

# Polycarbonate Blends with Styrene/ Acrylonitrile Copolymers

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## Synopsis

Lap shear adhesion between laminated sheets of polycarbonate and styrene/acrylonitrile copolymers exhibits a sharp maximum when the acrylonitrile content of the copolymers is in the range of 25–27% by weight. Observations of shifts in glass transitions of the two phases in melt-mixed polycarbonate/SAN blends suggest partial miscibility of one polymer in the other, and this solubility is at a maximum when the SAN copolymer has an acrylonitrile content in the same range causing maximum adhesion. Mechanical properties of injection-molded blends of polycarbonate with various SAN copolymers were also best when the acrylonitrile content was the same as that giving maximum adhesion. The partial miscibility behavior in blends as a function of acrylonitrile content of the copolymer is explained qualitatively in terms of a simple binary interaction model.

## INTRODUCTION

Commercial products based on blends of polycarbonate and ABS (acrylonitrile-butadiene-styrene) polymers were introduced a number of years ago.<sup>1-5</sup> These blends provide a useful combination of characteristics within a price structure which has allowed them to compete successfully with other products for certain applications.<sup>1,2,5</sup> This fact is scientifically interesting since quite often blends of arbitrarily chosen plastics have rather poor mechanical properties owing to thermodynamic immiscibility and to poor interfacial adhesion between the resulting phases,<sup>6,7</sup> making them unattractive for most commercial purposes. Toughness or ductility is usually the most deficient characteristic—quite often these properties exhibit a minimum when plotted vs. blend composition. The characteristics of specific ABS/polycarbonate blend products have been described,<sup>1-3,5</sup> but detailed investigations about how or why they deviate from the expectations for incompatible mixtures are quite limited.<sup>8</sup> Because of this situation, we recently investigated<sup>9</sup> the mechanical property-composition relationships for blends of polycarbonate with a particular ABS material. It was found that the notched Izod impact strength remained essentially at the level of pure ABS until about 50% polycarbonate was added and then increased rapidly upon further addition of the tougher polycarbonate. The blends have higher impact strength than pure polycarbonate when compared at low temperatures,<sup>8</sup> certain notch lengths,<sup>8</sup> or sample thicknesses of 0.25 in. or more.<sup>1,2</sup>

The purpose of this paper is to provide additional insight and knowledge

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about this blend system. The premise of this study was that the two components must interact in some way that leads to adhesion between them since lack of adhesion is frequently the demise of multiphase blends.<sup>10</sup> Because ABS is itself a two phase, rubber-toughened glass,<sup>11</sup> it was felt that the most fruitful course would be to eliminate the rubber and concentrate on the styrene/acrylonitrile copolymer (SAN) forming the matrix phase. The following reports on measurements of the adhesion between SAN and polycarbonate and the mechanical properties of their blends. The acrylonitrile content of the SAN copolymer has been varied over a wide range to gain further insight and to learn whether this might be a useful parameter for optimization. The results are interpreted in terms of the observed SAN-polycarbonate phase behavior.

### MATERIALS AND BLEND PREPARATION

The bisphenol-A polycarbonate used is a commercial product of the General Electric Co. designated as Lexan 131-111 which has  $\bar{M}_n = 13,300$  and  $\bar{M}_w = 34,200$ . A variety of SAN copolymers differing primarily in percent acrylonitrile were obtained through the courtesy of R. A. Mendelson of Monsanto and R. L. Jalbert of Borg-Warner. Pertinent information about these materials including molecular weights from gel permeation chromatography is given in Table I. Some were available only in small quantities, which limited the extent of experimentation possible. For the limit of zero acrylonitrile content, polystyrene was obtained from the Cosden Oil and Chemical Co. (designated 525) which has an unannealed heat distortion temperature of 74°C and a flow index of 5.0 g/10 min at ASTM D-1238-65T condition G. This material contains 3.0% oil.

Blends of polycarbonate and the various SANs were prepared by either of two methods depending on the quantity of sample available and the intended use. Test specimens for Instron or heat distortion measurements were prepared as follows. Pellets were combined in the desired proportions and dried for a minimum of 24 h at 100°C in a vacuum oven to prevent hydrolysis of the polycarbonate during melt processing. Melt blending was executed in a single screw laboratory extruder ( $D = 0.75$  in.,  $L/D = 20$ ). The extrudate from a rod die attached to the extruder was pulled through a water bath and immediately pelletized. The blended pellets were dried

TABLE I  
SAN Copolymers

Designation	Wt% AN	$\bar{M}_n$	$\bar{M}_w$	$[\eta]^a$ (dL/g)	Source
SAN-1	24.0	66,700	113,000	0.77	Monsanto
SAN-2	24.8	62,900	121,000	0.80	Monsanto
SAN-3	32.3	50,700	75,400	0.61	Monsanto
SAN-4	26.7	—	—	—	Monsanto
SAN-5	69.7	—	—	0.80	Monsanto
SAN-6	13.0	58,300	149,000	—	Borg-Warner

<sup>a</sup> Measured in DMF at 250°C.

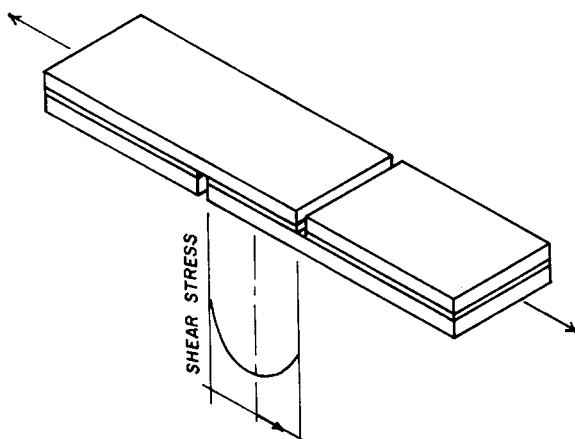


Fig. 1. Schematic illustration lap shear specimen formed from notched laminate. Variation of stress with position is indicated.

as before just prior to injection molding by a ram or a screw type machine into dog bone (ASTM D-638) and Izod (ASTM D-256) bars. The various heating zones for extrusion and molding were set in the range of 200–220°C for SAN or polystyrene and gradually increased with the polycarbonate content to the range of 270–290°C for pure polycarbonate.

Thin films for thermal analysis and dynamic mechanical testing were prepared by batch processing. After drying, pellets were mixed for 10 min at 60 rpm in a Brabender Plasticorder at temperatures ranging from 220°C to 270°C, depending on composition. The charge was transferred to a hydraulic press with platens heated to 175–225°C, depending on the blend composition where films ranging in thickness between 5 and 20 mils were pressed between Mylar films.

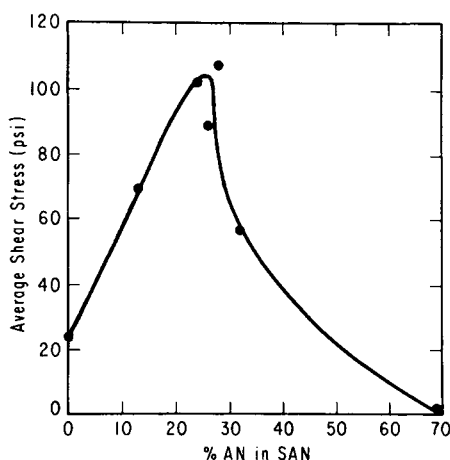


Fig. 2. Average shear stress at failure for polycarbonate/SAN laminates as a function of copolymer composition.

## INTERFACIAL ADHESION

Interfacial adhesion between polycarbonate and various SANs or polystyrene was measured using a lap shear specimen like that illustrated in Figure 1. In every case, polycarbonate was used for the two outer layers while the SAN or polystyrene was the inner layer of the three-piece sandwich. The sheets of each polymer used to form this laminate were compression-molded. The polycarbonate outer layer sheets were approximately 0.125 in. thick while the inner layer sheets were approximately 0.03125 in. thick. The three sheets were placed together in a mold and laminated at 190°C under pressure for about 8 min to form the bond at the two interfaces. At every point great care was needed to remove moisture otherwise bubbles formed. After lamination, 7 in.  $\times$  1 in. strips were cut and notched as shown so that a lap shear joint of 1 in.<sup>2</sup> area was formed. Only specimens with perfect-looking interfaces were used.

These specimens were placed in the grips of an Instron and pulled until separation occurred. Except for a few defective specimens, failure was the result of debonding at one of the interfaces. The loads required to shear the interface were recorded and used to calculate the average shear stress at failure. A shear stress distribution exists along the length of the joint as illustrated in Figure 1, and this distribution was calculated using a computer program developed from the analysis given by Goland and Reissner.<sup>12</sup> However, only the average stresses are discussed here for simplicity. These values are plotted in Figure 2 vs. the acrylonitrile content of the SAN inner layer. These data are most interesting in that a sharp maximum in adhesion with polycarbonate occurs between about 25% and 27% AN content of the SAN copolymer. The adhesion to polycarbonate at this optimal AN content is substantially higher than that of either polystyrene or SANs of high AN levels. This fact is consistent with other observations described later. It is significant to note that this optimum occurs near the azeotropic composition for styrene/acrylonitrile copolymerization, which is used to make many SAN copolymers and ABS polymers.

## PHASE BEHAVIOR

It is apparent from simple visual observations that SAN/polycarbonate blends are two phase materials; however, partial miscibility of one polymer in the other has been observed in blend systems<sup>5,13-17</sup> and was considered to be an important possibility in the present case. This issue was investigated by observation of glass transition behavior using differential scanning calorimetry and dynamic mechanical testing of blends formed from selected SANs and polystyrene with polycarbonate.

A Perkin-Elmer DSC-2 outfitted with a thermal analysis data station was employed for  $T_g$  detection using a scanning rate of 20°C/min. For all blends, two glass transitions were detected as expected; however, both were shifted towards each other consistent with the notion of partial miscibility. The DSC results are summarized in Table II by showing the  $T_g$  observed for the SAN and the polycarbonate phases of each blend. In each case, the  $T_g$  for the styrenic phase was higher than that of the pure styrenic polymer, as

TABLE II  
 Glass Transition Data for Blends of Polycarbonate and SAN Copolymers

Sample composition	Rheovibron		DSC	
	$T_{g,SAN}$	$T_{g,PC}$	$T_{g,SAN}$	$T_{g,PC}$
Polycarbonate	—	155	—	148
25% PS	110.5	153.1	93.6	142.0
50% PS	108.0	152.4	91.4	141.1
75% PS	104	N.T. <sup>a</sup>	87	140
Polystyrene	102.2	—	85.3	—
25% SAN-4	124.6	153.9	109	144
50% SAN-4	123.2	151.5	106.6	141
75% SAN-4	120.3	N.T.	105	140
SAN-4	116.0	—	100	—
25% SAN-3	123.5	154	109.8	144.5
50% SAN-3	122	150	108.9	141.1
75% SAN-3	117.5	N.T.	105.1	140
SAN-3	115.0	—	103.3	—
25% SAN-5	121.4	153.9	110	146.3
50% SAN-5	119.2	148.4	110	145.8
75% SAN-5	115.4	143.6	107	141
SAN-5	113.8	—	107	—

<sup>a</sup> N.T. = not tested since sample breaks in Rheovibron before  $T_g$  is reached.

would occur if some polycarbonate were dissolved in it. Likewise, the  $T_g$  for the polycarbonate phase was lower than that of pure polycarbonate consistent with some styrenic polymer dissolved in it.

The same compositions were examined using a fully automated Rheovibron at a frequency of 110 Hz. Figures 3 and 4 illustrate the type of results obtained by showing  $E'$  and  $E''$  vs. temperature for blends containing SAN-4. Again two glass transitions are evident but both are shifted toward each

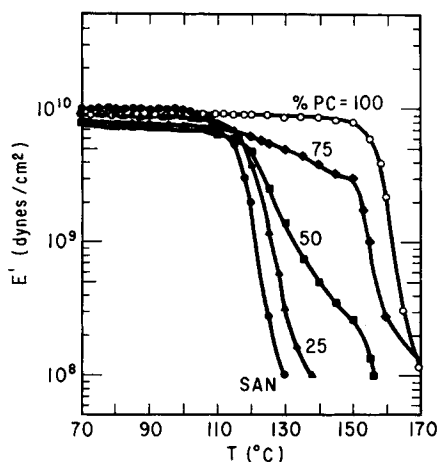


Fig. 3. Storage modulus as a function of temperature for polycarbonate/SAN-4 blends. 26.7% AN in SAN.

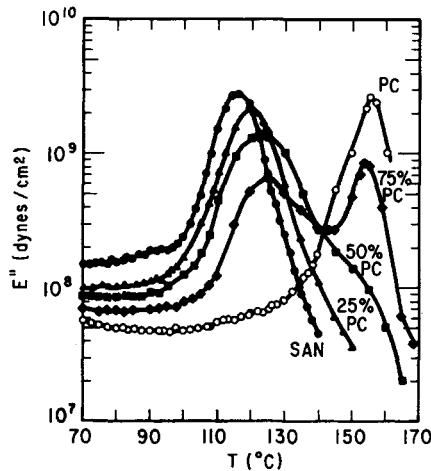


Fig. 4. Loss modulus as a function of temperature for polycarbonate/SAN-4 blends. 26.7% AN in SAN.

other as can be seen by noting the location of the  $E''$  peaks in Figure 4. The glass transitions noted are summarized in Table II. The trends are the same as seen by DSC.

For comparing the extent of partial miscibility (as judged by  $T_g$  shifts) as the % AN level of the SAN varied, two approaches are used. The first method compares the percent shift in the glass transition of the styrenic phase, using the following arbitrary definition for this quantity:

$$\% T_g \text{ shift} = \frac{T_g - T_{g,SAN}}{T_{g,PC} - T_{g,SAN}} \times 100 \quad (1)$$

where  $T_g$  is the glass transition observed for the styrenic phase in the blend and  $T_{g,SAN}$  and  $T_{g,PC}$  are the glass transitions of the pure polymers indicated

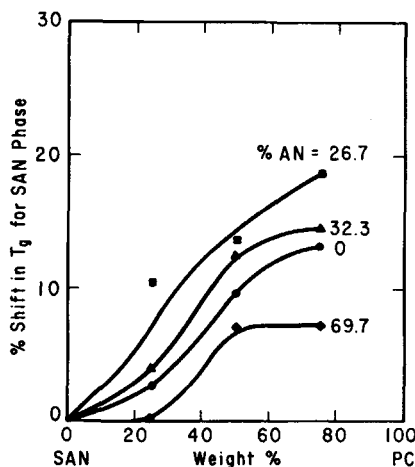


Fig. 5. Relative shift in  $T_g$  by DSC for SAN phase in blends with polycarbonate for SAN copolymers.

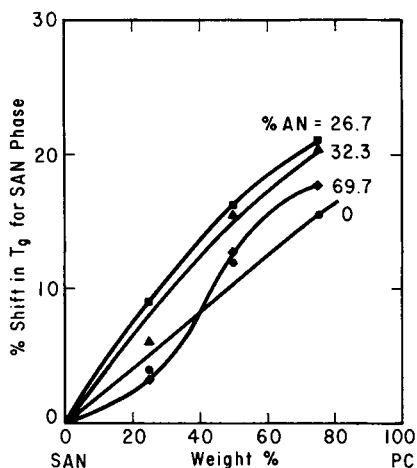


Fig. 6. Relative shift in  $T_g$  by Rheovibron for SAN phase in blends with polycarbonate for SAN copolymers.

by the subscripts. Figure 5 shows the DSC data reduced in this way plotted vs. percent polycarbonate in the blend for each of the four styrenic polymers examined. The greatest shift, regardless of polycarbonate level in the blend, occurs in the case of the SAN having 26.7% AN. This is in the same range where maximum adhesion was noted. An analogous presentation of the Rheovibron data is shown in Figure 6. While the plots in Figures 5 and 6 differ in some details, which are perhaps beyond the sensitivity of the measurements, the conclusion is exactly the same—the greatest shift in  $T_g$  occurs for the SAN containing 26.7% AN.

The second method attempts to assess more directly the amount of polycarbonate dissolved in the SAN phase and vice versa. It assumes the Fox equation would describe the  $T_g$ -composition relation if polycarbonate were fully miscible with the styrenic polymer. This is an ad hoc but useful assumption since a relation with no adjustable parameters is needed for such an analysis. By inverting the Fox equation

$$\frac{1}{T_g} = \frac{W_{\text{SAN}}}{T_{g,\text{SAN}}} + \frac{1 - W_{\text{SAN}}}{T_{g,\text{PC}}} \quad (2)$$

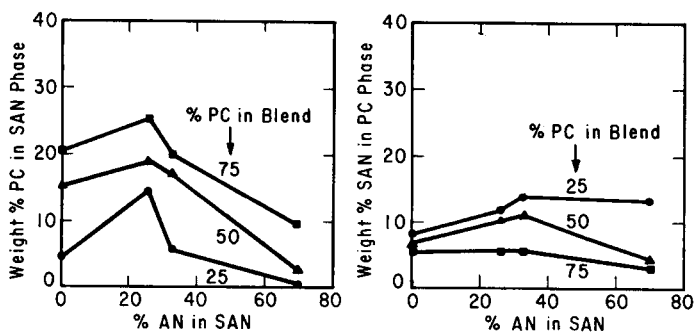


Fig. 7. Phase compositions for blends with SAN copolymers as computed from DSC data.

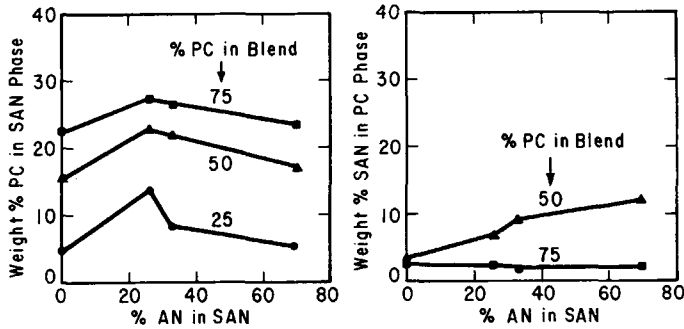


Fig. 8. Phase compositions for blends with SAN copolymers as computed from Rheovibron data.

the weight fraction of SAN in either the styrenic or polycarbonate phase can be computed

$$W_{\text{SAN}} = T_{g,\text{SAN}}(T_{g,\text{PC}} - T_g) / T_g(T_{g,\text{PC}} - T_{g,\text{SAN}}) \quad (3)$$

by inserting the  $T_g$  observed for the phase of interest. The results of these calculations are given in Figure 7 for the DSC data and in Figure 8 for the Rheovibron data. The estimated solubilities are plotted vs. the AN content of the SAN at a fixed level of polycarbonate in the blend. Based on this analysis, it appears that polycarbonate is more soluble in SAN than SAN is in polycarbonate. The latter results, shown on the right in both Figures 7 and 8, form a difficult pattern of behavior to interpret, but the former, shown on the left in each case, give a clear picture. Maximum solubility, for the materials tested, occurs when the SAN contains 26.7% AN.

### INJECTION-MOLDED BLENDS

Blends formed into test bars by injection molding as described earlier were examined for phase morphology, heat distortion behavior, and tensile mechanical properties. The results are given in this section.

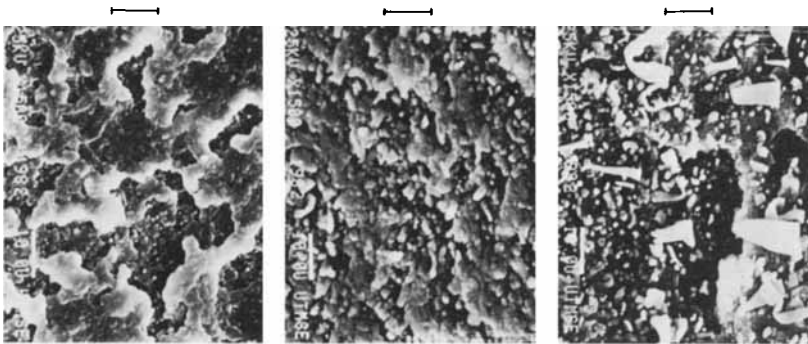


Fig. 9. Scanning electron photomicrographs of fracture surfaces of injection-molded polycarbonate/polystyrene blends. Percent polycarbonate: 80 (left), 40 (center), and 20 (right). Marker indicates 10  $\mu\text{m}$ .



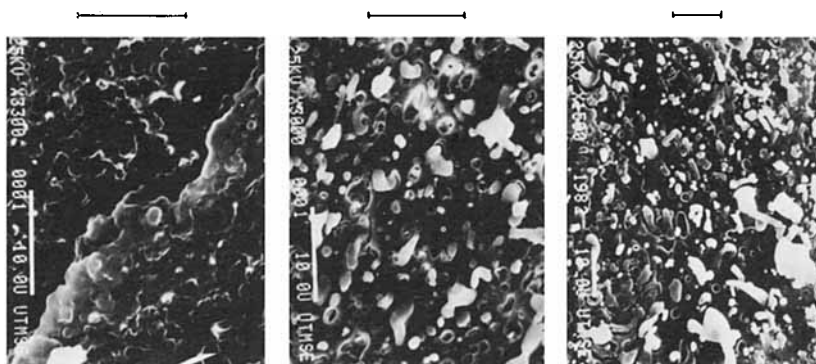


Fig. 10. Scanning electron photomicrographs of fracture surfaces of injection-molded polycarbonate/SAN-3 blends. Percent polycarbonate: 80 (left), 60 (center), and 40 (right). Marker indicates 10  $\mu\text{m}$ .

### Morphology

For examination of phase morphology, injection-molded Izod bars were cooled by immersion in liquid nitrogen and fractured while still cold by an Izod impact tester. These fracture surfaces were coated with gold and viewed end on by a Joel 35C scanning electron microscope using a beam voltage of 25KV. Figure 9 shows photomicrographs for various polycarbonate/polystyrene blends while Figure 10 shows similar pictures of the structure of polycarbonate/SAN-3 blends. Phase contrast is somewhat poor for the polystyrene blends (Fig. 9), and the topology of the fracture surface is rather irregular; however, close examination of the original photomicrographs clearly reveals cylindrical fibrils of one polymer in a continuous matrix of the other. For the blend containing 20% polycarbonate, the fibrils are somewhat less circular in cross section and tend to draw out on fracture. The same features are also apparent in Figure 10 for polycarbonate/SAN blends.

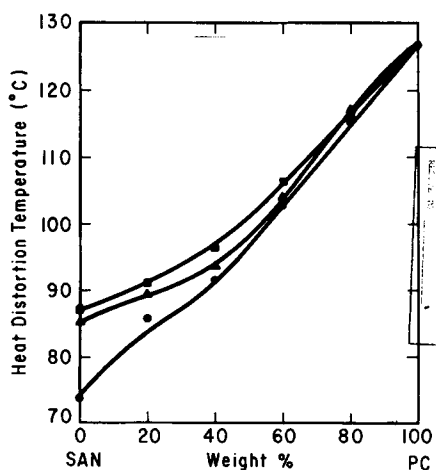


Fig. 11. Heat distortion temperature for blends of polycarbonate with SAN copolymers. % AN: (●) 0; (■) 24.8; (▲) 32.3.

Blends containing other SANs had quite similar morphologies. In every case, these injection-molded blends consist of a continuous phase of one polymer with the other polymer present in fibrillar form. The fibrils are typically  $1\ \mu\text{m}$  or less in diameter. These structures are the result of the flow patterns which exist in injection molding.

### Heat Distortion Temperature

Heat distortion temperatures were measured according to ASTM D-648 on  $0.125 \times 0.5$  in. Izod bars at 264 psi for selected blends at Borg-Warner Chemicals through the courtesy of R. L. Jalbert and A. T. Watson. The results are shown in Figure 11. The primary differences seen here are the result of the variation in heat distortion temperature for the different pure styrenic materials. Their blends with polycarbonate show values slightly less than additive. For this particular type of test, the effective thermal resistance of styrenic polymers, including ABS, can be significantly improved by blending with polycarbonate, even though these components are not completely miscible.

### Tensile Properties

Dog bone bars were tested in tension with an Instron at a crosshead speed of 0.2 in./min for modulus, using an extensometer with a 1-in. gauge, and 2 in./min for yield and failure properties. For the latter, strain was computed from crosshead travel and an effective gauge length.

Results are shown in Figures 12–14 for specimens tested after injection molding without further treatment. The moduli fall slightly below the additive line as shown in Figure 12 regardless of the type of styrenic polymer employed. The curve for polystyrene is similar in form to that reported by Kunori and Geil.<sup>18</sup> As expected, the SAN copolymers have somewhat greater stiffness than polystyrene. Figure 13 shows the yield on ultimate stress at failure for polycarbonate blends with four different styrenic polymers. Those having roughly 50% or more of the styrenic material did not yield,

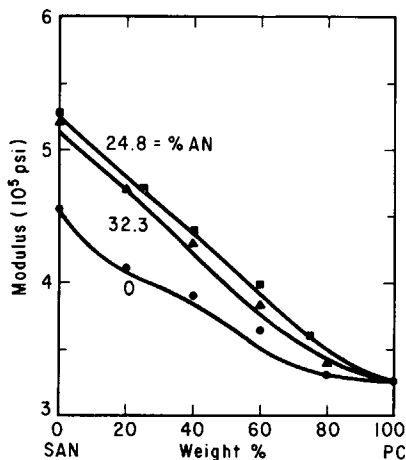


Fig. 12. Modulus of blends of polycarbonate with SAN copolymers.

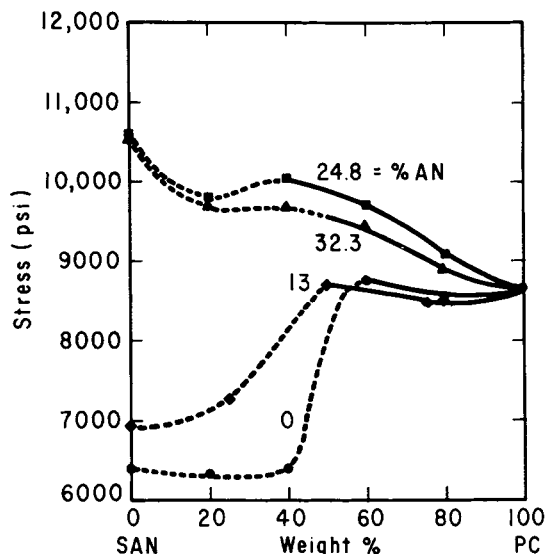


Fig. 13. Stress at yield or failure for blends of polycarbonate with SAN copolymers. (—) yield stress; (---) ultimate stress (no yield).

whereas those containing approximately 50% or more of polycarbonate did. Addition of SAN-2 and SAN-3 to polycarbonate raised the yield strength,<sup>19</sup> whereas the yield strength was effectively unchanged from that of pure polycarbonate when SAN-6 and polystyrene were added. When the latter two were the major components, failure strengths were significantly lower than the yield strength of polycarbonate which is in contrast to the results for SAN-2 and SAN-3 as the major components. Polycarbonate blends with the SAN having 24.8% AN were the strongest at all proportions. Unfortunately, the limited supply of SAN-4 did not permit preparation of injection molded blends from it.

The percent elongation at failure is shown in Figure 14 for each of these

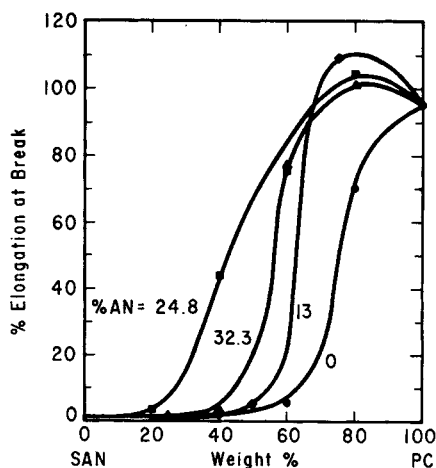


Fig. 14. Percent elongation at failure for blends of polycarbonate with SAN copolymers.

blend systems. There is a slight maximum at about 70–80% polycarbonate in every case except for blends with polystyrene. There is a precipitous decline in ductility for each blend system that occurs at a different polycarbonate content depending on the % AN of the styrenic polymer. Interestingly, blends with the SAN containing 24.8% AN remain ductile to lower polycarbonate contents than do blends made from styrenic polymers containing either more or less acrylonitrile. For this copolymer, blends containing as little as 40% polycarbonate necked and drew out to about 40% elongation, whereas at this polycarbonate level blends containing all of the other styrenic polymers failed in a brittle manner. This fact was regarded as highly significant and was explored further. Owing to orientation effects, glassy polymers are usually more ductile when prepared by injection molding than by other techniques which do not generate so much orientation such as compression molding.<sup>20</sup> To learn whether the high ductility noted above was simply a consequence of this effect, some samples were annealed at 170°C for 30 min in a cavity having the shape of the dog bone specimen. As described earlier,<sup>20</sup> this procedure permits relaxation of orientation without loss of the dog bone shape. Figure 15 compares the stress-strain diagrams for two blends thermally annealed in this way both of which contained 40% polycarbonate. The blend based on the SAN containing 32.3% AN failed in a brittle fashion as the corresponding blend did before annealing; however, for the case of the SAN containing 24.8% AN, necking occurred. While the ultimate elongation in the latter is less than that prior to annealing, this blend still has a very ductile character. This remarkable difference may be attributed to the superior interaction with polycarbonate of SANs having AN contents in the 25–27% range as seen in Figures 2 and 5–8.

Notched Izod impact strengths for the various blends were measured on 0.125 in. bars with the results shown in Figure 16. In every case, there is a marked loss of impact strength on adding styrenic materials to polycar-

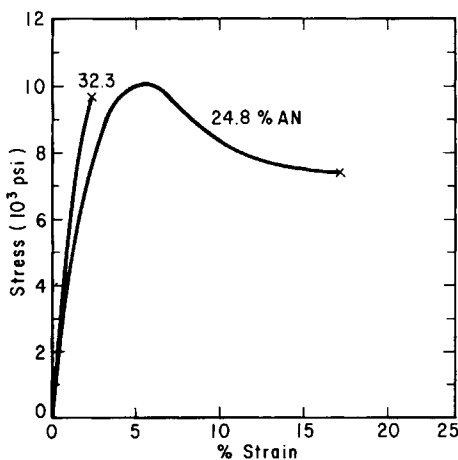


Fig. 15. Comparison of stress-strain diagrams for 40% polycarbonate blends with two different SAN copolymers. Both samples had been annealed, as described in the text, to relax orientation induced during injection molding.

bonate; however, blends with the SAN containing 24.8% acrylonitrile show the best retention of toughness. Data for the 13% AN copolymer are not shown for clarity. For polystyrene, the impact strength actually goes through a minimum as found by Kunori and Geil,<sup>18</sup> although this feature is somewhat masked by the linear ordinate used in Figure 16. Keskkula and Pettis<sup>19</sup> have examined the detailed shape of the impact strength vs. composition curve for blends containing 75% polycarbonate or more using an SAN having 30% acrylonitrile. They found the decline not to be as precipitous as suggested in Figure 16; however, our curves in this region were arbitrarily drawn and cannot be compared since no data were obtained between 80 and 100% polycarbonate. The central point in Figure 16 is the optimum impact strength, which is obtained for the SAN containing 24.8% acrylonitrile.

### SUMMARY AND CONCLUSIONS

It has been demonstrated by lap shear experiments with laminates that adhesion between polycarbonate and styrene/acrylonitrile copolymers goes through a sharp maximum when the acrylonitrile content of the latter is in the range of 25–27% by weight. While all of these copolymers form two phase blends with polycarbonate, glass transition observations suggest some solubility of polycarbonate in SAN and vice versa. The maximum extent of this solubility occurs for copolymers having approximately the same acrylonitrile contents which give maximum adhesion. We believe that this partial solubility is responsible for the adhesion results observed. The mechanical properties of polycarbonate/SAN blends are also best when the SAN contains acrylonitrile levels in this same range. Assuming that interfacial adhesion in blends will parallel that observed for laminates, then the better mechanical properties may be directly attributed to the superior adhesion at the optimum acrylonitrile level.

The remaining question is why should there be an optimum acrylonitrile content for maximum partial solubility. We believe that the answer to this question is basically the same as why certain copolymers form completely

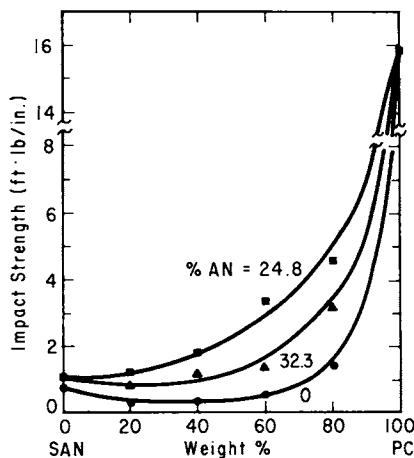


Fig. 16. Notched Izod impact strength for blends of polycarbonate with SAN copolymers.

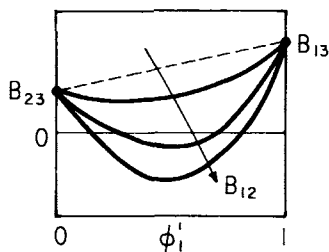


Fig. 17. Schematic of interaction energy density  $B$  as predicted by binary interaction model, eq. (3). Dashed line is for  $B_{12} = 0$ . Arrow indicates trends as  $B_{12}$  becomes a larger positive number.

miscible blends with other polymers when the homopolymers made from the two monomers of the copolymer are not similarly miscible.<sup>21</sup> A good example of the latter is blends of poly(methyl methacrylate) with SAN copolymers. These mixtures may be completely miscible when the acrylonitrile content of the latter is in the approximate range of 9–28% by weight,<sup>22</sup> while copolymers outside this range are not completely miscible with poly(methyl methacrylate). Other examples of this effect may also be cited.<sup>21</sup> One explanation for this has been offered in terms of a simple binary interaction model.<sup>21</sup> This model states that the interaction energy density  $B$  between a copolymer, composed of monomers 1 and 2, and another polymer, species 3, is related to the three possible binary interaction parameters  $B_{ij}$  according to the following:

$$B = B_{13}\phi'_1 + B_{23}\phi'_2 - B_{12}\phi'_1\phi'_2 \quad (4)$$

where  $\phi'_1$  and  $\phi'_2$  are the volume fractions of monomers 1 and 2 in the copolymer. In the typical case, all binary pairs will mix endothermically, i.e., each  $B_{ij}$  in eq. (4) is positive. Consequently,  $B$  vs.  $\phi'_1$  tends to exhibit a minimum as shown in Figure 17. In the case

$$B_{12} > \left( \sqrt{B_{23}} + \sqrt{B_{13}} \right)^2 \quad (5)$$

$B$  dips below the line  $B = 0$  over a certain range of  $\phi'_1$  values, as shown in Figure 17. In this region, polymer 3 will be miscible completely with copolymers of 1 and 2. This would explain the miscibility of PMMA with certain SAN copolymers. For  $B_{ij}$  values where eq. (5) is not fulfilled,  $B$  will simply form a minimum at a certain  $\phi'_1$  but remain positive. This minimum would correspond to a maximum in the extent of partial miscibility between polymer 3 and the copolymer. In terms of this model, the present results would be explained, at least qualitatively, by such a minimum in  $B$  for polycarbonate and SAN mixtures when the AN content is about 25–27% on a weight basis.

It is important to note that the range of acrylonitrile contents, i.e., 25–27%, where SAN copolymers have maximum adhesion with polycarbonate corresponds rather closely with the azeotropic composition for this system and to the composition of many commercial SANs. These same copolymers

are often used as the matrix phase for ABS materials. This interaction and the resulting adhesion is believed to be one of the reasons why ABS can be successfully alloyed with polycarbonate. It follows that the best types of ABS for this purpose would be ones whose matrix phase have this optimum AN content.

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