

Blends of Imidized Acrylic Polymers with SAN Copolymers and with PVC

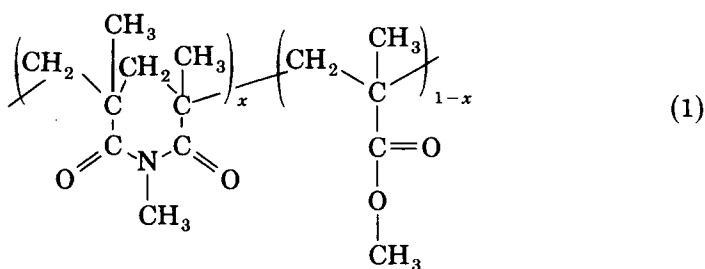
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Synopsis

A series of imidized acrylic polymers of varying structural composition generated by reaction of methylamine with poly(methyl methacrylate) were blended with a range of styrene/acrylonitrile or SAN copolymers (0–33% AN) and with poly(vinyl chloride). On the basis of glass transition behavior determined by differential scanning calorimetry, some but not all imidized acrylic structures were found to be miscible with PVC and with SAN copolymers within a limited window of AN levels. Acid functionality in the imidized acrylics appears to hinder their miscibility with SAN rather significantly and with PVC to a lesser extent. Miscible SAN blends showed lower critical solution temperature behavior whereas miscible blends with PVC did not up to the highest attainable temperatures. The composition factors that influence the phase behavior are described and interpreted in terms of possible mechanisms.

INTRODUCTION

A series of thermoplastic polymers derived from reaction of methylamine with poly(methyl methacrylate) (PMMA) have recently become commercially available. These materials can be made by a number of processes, including that stated in the patent to Rohm and Haas Co.,¹ and in their simplest form they may be thought of as a methyl glutarimide/methyl methacrylate copolymers, i.e.,



Other variations of interest include materials that may be described as terpolymers where the third unit is methacrylic acid. The latter may also include structures where adjacent units of this type exist in the anhydride form. The imidized acrylics have the clarity and processability of acrylics, but they also offer higher heat distortion temperature, modulus, and chemical resistance. This study was motivated by the possibility, as suggested by recent

patents,²⁻⁴ for improving the thermal, barrier, and optical properties of other polymers that may form miscible blends with the imidized acrylics.

Candidate polymers for blending with the imidized acrylics include those that form miscible blends with PMMA and other polymers having a high concentration of carbonyl groups [see eq. (1)], as this seems to be a factor related to achieving the favorable interaction needed. This article focuses on two polymers meeting these requirements, viz., styrene/acrylonitrile copolymers or SAN and poly(vinyl chloride) or PVC. A great deal of recent work has shown that, for a limited range of acrylonitrile contents, SAN copolymers form miscible blends with PMMA⁵⁻⁷ and other carbonyl-containing polymers.⁸⁻¹¹ Of particular relevance here is the recent report that copolymers of MMA and *N*-phenylmaleimide form miscible blends with an SAN copolymer.¹² While the interactions in blends are complex, recent theories¹³⁻¹⁵ have shown that repulsive interactions between the various monomeric units in such copolymers can assist miscibility to the point where selected copolymers form miscible blends with other polymers even when the corresponding homopolymers do not. There is also a great deal of literature showing that PVC is miscible with a number of carbonyl-containing polymers.¹⁶⁻¹⁸ Some reports suggest that PMMA is miscible with PVC depending on the tacticity of the acrylic.^{19,20} While there is some dispute about this pair^{21,22} and more definitive work is needed to clarify all of the issues, it seems clear that significant miscibility does exist.

This article examines the phase behavior of blends of SAN copolymers (containing 0-33% AN) and of PVC with a number of imidized acrylics having a broad range of functionality, i.e., with varying imide to MMA ratio and with potentially reactive acid and anhydride units. Miscibility was judged on the basis of whether a single glass transition was found or not by differential scanning calorimetry. For some blends, optical cloud point observations indicative of lower critical solution temperature (LCST) behavior provided a useful qualitative guide to the net interactions involved.

MATERIALS AND EXPERIMENTAL PROCEDURES

Table I lists some of the various imidized acrylic polymers used in this study along with pertinent information about their compositions and molecular weights. The materials with a commercial designation are available from the Rohm and Haas Co., while those without such a designation represent experimental materials. The second column in Table I gives a designation scheme used here that has the following meaning. The three digit numerical code indicates the measured Vicat temperature (°C) for each sample. The letter code C designates materials that may be thought of as copolymers having the structure shown by eq. (1). The columns labeled imide and MMA indicate the proportions of these two types of repeat units in weight percent. The letter code A designates the more complex structures mentioned earlier also having free acid or anhydride repeat units, and the columns so labeled indicate their weight percents. In two instances, the letters H and L are used to distinguish high and low molecular weight versions of copolymers that are essentially identical in composition.

The wide range of SAN copolymers used here is described in Table II with information about their molecular weights where available. The PVC used

TABLE I
Imidized Acrylic Polymers Used in This Study

Commercial designation	Designation used here	Wt%					$\bar{M}_n \times 10^{-3}$	$\bar{M}_w \times 10^{-3}$
		Imide	Free acid	Anhydride	MMA			
Paraloid EXL 4245	145C-H	72	0	0	28	64	135	
None	160C	85	0	0	15	59	130	
Paraloid HT 510	145C-L	71	0	0	29	31	81	
None	140A	53	2	1	44	74	187	
Paraloid EXL 4150	150A	72	3	1	24	66	140	
None	160A	78	4	1	17	63	136	
Paraloid EXL 4170	170A	85	5	Nil	9	61	134	
None	180A	91	6	Nil	3	60	135	

was obtained from the Diamond Shamrock Corp. It was found to have $\bar{M}_n = 99,000$ and $\bar{M}_w = 213,000$ by gel permeation chromatography, where the calibration was based on PMMA standards.

Blends of the various imides and the various SANs were prepared by either solution casting or melt blending. In the case of solution casting, the polymers were carefully weighed in the appropriate proportions and simultaneously dissolved in CH_2Cl_2 to give solutions containing approximately 3–5% by weight of total polymer. The solutions were poured into aluminum pans and covered with aluminum foil. Small holes were punched in the foil to retard the initial rate of solvent evaporation. These solutions were allowed to dry for 2 days under a hood at room temperature. They were then placed in a vacuum oven and dried for 3 days at 85°C followed by 3 days at a temperature just above the glass transition temperature of the imidized acrylic. This ensured complete evaporation of the solvent and avoided phase separation for some mixtures that exhibited cloud points at temperatures close to their glass transition temperatures. A Brabender Plasti-Corder equipped with a 50 cm^3 sample chamber was used for melt blending at 200°C and 40 rpm.

Blends of the imidized acrylics with PVC were prepared by the same procedure with the following exceptions. A stabilizer for the PVC (a lead-based compound designated as Halstab 31 from Hammond Lead Products, Inc.) was added to the solutions in the amount of approximately 0.1% by weight of polymer. The solvent used was tetrahydrofuran (THF). After initial drying at 85°C , the solutions were heated to 180° for only 2 h to avoid dehydrochlorination of the PVC. Glass transition analysis of the blends indicated that this was sufficient to remove all of the solvent.

Two approaches were used to assess the phase behavior of blends of the various imidized acrylics listed in Table I with the range of SAN copolymers shown in Table II and with poly(vinyl chloride). First, thermal analysis was used to determine whether these mixtures exhibited one or two glass transition temperatures. Second, blends that were found to have a single glass transition temperature were examined visually for any indications of phase separation upon heating, i.e., LCST behavior.

Thermal analysis was carried out using either a Perkin-Elmer DSC-2 or DSC-7 (both were equipped with a thermal analysis data station) at a heating rate of $20^\circ\text{C}/\text{min}$ with samples weighing 5–15 mg. Three temperatures char-

TABLE II
Poly(styrene-co-acrylonitrile) Polymers Used in This Study

(Wt %) AN	Designation	Source	$\bar{M}_n \times 10^{-3}$	$\bar{M}_w \times 10^{-3}$
0.0	PS	Cosden Oil and Chemical Co.	100	350
5.7	SAN 5.7	Asahi Chemical Industry Co., Ltd.	87.7	212
9.5	SAN 9.5	Asahi Chemical Industry Co., Ltd.	NA ^a	NA ^a
11.5	SAN 11.5	Asahi Chemical Industry Co., Ltd.	NA ^a	NA ^a
13.5	SAN 13.5	Asahi Chemical Industry Co., Ltd.	NA ^a	NA ^a
14.7	SAN 14.7	Asahi Chemical Industry Co., Ltd.	82.7	181
15.5	SAN 15.5	Asahi Chemical Industry Co., Ltd.	NA ^a	NA ^a
19.5	SAN 19.5	Asahi Chemical Industry Co., Ltd.	NA ^a	NA ^a
25.0	SAN 25	Asahi Chemical Industry Co., Ltd.	NA ^a	NA ^a
30.0	SAN 30	Dow Chemical (U.S.A.)	NAa	NAa
33.0	SAN 33	Dow Chemical (U.S.A.)	NAa	NAa

^a NA = Not available but designed to be similar to SAN 5.7 and SAN 14.7.

acteristic of the glass transition were computed from each thermogram, namely, the onset, midpoint, and completion temperatures of the glass transition region as described previously.²³ The method reported is clearly indicated on each figure.

The temperature at which phase separation occurred on heating, or the cloud point, for blends exhibiting LCST behavior was obtained by a method that has been described in full detail elsewhere.²⁴

BLENDS WITH SAN COPOLYMERS

Glass Transition Behavior

Since there are 88 possible binary combinations of the imidized acrylics (Table I) and the SAN copolymers (Table II), this presentation will be restricted to selected blend proportions and copolymer compositions that serve to illustrate the results found. The conclusions reached about miscibility

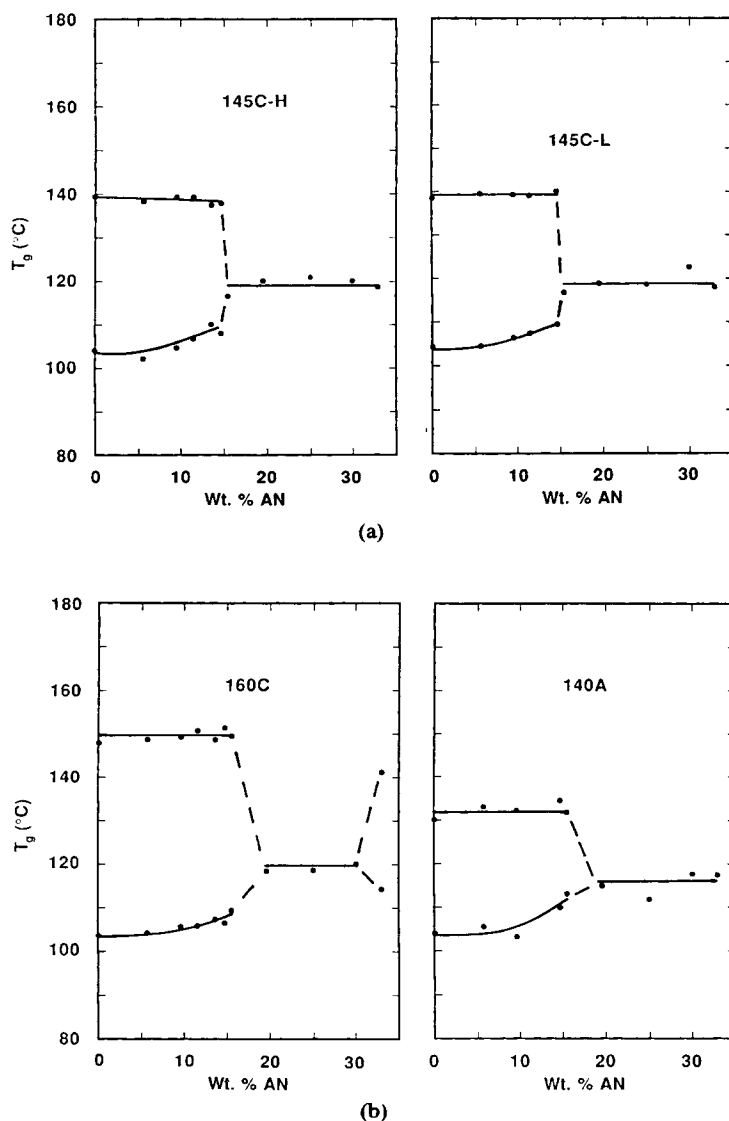


Fig. 1. Glass transition temperatures (onset) for 50/50 blends plotted vs. the AN content of the SAN copolymer for those imidized acrylics showing a miscibility window (regions of one T_g shown here).

are representative of all other blends examined during the course of this investigation.

First, 50/50 blends for each of the 88 combinations were examined by DSC for glass transition behavior. For four of the imidized acrylics, blends with some of the SAN copolymers exhibited a single glass transition while the other four gave blends with two glass transitions for all of the SAN copolymers in Table II. These results are summarized in Figures 1 and 2 by plotting the observed glass transitions (onset temperature) for the 50/50 blends vs. the wt % acrylonitrile in the SAN copolymers. As seen in Figure 1, the 145C-H

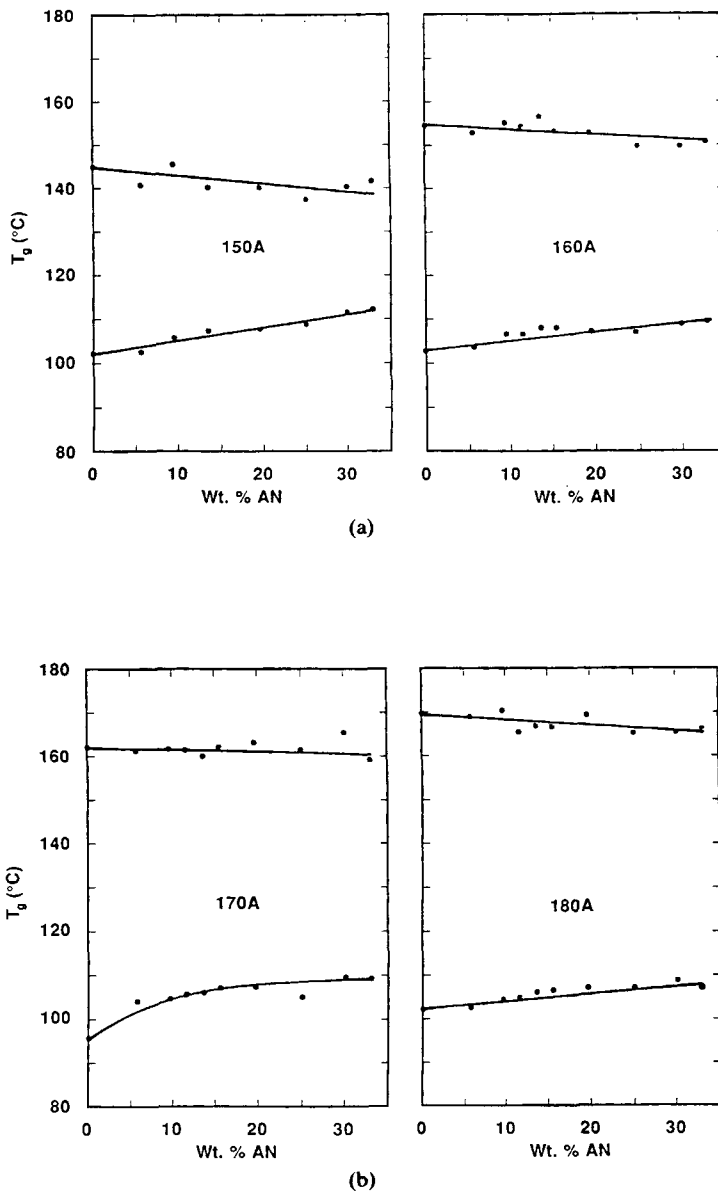


Fig. 2. Glass transition temperatures (onset) for 50/50 blends plotted vs. the AN content of the SAN copolymer for those imidized acrylics not showing a miscibility region.

and 145C-L give single T_g blends with SAN copolymers containing 15.5% AN or more up through the highest level available (33% AN) while 140A shows similar behavior from 19.5% AN. On the other hand, blends with 160C show single T_g behavior from 19.5 to 30% AN; however, its blends with SAN 33 have two T_g 's. Each of the imidized acrylics form blends with two glass transitions when the SAN copolymer contains less AN than indicated above. While not tested, it seems likely that none of the imidized acrylics would form miscible blends with a polyacrylonitrile homopolymer. Thus, we expect that

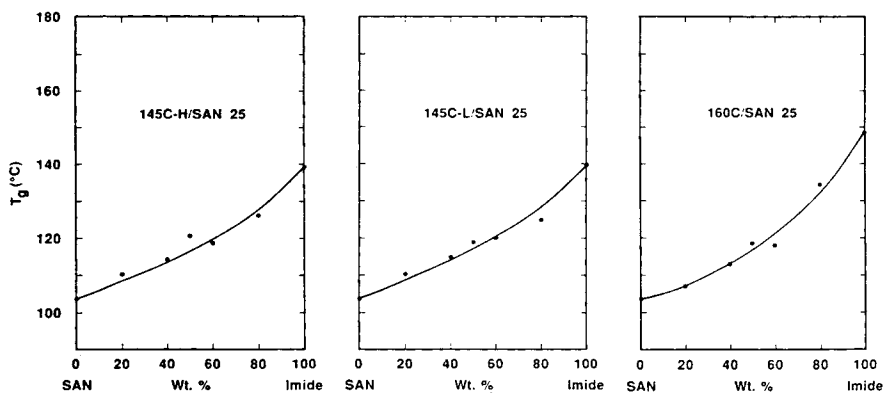


Fig. 3. Examples of T_g (onset) vs. blend composition relations for blends with an SAN containing 25% AN.

145C-H, 145C-L, and 140A would lead to blends with two T_g 's at some higher AN level, as 160C does, if such SAN copolymers had been available for this study. Consequently, we believe that these four imidized acrylics form miscible or single T_g blends with SAN copolymers only over a limited range of AN contents, while phase separated or two T_g blends are formed on either side of this AN range. Many examples of miscibility windows have been observed for blends of copolymers with other polymers.¹⁵ Recent models¹³⁻¹⁵ suggest that repulsive interactions between the two types of monomer units of the copolymer are a factor in such thermodynamic phase behavior.

Each blend in Figure 1 was prepared by the solution casting technique described earlier except for those containing 140A. Solution-cast blends containing the latter polymer exhibited two T_g 's corresponding to those of the pure components for every SAN copolymer. Melt processed blends, however, were found to be miscible with certain SAN copolymers as shown in Figure 1. This indicates that the solvent selected, CH_2Cl_2 ; for casting blends does not lead to equilibrium mixtures for this pair as has been observed for some other systems.^{6, 25-27} In all other cases, melt blending led to the same result as solution casting.

Figure 3 shows the glass transition behavior for blends of three of these imidized acrylics with SAN 25 (this AN level corresponds to the midpoint of miscibility range) over the entire range of component proportions. These results are typical of completely miscible blend systems. Similar data are not shown here for 140A since the supply of this material was too limited to prepare every composition by melt processing; however, selected blends at compositions other than 50/50 did show single T_g behavior.

The four imidized acrylic materials for which results are given in Figure 2 showed two glass transitions when solution blended with every SAN copolymer listed in Table II. Similar results were obtained when the blends were prepared by melt mixing. Thus, these four materials do not appear to form miscible blends with SAN copolymers regardless of AN content, at least up to the 33% level. Apparently, the acid functionality of these compositions is responsible for their failure to homogeneously mix with SAN copolymers, possibly because of the large energy needed to break up hydrogen bonding

TABLE III
Effect of Free Acid Composition on the Miscibility of Imidized Acrylic Polymer with SAN^a

Free acid (wt %)	Glass transition behavior ^b	Visual appearance
Nil	1 T_g (120)	Clear
1.0	2 T_g 's (115.8, 145.2)	Almost clear
2.5	2 T_g 's (110.2, 155.5)	Translucent
3.9	2 T_g 's (110.1, 160.8)	Opaque
4.6	2 T_g 's (109.2, 157.7)	Opaque

^aThe imidized acrylic polymers employed had a constant imide content of 83% by weight and were mixed in 50/50 proportions with an SAN containing 25% AN by weight (Tyril 1000) using a Brabender mixing device.

^bGlass transitions were measured by DSC, and the values in parenthesis represent the onset of the T_g region in °C.

interactions between the carboxyl groups. The effects of acid functionality on miscibility is shown more dramatically in Table III. In this example, a series of imidized acrylics having a constant imide content of 83% by weight, but a variable level of acid functionality were melt mixed with an SAN containing 25% by weight of AN (Tyril 1000 from Dow Chemical Co.). As expected from the results above, the material with no acid functionality forms a clear blend with a single T_g , indicating miscibility. At only 1% acid, the blend is nearly transparent but exhibits two T_g 's. However, since these T_g 's are significantly shifted from those of the pure components, it is apparent that some partial miscibility exists. With higher acid contents, the blends become progressively more opaque, and the two T_g 's correspond more closely to those of the pure components.

On the other hand, the material designated 140A contains 2% acid but is miscible with SANs containing 19.5% or more of AN as seen in Figure 1. However, this material contains only 53% by weight of the imide structure while all others in Table I contain 72% or more. The high MMA content of 140A is evidently the factor that offsets the detrimental effect of the relatively low acid content of this polymer. Recall that pure PMMA is miscible with SANs containing 9.5–33% AN, as reported earlier.⁶ The wide range of structural variables of this class of polymers will permit design of compositions meeting various requirements simultaneously.

LCST Behavior

Most of the miscible systems described above were examined for phase separation on heating since miscible blends frequently show such behavior.²⁴ Figure 4 shows complete cloud point curves for blends of 145C-H, 145C-L, and 160C with several SAN copolymers. In some cases, the cloud points were so high that phase separation could not be observed before thermal degradation or depolymerization occurred, whereas, in marginal cases, estimates of the cloud point could be made by minimizing the time the samples were at excessively high temperatures. The effect of AN content on these results may be seen more clearly in Figure 5, where the cloud points for 50/50 blends are plotted vs. the AN content of the SAN. In each case, there is a maximum cloud point at about 25% AN. This is consistent with experimental results

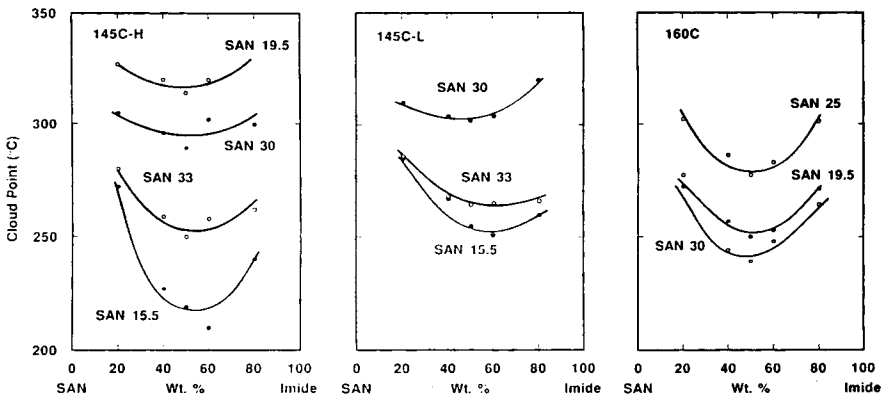


Fig. 4. Cloud point curves for selected SAN/imidized acrylic blends.

observed for many other blend systems involving copolymers⁶ and with the prediction of recent theories¹³⁻¹⁶ (with the reasonable hypothesis that the cloud point will be higher the more favorable the interaction between the two components).

For a given AN level, the various imides give cloud points in the following rank order: 145C-L > 145C-H > 160C. Note that for 19.5 and 25.0% AN, the cloud points for blends with 145C-L were too high to measure. Since 145C-L and 145C-H are essentially identical in composition, the higher cloud points for the former evidently reflect its lower molecular weight. The lower cloud

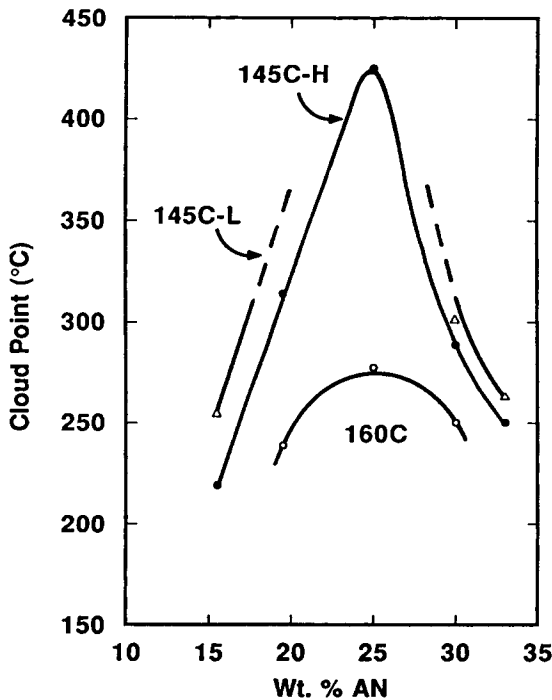


Fig. 5. Cloud points for 50/50 blends plotted vs. the AN content of the SAN copolymer for three imidized acrylics.

points for 160C through 145C-H represents a compositional effect (lower MMA or higher imide level) since these two have rather similar molecular weights. At this point it is important to remember that these imidized acrylics should also be viewed as copolymers [see eq. (1)]. Based on previous work,⁶ it is known that pure PMMA is miscible with a wide range of SAN copolymers, but it is quite possible that the hypothetical structure of 100% imide units would not be similarly miscible. The intramolecular interactions between the MMA and imide units will enhance the tendency for miscibility with other polymers if it is repulsive,¹⁵ as we might expect. Phase behavior for blends of imidized acrylics with SANs is determined by the appropriate summation of all intermolecular and intramolecular interactions and can be represented on a 2-dimensional composition map, as reported for other copolymer-copolymer systems,^{10, 11, 28-30} where the two coordinates are % imide and % AN. From this work, it has not been possible to locate the optimum composition coordinates for the most favorable interactions between these two copolymer types owing to the limited range of imidized acrylics available for study. However, for a fixed AN level (e.g., 25%), we can speculate that an indicator like the LCST would vary in the following way as the % imide is changed. It would start at a finite level for 0% imide (or pure PMMA), then rise to a maximum, and go down to a level that precludes miscibility prior to reaching 100% imide.

BLENDS WITH PVC

Glass Transition Behavior

The glass transition behavior for blends of each of the imidized acrylics in Table I (except 140A) with PVC is summarized in Figures 6 and 7. The open circles indicate the midpoint of the glass transition region while the solid points below and above this correspond, respectively, to the onset and completion temperatures. This information is presented to give an indication of the breadth of the T_g region, which will be an important factor in following discussions. For blends that exhibited two glass transitions, these measures are given for both transitions.

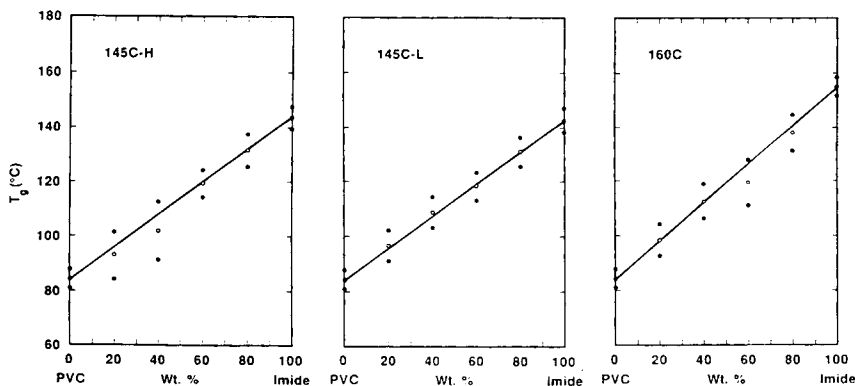


Fig. 6. Glass transition temperature behavior for blends of PVC with imidized acrylics having no acid functionality: (○) midpoint of T_g region; (●) onset (above) and completion (below) temperatures.

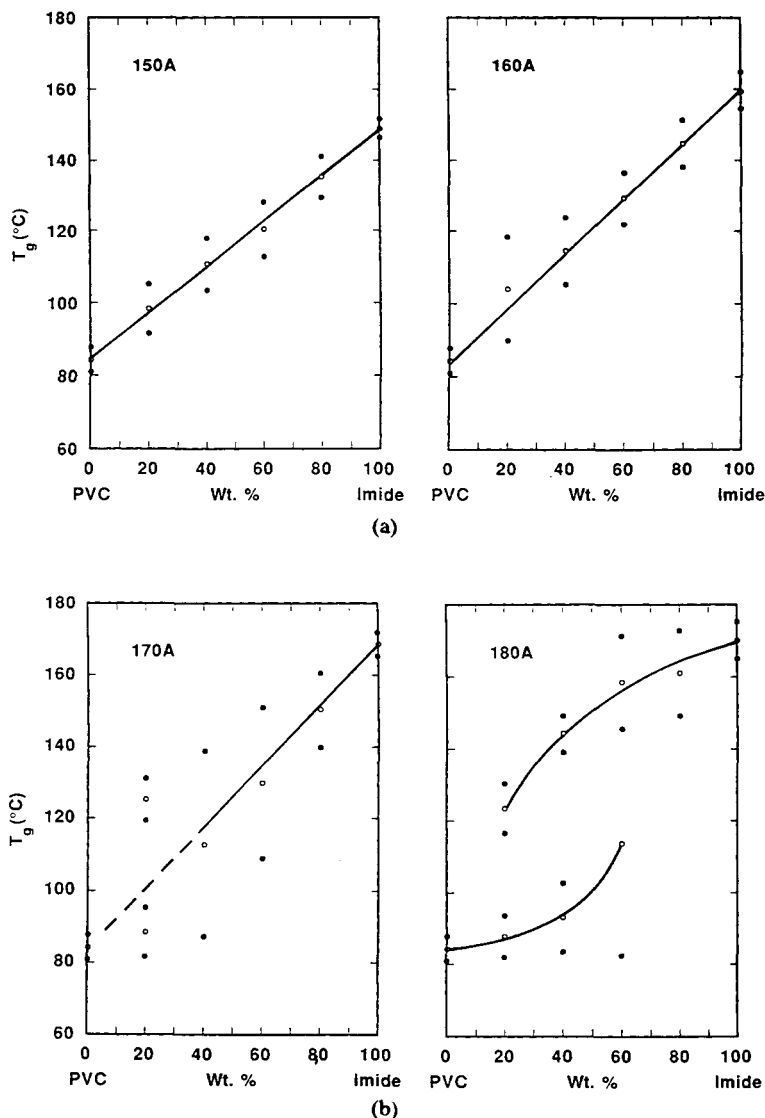


Fig. 7. Glass transition temperature behavior for blends of PVC with imidized acrylics having acid functionality.

As seen in Figure 6, blends of PVC with the three imidized acrylics having no acid functionality (145C-H, 145C-L, and 160C) exhibit a single, composition-dependent glass transition temperature. The transitions for the blends are somewhat broader than those of the pure components, as expected, but not unusually so.²⁵

Figure 7 shows the T_g behavior for blends of PVC with the imidized acrylics having acid functionality. As the numerical code (Vicat temperature) increases in this series of polymers, both the acid content *and* the imide level increase as seen in Table I. The first two, 150A and 160A, form miscible blends with PVC over the entire composition range. The glass transitions are of similar breadth as in Figure 6 except for the blend containing 20% 160A, whose

transition is unusually broad. On going to 170A, the 20% blend has two glass transitions; however, within our ability to resolve them by DSC, all other proportions of PVC and 170A appear to have single but very broad T_g 's. Blends of PVC with 180A show even more distinct evidence for phase separation. These blends exhibit a glass transition near that of PVC, which broadens with imide content and is not detectable in the 80% blend. There is also a higher T_g indicative of a phase rich in imidized acrylic. This transition drops considerably from that of the pure component as PVC is added. Thus, in going from 150A to 180A, there is a steady progression toward immiscibility, as seen by the results in Figure 7.

Because of the coupling of the acid and imide contents in this series, it is not possible to conclude from Figure 7 which is the most responsible factor for the trends seen. However, comparison of results for 170A and 160C provides a better picture. These polymers have the same imide content and essentially the same molecular weights, but they differ in acid level and MMA content. PVC is clearly miscible with 160C but much less so with 170A. We suggest that the primary issue here is the acid functionality of the latter, although the difference in MMA levels may also be a factor.

From the point of view of achieving miscibility, it is quite clear from Figures 2 and 7 that PVC is more tolerant of acid functionality in the imidized acrylic than SAN copolymers are. The causes for this are not entirely clear; however, since PMMA evidently has more limited miscibility with PVC¹⁷⁻²² than with SAN, attractive interactions with this structural part of the imidized acrylic is not the cause.

LCST Behavior

It was not possible to observe phase separation on heating for any PVC blends since rapid dehydrochlorination occurred, in spite of use of a stabilizer, before any cloud point. From a purely scientific point of view, this is unfortunate since it deprives us of a useful qualitative approach for assessing interactions between PVC and the various imidized acrylic polymers.

SUMMARY

The glass transition results presented here demonstrate that a range of imidized acrylic polymers form miscible blends with styrene/acrylonitrile copolymers over specific ranges of AN contents, whereas none of the former are miscible with polystyrene and are presumed not to be miscible with polyacrylonitrile. Cloud points for these blends reach a maximum temperature when the AN level of the SAN copolymer is about 25% by weight. Thus, intramolecular repulsion between styrene and acrylonitrile units within the copolymers appears to be a significant factor promoting miscibility. Acid groups in the imidized acrylic appear to hinder immiscibility with SAN presumably due to the unfavorable energy of breaking up hydrogen bonding between these units. By appropriately balancing all of the structural variables, however, it is possible to design imidized acrylics having acid functionality to be miscible with SAN copolymers.

Poly(vinyl chloride) also forms miscible blends with many of the imidized acrylics examined here, and miscibility seems to be less hindered in this case by the presence of acid groups. The latter probably reflects greater opportuni-

ties for more favorable net interaction of these groups with PVC (e.g., hydrogen bonding) than with SAN relative to the hydrogen bonds that may exist among these acid groups within the imidized acrylic. The mechanism for miscibility with PVC probably relates to the high carbonyl density of the imidized acrylics and repulsive interactions among the various units in the imidized acrylics. None of the blends with PVC showed phase separation on heating prior to temperatures where PVC becomes chemically unstable.

Properly selected imidized acrylics can significantly raise the glass transition of SAN and PVC compositions and may offer a variety of other advantages including sites of chemical reactivity.

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