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Melt Viscosities and Mechanical Properties of PC/SAN Blends

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Melt viscosities and mechanical properties of polycarbonate (PC), styrene-acrylonitrile copolymer (SAN), and their blends were studied. PC and SAN were melt blended at different compositions using a single screw extruder. Melt viscosities were measured with a capillary theometer. The melt viscosity test showed that increasing the PC content increases the viscosity of the blends, and melt viscosities of blends are intermediate between the values of PC and SAN. Standard mechanical property tests were performed on injection molded ASTM test specimens. Tensile strength and modulus increase but elongation at break decreases when the SAN content increases. Notched Izod impact strength decreases rapidly with the addition of SAN into PC. Unnotched impact strength is higher than notched impact strength and is less sensitive to the addition of SAN.

Keywords: Polycarbonate; SAN; viscosity; blends; mechanical properties

INTRODUCTION

The development of new plastics does not always require new molecules to meet the needs of new materials. Blending of existing polymers can be implemented more rapidly and economically than the development of new polymers. Polycarbonate (PC) and styreneacrylonitrile copolymer (SAN) have long been used as commercial polymers, but PC is an expensive polymer, which is also restricted in some applications because of its low processibility and high viscosity. Adding SAN to PC results in a blend that will be easier to process,

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having a high modulus and will provide a less expensive material than PC alone. Several studies have been conducted on various properties of PC/SAN blends [1-9]. The present work attempts to evaluate the rheological and mechanical properties of these blends.

The relative reactivity of styrene and acrylonitrile, as indicated by the difference in reactivity ratio, makes the copolymerization subject to a high degree of composition drift and inhomogeneity if the drift is not compensated for by an adjustment of monomer ratio. In order to avoid composition drift, SAN copolymers are often manufactured near the azeotropic point, where monomer and polymer have the same composition (0.62 mole fraction or 76 wt% styrene). Based on a three dimensional solubility parameter calculation it has been pointed out by Huang and Wang [1] that the optimum composition for the best miscibility between SAN and PC is about 0.25 mole fraction of acrylonitrile (AN). This number is near the composition of commercial SAN and accounts for the success of some PC/SAN and PC/ABS blends.

Keitz et al. [2] studied several properties of PC/SAN blends using SANs with different AN content. The lap shear adhesion, tensile properties, and notched Izod all show that the adhesion between PC and SAN copolymers exhibits a maximum when the SAN contains 25 wt% AN. While all of blends showed two-phase morphology in scanning electron micrographs, results of differential scanning calorimetry (DSC) and Rheovibron suggest partial miscibility of one polymer in the other. The solubility of PC in SAN is at a maximum when the SAN copolymer has an AN content in the same range causing the maximum adhesion. It is believed that this partial solubility is responsible for the adhesion results observed. The mechanical properties of PC/SAN blends are also best when the SAN contains AN level in the same range.

Thermal analysis of polymer blends has shown that a miscible polymer blend will exhibit a single glass transition temperature (T_g) between the T_gs of the pure components, while for partially miscible systems the T_gs approach each other, but remain separated. Kim and Burns [3] studied miscibility of PC/SAN blends and PC/ABS blends by determining the T_g and the change of specifc heat at T_g . Two glass transition regions are founded in the blends, and PC and SAN are reported to be partially miscible. The T_{gs} of PC in PC/SAN blends are observed to decrease with an increase in the SAN weight fraction. From the measured T_g and specific heat increments at the T_g , SAN appears to dissolve more in the PC-rich phase than does PC in the SAN-rich phase. Similar results are observed in the PC/ABS blends.

Callaghan *et al.* [4] reviewed studies on miscibility of PC/SAN and made thermodynamics analysis based on Flory-Huggins and Sanchez-Lacombe equation of states theories. They prepared low molecular weight PC and SAN for critical molecular weight analysis in which the molecular weight of PC and SAN were reduced until their blends become miscible. The results also predict an optimum AN composition at 25 wt% for miscibility.

Skochdopole *et al.* [5] studied the properties and morphology of some injection-molded PC/SAN blends. The mechanical and thermal properties of PC/SAN blends are correlated with the morphology of the blends. The notched Izod impact toughness of the PC falls off rapidly with the addition of 10 wt% SAN or more. The dart impact toughness remained high up to 30-40 wt% SAN and dropped rapidly above this SAN concentration. The tensile strength and modulus have more linear dependence on composition. The results of T_g by DSC measurement show the presence of two phases over the entire concentration range and a small solubility of each in the other.

Takahashi *et al.* [6] studied the rheology of the molten PC/SAN blend at extremely high shear rate up 10^7 s^{-1} . The first Newtonian region, non-Newtonian region, and the second Newtonian region take place in the compound systems in a way very similar to those in the parent polymers, PC and SAN. Takahashi *et al.*, also studied the influence of high shear rate processing on the compatibility of PC and SAN blends. Incompatible blends of SAN 30%/PC 70% were extruded with an extremely high shear rate up to 10^7 s^{-1} for several runs. The apparent volume fraction of the spherical minor constituent estimated from the microscopic photographs decreases with increasing shear rate. The decrease of the fractional volume of SAN must be dispersed in PC like a compatible constituent. SEM observation revealed that dimple fracture of microsize took place on the SAN sphere in the PC matrix. And at the bottom of the dimple, small particles of size 50 nm that would be composed of PC were present. This

suggests that PC is also dispersed in SAN like a compatible constituent in the blends exposed to the extremely high shear rate. The fraction of the spherical minor constituent was decreased with the repeated runs. After the fifth run, the blend appeared uniform and structureless. This also suggests that the constituents are forced to be mutually compatible. The dynamic loss tangent gave two peaks with one corresponding to that of SAN and another attributed to a modified PC. The latter shifted to lower temperatures with each additional extrusion run. These also show that some SAN is mixed into PC in a compatible form. This study concluded that the compatibility of PC/SAN blends is enchanced in extremely high shear rate processing.

McLaughlin [7] studied the influence of microstructure on the dynamic mechanical behavior of the blend of PC with SAN. He compared the torsional response of both injection and compression molded samples. The samples from compression molding show two peaks and a shift in T_g of PC which indicates partial miscibility. In injection molded samples a separated tan δ shows up between peaks of SAN and PC. This is a spurious loss peak appearing in multilayer structure. Microstructural characterization of the materials was accomplished by transmission electron microscopy and scanning electron microscopy. Microstructural pictures confirm a difference between microstructure of compression molded samples and injection molded samples. For the injection molded samples the author also reported an increase of ultimate elongation when PC concentration exceeds 50%.

The effect of phase morphology on properties of PC/SAN blends was reported by Quintens *et al.* [8]. A 70/30 and a 60/40 PC/SAN blend were intensively melt mixed and injection molded then subject to an annealing process at 200°C up to 18 min. The size of phase is represented by Chalkey parameter, which is related to the relative surface fraction. After annealing the 60/40 blend changes from a fine dispersion structure to a coarse co-continuous network. The 70/30 blend changes from a co-continuous structure to a dispersion of SAN in a PC matrix. Elongation at break of both blends was found to decrease when phase size increases during the annealing process and the samples change from ductile to brittle. However, the tensile strength remains constant for annealed samples.

EXPERIMENTAL

Materials

Lexan 141 grade polycarbonate was obtained from General Electric Company. Kustran DN-79 styrene-acrylonitrile copolymer was obtained from Monsanto Company. Its AN content is 32 wt% and Mw is 95,000. Three blends of SAN and PC were compounded. The weight percentage compositions of the blends were 25/75, 50/50, and 75/25, respectively.

Sample Preparation

Mixture of plastics pellets were drum mixed and dried for four hours at 93°C (200°F) before extrusion. A Brabender single screw extruder was used for compounding. The machine has a screw with a length of 16.5 inches and an effective L/D of 22:1. The screw is a square pitch type with a compression ratio of 2.95. The processing temperature was controlled at the minimum temperature (280°C) that would melt both resins yet would avoid degradation. The extrudates were cooled in a water bath then palletized for injection molding and melt viscosity measurements. Injection molding was conducted on a Negri Bossi & Company V 2/78, 18-ton capacity injection molding machine with a 0.67 oz shot size. ASTM D638 type I tensile bars were molded. All materials were dried for four hours at 93°C before being molded. The mold temperature was 75°C.

Viscosity Measurement

A Gottfert Rheograph 2001 is used for measurement of melt viscosities. A capillary die with a length of 20 mm and 1 mm diameter was used. The barrel has a 15 mm diameter, and the maximum pressure is 2000 bars. Several shear rates were selected and stresses at steady state were measured. From the flow rate of melts through the die at different stresses the Rabinowitsch correction was made to obtain corrected viscosities and corrected shear rates. Measurements were taken at temperatures between 240°C to 280°C and at shear rates between 50 sec^{-1} to 4000 sec^{-1} .

Measurement of Mechanical Properties

Tensile properties were measured on an Instron machine, Model 6025, which is equipped with a maximum load cell capacity of 10,000 pounds. ASTM D638 procedures were followed. The jaw speed used in this testing was 2.0 in/min. The flexural properties were tested in accordance with ASTM D747 with a 6 kg-cm capacity Tinus Olsen stiffness tester of the cantilever beam type. This method provides a means of determining stiffness for plastics by measuring the force and angle of the bend of the cantilever beam.

The impact resistance test was determined in accordance with ASTM D256A. The specimen is held as a vertical cantilever beam and is broken by a swing of the pendulum from the centerline of the notch and on the same face as the notch. The unnotched method is similar to notched test, except that the specimen is reversed in the vise of the machine 180° to the usual striking position, such that the pendulum impacts the specimen on the face opposite the notch. An Izod Impact Tester, Model No. TMI 43-1, made by Testing Machine Inc., was used.

RESULTS AND DISCUSSION

Melt Viscosity

The viscosity of PC at different temperature is shown in Figure 1 and the viscosity of SAN is shown in Figure 2. These curves illustrate that temperature and shear rate have an important effect on the melt viscosity of those polymeric melts, just as they do with other polymeric materials. Elevated temperatures facilitate molecular uncoiling and disentanglement, thus lowering the melt viscosity of polymers.

The rheology curves for PC, SAN, and their blends at shear rate $100 \sec^{-1}$ and $1000 \sec^{-1}$ are shown in Figures 3 and 4 for temperature of 240°C and 280°C, respectively. These results show that SAN has a lower melt viscosity than PC. An increase in PC content leads to an increase of viscosity but the increase in not close to a linear one. An addition of 25 wt% of SAN to PC reduced the melt viscosity by more than 50%. For example, while the melt viscosity of PC at shear rate $100 \sec^{-1}$ and 240°C is 3960 Pa-sec, for the blend of 75 wt% PC the viscosity became 1420 Pa-sec at the same condition. In order to



FIGURE 1 Melt viscosity of polycarbonate at 240°C (\circ), 260°C (+), and 280°C (Δ).



FIGURE 2 Melt viscosity of SAN at 240°C (\circ), 260°C (+), and 280°C (Δ).



Weight Fraction of PC

FIGURE 3 Melt viscosity of PC, SAN, and their blends at 240°C. (•): Shear rate = 100 sec^{-1} , (o): Shear rate = 1000 sec^{-1} .



FIGURE 4 Melt viscosity of PC, SAN, and their blends at 280°C. (\blacktriangle): Shear rate = 100 sec⁻¹, (\triangle): Shear rate = 1000 sec⁻¹.

decrease the melt viscosity of PC for such processes as blow molding or injection molding, an addition of SAN in the range around 25 wt% would create a noticeable effect. The viscosity curves in Figure 3 appeared to be similar to each other. The distance between two curves represents decrease of viscosity due to shear rate increase from 100 sec^{-1} to 1000 sec^{-1} . This distance is smaller in Figure 4. The Non-Newtonian behavior is reduced as temperature increases from 240°C to 280°C.

The melt viscosity of the blend with 75 wt% SAN is very close to that of pure SAN. One explanation on the deviation of melt viscosity from linear rule is that SAN dissolves more in the PC-rich phase than PC dissolves in the SAN-rich phase. This has been established by observing change of T_{gS} of PC and SAN from results of DSC study [2-5]. Dissolution of SAN into PC can be considered having plasticization effect in the PC phase, while minor PC in low PC blends simply disperses into the SAN phase and does not increase viscosity until the PC itself becomes more or less a continuous phase.

Tensile and Flexural Properties

Mechanical properties are reported in Table I. Of the various measurements of mechanical properties conducted on materials, stress-strain behavior in tension is one of the most common. Tensile properties are determined by pulling a sample at a constant rate. The resulting stress is measured and recorded as a function of strain. Tensile strength is calculated from the load at yield or break. The tensile strength of PC is 8870 psi at yield and is 8300 at break. For SAN there is no yield and the tensile strength is 10400 psi. SAN has higher tensile strength and tensile modulus than PC. The tensile strength at break of blends increases with increasing SAN content. For blends with less than 50 wt% PC there were no true yielding phenomena observed. For elongation at break a steep decrease is observed when SAN is added into PC. At 50 wt% and higher content of SAN the elongation is essentially the same as SAN.

The tensile properties of PC and several SANs with different AN content were reported by Keitz *et al.* [2]. Tensile modulus of polystyrene and SAN is higher than PC while tensile strength of polystyrene and SANs with low AN content is lower than PC. The tensile strength

Properties	Composition (PC/SAN)				
	100/0	75/25	50/50	25/75	0/100
Tensile Strength At Yield (psi)	8870	10220	no yield	no yield	no yield
Tensile Strength At Break (psi)	8300	7570	10330	10400	10400
Elongation at Yield (%)	10.6	8.8	no yield	no yield	no yield
Elongation at Break (%)	139	54	7.9	4.8	4.5
Flexural Modulus (Kpsi)	131	155	182	193	201
Notched Impact (ft-1bf/in)	18.4	0.7	0.3	0.3	0.24
Unnotched Impact (ft-1bf/in)	38	33	7.0	3.0	2.4

TABLE I Mechanical properties of PC/SAN blends

of PC/SAN blends shows a different behavior depends on AN content of SANs.

However, the elongation behaviors are similar. They remain roughly constant up to 25 wt% SAN then increase quickly around 50 wt%. This result corresponds quite well with the composition range where PC remains as a continuous phase and would be expected to have its good elongation dominate the blend performance.

The results of flexural modulus show that flexural modulus increases when SAN content increases. As SAN content increases the modulus increases quickly until SAN becomes the continuous phase around 50 wt%. After this range the increase of modulus becomes slower.

Impact Resistance Testing

Impact strength is the energy required to break a material using very rapid loading rates. The values for a given polymer can vary over a wide range because impact strength is sensitive to many variables. These include crystallite size, molecular weight, molded in stress, and the presence of fillers, reinforcing agents, and cracks.

The notched Izod impact strength fell off rapidly when small amounts of SAN were added to PC. Keitz *et al.* [2] and Keskkula and Pettis [9] showed the drop-off in notched Izod impact strength when even 10% SAN is added to PC. Kunori and Geil [10] reported a similar drop-off in notched Izod impact strength with the addition of only 2% polystyrene into PC. This behavior can be attributed to the initiation of cracks in the brittle polystyrene or SAN phase that carry on into the PC phase. Such behavior discourages shear yielding of the PC in the presence of a sharp notch. In the notched specimen cracking takes place in the immediate neighboring of the notch followed by brittle failure. Unnotched impact results are higher than those of notched impact are. They are also less sensitive to the addition of SAN. The blend with 25 wt% SAN maintains a higher unnotched impact value and the value drops around 50 wt% of SAN. Skochdopole *et al.* [5] reported a similar trend for dart impact toughness of PC/SAN blends. Since dart impact test does not have initial cracks on the surface a result similar to the unnotched impact test indicates that a same mechanism is involved.

CONCLUSIONS

Several blends of PC and SAN were prepared using a single screw extruder. Their melt viscosity, tensile, flexural stiffness, and Izod impact properties were tested. Among the melt viscosity of PC, SAN, and their blends, PC had the highest viscosity, and increasing the SAN content of the blends reduces the viscosity. Tensile strength increases with increasing SAN content. The elongation decreases with increasing SAN content. Flexural modulus resembles tensile strength, being increased with the increasing SAN content. For notched Izod impact test, the impact properties of blends decreases quickly at 25 wt% of SAN to a value close to pure SAN. For the unnotched Izod impact test, the results were higher than the notched test for all compositions, and the blend with 25 wt% retains a high value in unnotched impact test as compared to notched test.

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