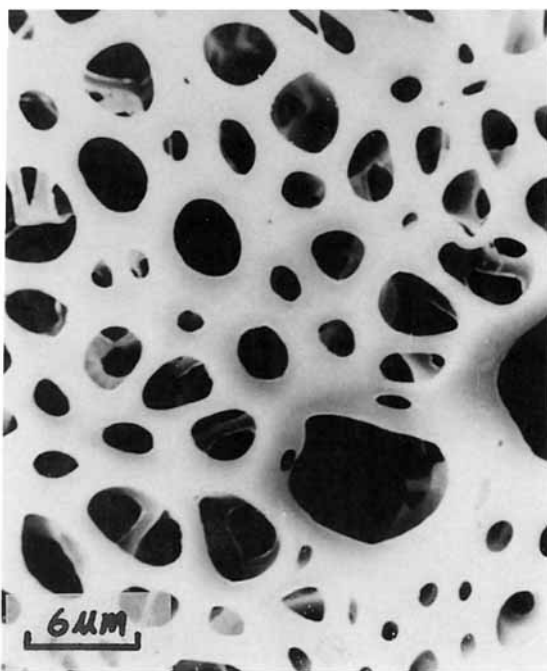


Figure 7 The network morphology of ATS-C/thermoplastic semi-IPNs. Cured condition: room temperature (RT) to 250°C for 0.5 h to 330°C to RT.

linking degree of the network, the mechanism of phase separation, and the molecular mobility of the system.²⁷ The differences among the semi-IPNs based on ATS-C are due to the thermoplastic resin. Because the thermoplastic resin does not affect the cure reaction of ATS-C, the formation of different morphologies depends mainly upon the nature of the thermoplastic resin. The size of dispersed particles decreases with increasing of the T_g of the thermoplastic due to the low molecular mobility. It is well known that phase separation competes with the cure reaction. The rate of phase separation at a temperature not far from T_g depends very much on the temperature.²⁴ For the ATS-C/PES-C semi-IPN, because of the higher T_g or viscosity of PES-C, the rate of phase separation is expected to be much reduced, whereas the rate of the cure reaction of ATS-C is not reduced so much. Therefore, the ATS-C phase is present in the form of network morphology and small particles, which cannot be observed by SEM.

CONCLUSIONS

A series of high-performance semi-interpenetrating polymer networks have been prepared from a cross-linkable acetylene-terminated sulfone (ATS-C) oligomer and a high glass transition temperature (T_g) thermoplastic bisphenol A polysulfone (PSF), a poly(ether ketone) bearing a phthalidylidene group (PEK-C), and a poly(ether sulfone) bearing a phthalidylidene group (PES-C). The results of dynamic mechanical analysis show that all the blends studied are compatible before the cure of ATS-C. The T_g of each uncured blend is between the T_g of both components, and the addition of ATS-C results in a dramatic reduction in the T_g of the thermoplastic and decreases with increasing ATS-C content. During the dynamic or isothermal cure, phase separation between the thermoplastic and cross-linked ATS-C takes place due to a rise in molecular weight of cured ATS-C. The extent of phase separation depends on the cure conditions. As curing proceeds, the initially miscible blend passes through stage of partial compatibility to achieve a fully incompatible semi-IPN. At the temperature of dynamic cure, $T_{max} < 240^\circ\text{C}$ (with heating rate of $3^\circ\text{C}/\text{min}$), or at the temperature of isothermal cure, $T_{iso} < 210^\circ\text{C}$ (for 1 h), cured blends are compatible. Whereas partial compatible blends are obtained as $T_{max} = 270\text{--}300^\circ\text{C}$ or $T_{iso} = 210\text{--}270^\circ\text{C}$, the T_g of the thermoplastic phase in semi-IPNs is lower than the T_g of the pure component. As $T_{max} = 300\text{--}360^\circ\text{C}$

or $T_{\text{iso}} = 270\text{--}300^\circ\text{C}$, a fully incompatible semi-IPN can be obtained and the T_g of the thermoplastic phase in semi-IPNs is equal to the T_g of the corresponding neat thermoplastic. When cured at $T_{\text{max}} > 360^\circ\text{C}$ or $T_{\text{iso}} > 300^\circ\text{C}$, the network density of ATS-C is high enough to immobilize the thermoplastic and cross-linked components and the T_g of the thermoplastic phase in semi-IPNs is higher than the T_g of the pure component. SEM observation of the fractured-etched surface indicates the formation of a co-continuous two-phase structure in semi-IPNs. The connected globule and network morphology of the cured ATS-C phase are dispersed in the matrix of the thermoplastic resin phase. The size of the dispersed particles decreases with increasing T_g of the thermoplastic.

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