

Polybutylene Terephthalate/High-Density Polyethylene Alloys. I. Morphological Studies

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SYNOPSIS

Polybutylene terephthalate (PBT) is a commercially successful thermoplastic polyester but has certain drawbacks such as low impact strength and low melt strength. An attempt has been made to modify the properties of PBT by blending it with polyolefin such as high-density polyethylene (HDPE). Since PBT and HDPE are incompatible, an ionomer has been used as a compatibilizer to form an alloy. Alloys of PBT and HDPE with varying amounts (2–8%) of ionomer were prepared by melt blending. The ultimate mechanical properties improved significantly on the addition of the ionomer due to an increase in interfacial adhesion between PBT and HDPE. DSC studies show that the presence of ionomer facilitated the crystallization of PBT in the alloy. DMTA studies show that more of PBT (amorphous) is going in to the HDPE-rich phase in the presence of ionomer. The morphology of the alloys was studied using scanning electron microscopy (SEM), polarizing microscopy (PM), and small-angle light scattering (SALS). They showed improved dispersion of HDPE domains in PBT matrix with increasing ionomer content and change in the type of superstructure on adding the ionomer. It has been shown that an alloy of PBT and HDPE with improved mechanical properties and homogeneous morphology can be made with use of ionomer as a compatibilizer. Such alloys are cost effective and can find use in several engineering applications.

INTRODUCTION

Blending is an economically viable and versatile way in which new materials can be produced to have a wide range of properties.^{1–3} There has been considerable interest in the phenomenon of miscibility in the binary polymer blends. The majority of the polymers are immiscible at the molecular level as determined by laws of thermodynamics and consist of two distinct phases. It is possible to render such an immiscible blend compatible by adding a third polymer that would promote interactions across the matrix-dispersed phase interface. Most published papers on this topic have considered immiscible polymer blends with small amounts of block or graft copolymers added to act as compatibilizing agents.^{4,5} In the latest of a series of papers, Teyssie and co-

workers^{6,7} observed a significant reduction in the dispersed-phase size and an increase in the interfacial adhesion as a result of melt-blending polyethylene and polystyrene with as little as 2% by wt of poly(butadiene-*b*-styrene) copolymer.

The object of this study is to determine the compatibilizing effect of an ionomer poly(ethylene-*co*-sodium methacrylate) on the morphology of a polyolefin/polyester blend. The polyolefin and the polyester chosen were high-density polyethylene (HDPE) and polybutylene terephthalate (PBT), respectively. Some of the similar compatibilized systems that have been studied recently are those of PET/HDPE blends where a block copolymer of styrene and butadiene act as an effective compatibilizer. The rubber block exhibits a certain miscibility with the polyolefin hydrocarbon backbone, whereas the end blocks of styrene having a common aromatic character of PET^{8,9} will be miscible with the latter. Compatibilized blends of PP or PE with PET have also been reported where PP/PE is the

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major component and PET is the minor component. Acrylic acid-grafted polypropylene (PP-*g*-AA) was seen to act as an effective compatibilizer for this system.^{10,11} Although the compatibilization of blends by the presence of graft copolymers has been well studied, little work has been done on compatibilized ternary blends such as polyester/ionomer/polyolefin.

The choice of ionomer as compatibilizer is based on the miscibility or reactivity of its segments with at least one of the blend components. Nonreactive copolymers have segments that are capable of specific interactions with each of the blend components, their miscibility often dictated by their closely matched solubility parameters. The mode of compatibilization in PBT/HDPE/ionomer blend is based on specific interactions that the ionomer has with the blend components. The polyethylene backbone is a common feature of both HDPE and the ionomer and, hence, will be miscible. On the other hand, carboxyl group of PBT will have affinity to link with (COO)⁻ group present in the side chains of the ionomer. Thus, the ionomer will act as a bridge holding the polyester and polyolefin components of the blend to give a compatibilized blend.

In this paper, the influence of interfacial modification on the morphology of a PBT/ionomer/HDPE ternary blend is studied by scanning electron microscopy (SEM), polarizing microscopy (PM), and small-angle light scattering (SALS) techniques. The crystallization behavior of these blends was studied with the help of differential scanning calorimetry (DSC) and wide-angle X-ray diffraction (WAXD). Dynamic mechanical thermal analysis (DMTA) was also done in order to understand finer details regarding the degree of compatibility in the blends with the ionomer.

EXPERIMENTAL

Material

Polybutylene terephthalate (PBT) (tradename ARNITE T-006) was obtained from Cenka Plastics Ltd., India. High-density polyethylene (HDPE) (tradename Hostalan) with an MFI value of 10 was obtained from Polyolefins Industries Ltd. The compatibilizing agent was an ionomer with the tradename SURLYN 8660 obtained from DuPont (U.S.A.). This ionomeric resin is a random terpolymer consisting of polyethylene backbone as the major component and neutralized as well as unneutralized pendant carboxylate groups. First, a co-

polymer of ethylene with 3–10 wt % of methacrylic acid is made and then 20–80% of acid groups are neutralized by either sodium or zinc.

Sample Preparation

In the ternary blend of PBT/HDPE/Ionomer, the polyester (PBT) and polyolefin (HDPE) were used in the ratio of 80/20 (wt/wt) for all compositions, while the ionomer content was chosen as 2, 4, and 8% by wt (Table I). All the blends were prepared by the melt-mixing technique using a single-screw extruder of L/D 20, model Betol DM 1820. Vacuum-dried polymer granules of PBT, HDPE, and Surlyn were first dry mixed to the various compositions as shown in Table I. These were then melt blended in the extruder at 240°C and 30 rpm. Extruded strands of the blends were quenched in cold water and granulated and the granules dried before further processing.

For morphological studies, dried granules of blend as well as pure polymers were compression molded to make 10 mL film on a Carver laboratory hydraulic press at 10,000 psi pressure and 240°C temperature. For SALS as well as optical microscopy studies, the compression-molded films were sandwiched between two microscopic coverslips and melted in a silicone oil bath (240°C) and then quenched to 0°C to give thin films. For SEM studies, injection-molded bars fractured in liquid nitrogen were used. Sheath and core areas of the ternary blend specimens were separately observed. The SEM micrographs were recorded on Stereoscan, Model S4-10, of Cambridge Scientific instruments Ltd. at a magnification of about 1000.

For SALS studies, thin-molded films were examined by using a photographic SALS apparatus¹² with a 5 MW He-Ne source. The *H*_v light-scattering

Table I Blend Compositions

Sample No.	% Weight		
	PBT	HDPE	Ionomer
1	100	—	—
2	—	100	—
3	—	96.0	4
4	80	20	—
5 ^a	78.4	19.6	2
6 ^a	76.8	19.2	4
7 ^a	73.6	18.4	8

^a PBT/HDPE: 80/20 ratio is fixed in all cases.

patterns were recorded on NP22 ORWO plain film and scanned by a Joyce and Loebel microdensitometer. Phase morphology of the blends was also studied on Leitz polarizing microscope.

WAXD studies were carried out on a RIGAKU (D/max-B series) X-ray diffractometer.

For DSC studies, a DuPont 1090 differential scanning calorimeter was used with a heating rate of 10°C/min in nitrogen atmosphere up to 250°C to obtain the heating curve. The samples were held at this temperature for 10 min to eliminate any previous history and then subsequently cooled at 10°C/min up to 50°C to obtain the cooling curve.

DMTA studies were done on a dynamic mechanical thermal analyzer made by Polymer Laboratories. The experiment was carried out in the temperature range of -120 to 100°C at the heating rate of 5°C/min. Test specimens in the shape of rectangular bars were cut out from the injection-molded specimens according to the specification prescribed for the instrument. Samples were scanned with an imposed frequency of 5 Hz in the bending mode and storage modulus; loss modulus and $\tan \delta$ were recorded as a function of temperature. The T_g is defined as the temperature corresponding to the maximum in E'' or $\tan \delta$ ($\tan \delta = E''/E'$) at the main relaxation that marks the onset of main-chain segmental mobility corresponding to glass transition. All the pure components along with the blend samples were also tested by DMTA under similar conditions for a valid comparison.

RESULTS AND DISCUSSION

DSC

Figures 1 and 2 show some representative DSC thermograms of the melt-blended granules during heating and cooling, respectively. Comparisons of various parameters determined from these curves are given in Table II. From the table, it can be seen that the melting temperature of the PBT component is about the same as that of pure PBT, in both the compatibilized and the incompatibilized blends. The melting temperature of the HDPE component shows a slight decrease in the blend systems. This suggests that crystallite size is slightly decreased for the HDPE component in the blends, whereas that of PBT component remains unchanged. This is because some of the amorphous PBT is probably going into the HDPE-rich phase, whereas not much of the amorphous HDPE is going into the PBT-rich phase.

Hence, the melting temperature of PBT is not affected but that of HDPE shows a decrease. On the addition of Surlyn to the blend, the T_c for the PBT component shifts to a higher value, which suggests that addition of ionomer to the PBT/HDPE blend has facilitated crystallization. In other words, the degree of crystallinity increases on addition of ionomer, which is also reflected in the higher ΔH_c values of the PBT component in the compatibilized blends. T_c of the HDPE component does not change on addition of Surlyn, but we do observe a decrease in ΔH_c component.

The width of the crystallization exotherm ΔT_c is found to increase on addition of ionomer to the blend. ΔT_c is reported to be a measure of crystallization rate.¹³ Hence, we can say that addition of ionomer has facilitated crystallization and, thus, increased the rate of crystallization in the blend. The high rate of crystallization at higher Surlyn content may be the cause of formation of imperfect crystals; hence, ΔH_f is reduced in the compatibilized blend. Finally, it can be seen that the blend with 8% Surlyn shows a separate melting peak for the ionomer component and the T_m of this component is depressed in the blend system as compared to pure ionomer. Such a melting-point depression can be attributed to a favorable free energy of mixing between blend components.¹⁴ On this basis, we can say that the ionomer (Surlyn) is compatible with the PBT/HDPE blend system.

WAXD

Figure 3 gives intensity vs. 2θ scan for pure PBT and HDPE as well as for the blends. PBT crystallizes in two crystalline modifications: α and β forms. The latter is formed due to mechanical deformation. The pure PBT film has four strong peaks at $2\theta = 17.4^\circ$, 20.7° , 23.5° , and 25.3° . The strongest peak is around 23.5° .¹⁵ Pure HDPE as shown in Figure 3 has a strong peak at 21.7° and a less intense but distinct peak at 24° . Occurrence of all characteristic peaks of PBT in the diffraction patterns of the blends without any additional peak indicates that the PBT crystallization pattern is not affected in the blends. However, it is seen that the peak at 21.6° becomes more intense as compared to that at 23.5° in the case of pure PBT. This may be because HDPE also crystallizes in the blend and overlapping of peaks results in higher intensity of the peak at 21.6° in the blend. In the compatibilized blends at ionomer contents (2% and 4%), the peak at 22.0° and 20.8° , respectively, are more intense than are the others.

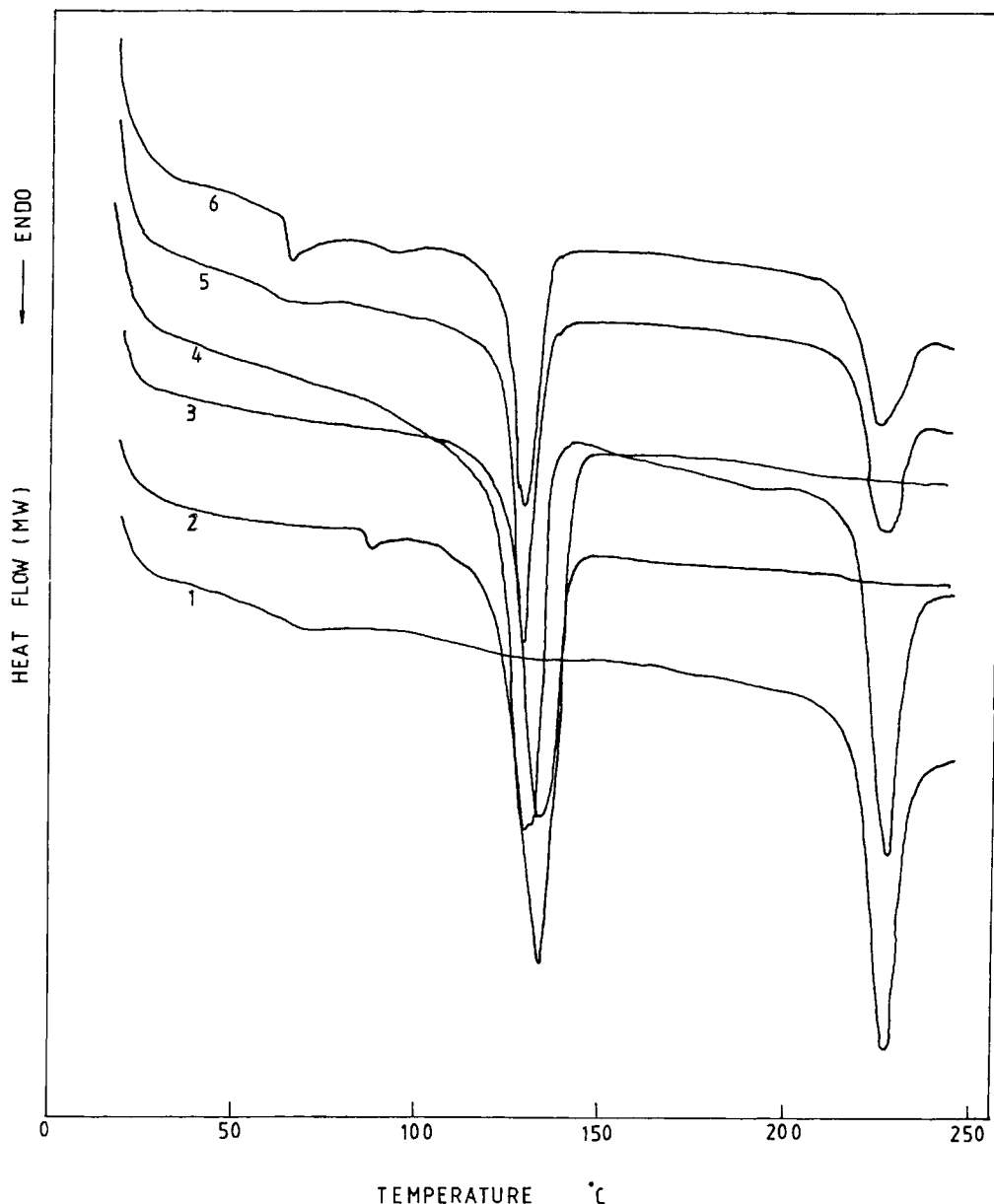


Figure 1 DSC thermograms for PBT/HDPE/ionomer blend samples (heating cycle). (1) PBT; (2) HDPE; (3) HDPE/ionomer (4%); (4) PBT/HDPE; (5) PBT/HDPE/ionomer (4%); (6) PBT/HDPE/ionomer (8%).

At 8% ionomer content, the peak at 23.4° is more intense than are the others, the same as that of pure PBT. The peaks at 23° and 17.4° are characteristics of α forms of PBT. Hence, we can say that on blending the β form of PBT becomes more dominant for low ionomer content, whereas at higher ionomer content (8%), again it is the α form that predominates.

Degree of crystallinity (X_c) was calculated from these diffractograms using the method described in the literature.^{16,17} The experimental I vs. 2θ curves

were converted into IS^2 vs. S curves. An amorphous scattering curve was drawn in accordance with method suggested by Sotton.¹⁶

The degree of crystallinity (X_c) approximately calculated for these samples as above, varies with blend composition as shown in Table III. It is seen that blending has improved the overall crystallinity of the sample in the compatibilized blends; the composition with 4% ionomer content shows the highest degree of crystallinity. This is also reflected in its high ΔH_c values as seen in the DSC results.

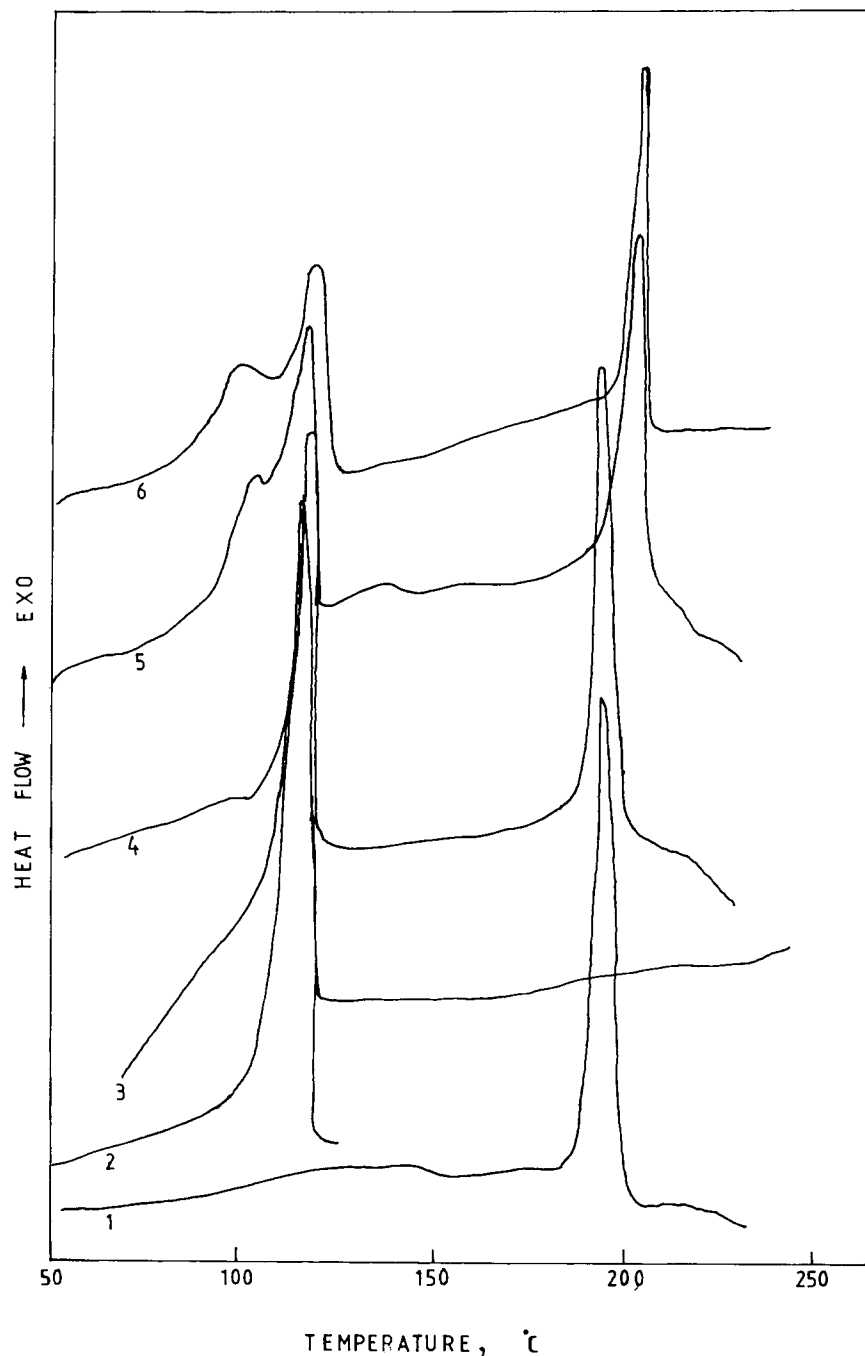


Figure 2 DSC thermograms for PBT/HDPE/ionomer blend samples (cooling cycle). (1) PBT; (2) HDPE; (3) HDPE/ionomer (4%); (4) PBT/HDPE; (5) PBT/HDPE/ionomer (4%); (6) PBT/HDPE/ionomer (8%).

DMTA

Pure polymers and their blends were subjected to a sinusoidal force of known magnitude in the bending mode, and the resultant displacement in the sample was measured. Storage modulus (E'), loss modulus (E''), and $\tan \delta$ (δ being the phase angle) were de-

termined as a function of temperature. T_g values of the blends were determined from the DMTA traces and are shown in Table IV. These were evaluated with respect to the blend composition.

Pure PBT shows α -relaxation peaks at 63 and 64°C, respectively, on $\tan \delta$ and E'' plots as is shown in Figure 4(i). Pure HDPE shows γ -relaxation

Table II Tabulation of Data from DSC Thermograms of PBT/HDPE/Ionomer Blend Samples

Sample	T_m (°C)	ΔH_f^a (J/gm)	ΔH_c^a (J/gm)	T_c (°C)	ΔT
PBT	227.2	25.9	37.1	194.3	5.0
HDPE	134.6	83.8	137.3	115.0	6.0
HDPE + ionomer (4%)	134.4	96.0	172.0	115.2	6.0
PBT/HDPE	227.6	24.8	38.7	193.7	4.5
	129.0	139.0	163.0	117.5	5.0
PBT/HDPE/ionomer (4%)	226.8	14.5	31.2	205.0	8.0
	129.0	98.0	150.5	116.8	—
PBT/HDPE/ionomer (8%)	226.8	14.3	31.2	205.0	8.0
	130.5	96.7	131.0	114.4	—
	67.3	18.6	—	—	—
Surlyn	98.3	30.3	—	—	—

^a Data normalized to per gram of PBT and per gram of HDPE in the blend.

peaks at -99 and -100°C , respectively, on $\tan \delta$ and E'' plots as is shown in Figure 4(ii). Figure 4(iii-v) shows DMTA traces for blend samples.

All the blends show two glass transition temperatures (T_g) that confirm the presence of two-phase systems. This shows that the PBT/HDPE system

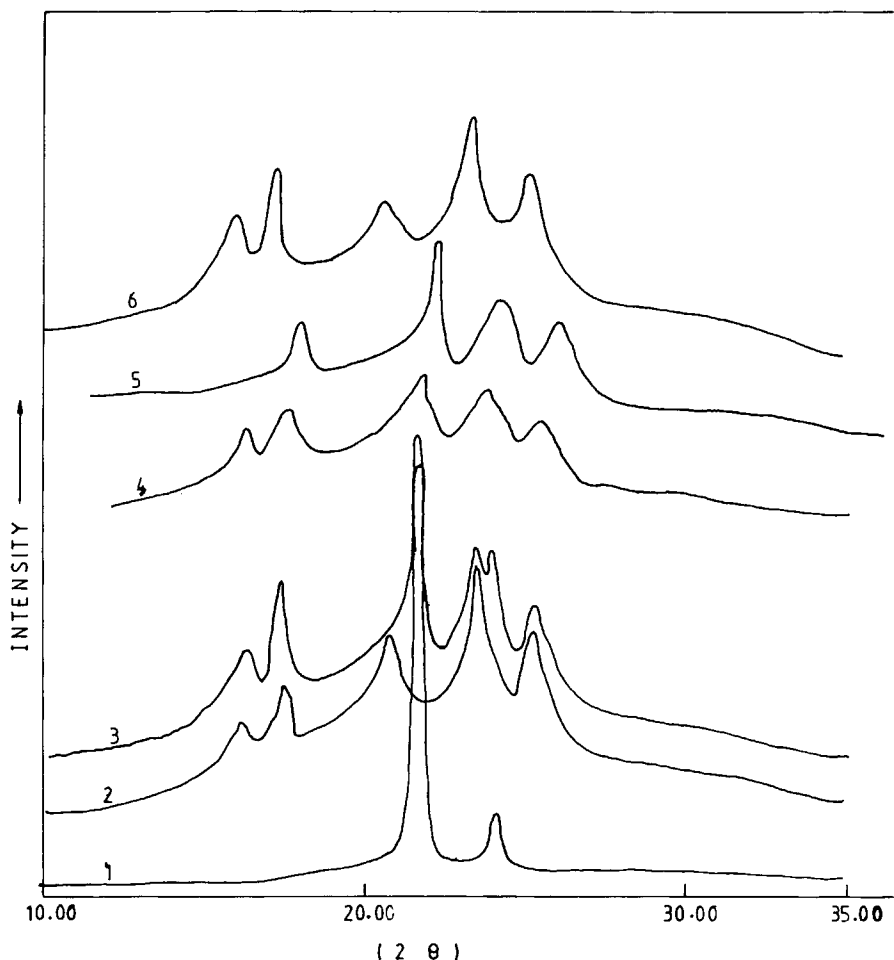


Figure 3 WAXD I vs. 2θ plots for PBT/HDPE/ionomer blend samples. (1) HDPE; (2) PBT; (3) PBT/HDPE; (4) PBT/HDPE/IONO (2%); (5) PBT/HDPE/IONO (4%); (6) PBT/HDPE/IONO (8%).

Table III Peak Positions and Their Relative Intensity Data Obtained from WAXD Study of PBT/HDPE/Ionomer Blends

Sample	2θ (Intensity)				X_c % Crystallinity
	PBT	17.4 (1036) ^a	20.7 (1355)	23.5 (1822)	25.3 (1359)
HDPE	—	21.7 (9060)	24.0 (1695)	—	69
PBT/HDPE	17.3 (1439)	21.6 (2322)	23.4 (1600)	25.3 (1238)	54
PBT/HDPE/ionomer (2%)	17.6 (957)	22.0 (1160)	24.1 (1089)	25.5 (877)	48
PBT/HDPE/ionomer (4%)	16.6 (529)	20.8 (862)	23.2 (606)	24.7 (534)	56
PBT/HDPE/ionomer (8%)	17.3 (961)	20.4 (819)	23.4 (1185)	25.0 (953)	49

^a All values given in parentheses are the relative intensities.

is not a totally miscible one as is also predicted by their solubility parameter values [PBT = 10.7 (cal/cc)^{1/2}, HDPE = 9.02 (cal/cc)^{1/2}]. The two T_g values are around 66°C for the PBT-rich region and around -63°C for the HDPE-rich region. T_g for the PBT-rich region is not far from the T_g of pure PBT for both compatibilized as well as uncompatibilized systems. T_g for the HDPE-rich regions has significantly shifted to higher temperatures for both the blend systems, which means that PBT is dissolving in the HDPE-rich phase to a greater extent than is the HDPE dissolving in the PBT-rich phase. A similar result has been interpreted for PS-polybutadiene blends by Kim and Burns.¹⁸ This can also be

confirmed by looking at the peak height value of the PBT-rich phase, which is higher for PBT/HDPE (in terms of $\tan \delta$ value) and decreases on addition of ionomer. This, again, indicates that more PBT is going into the HDPE-rich phase on addition of ionomer. Pure HDPE shows three relaxation temperatures as reported in the literature¹⁹: the α peak in the 30–80°C range, the β peak in the 0–60°C range, and the γ peak in the -100 to -140°C range.

The absence of the γ -relaxation peak in the blends and, instead, the presence of a transition peak that falls in the β -relaxation range can be explained on the basis of inhibition of motion of small-chain segments of HDPE in the amorphous phase due to

Table IV Results of DMTA Regarding T_g Transitions in PBT/HDPE/Ionomer Blends

Sample	Peak 1		Peak 2		Peak 3	
	Tan δ	E''	Tan δ	E''	Tan δ	E''
PBT	63	64	—	—	—	—
HDPE	—	—	—	—	-99	-100
PBT/HDPE	66 (.12)*	62	-63 (.027)	-69	—	—
PBT/HDPE/ionomer (4%)	70 (.096)	64	-67 (.027)	-69	—	—
PBT/HDPE/ionomer (8%)	71 (.011)	64	-70 (.024)	-72	—	—

^a Peak height in terms of $\tan \delta$ value.

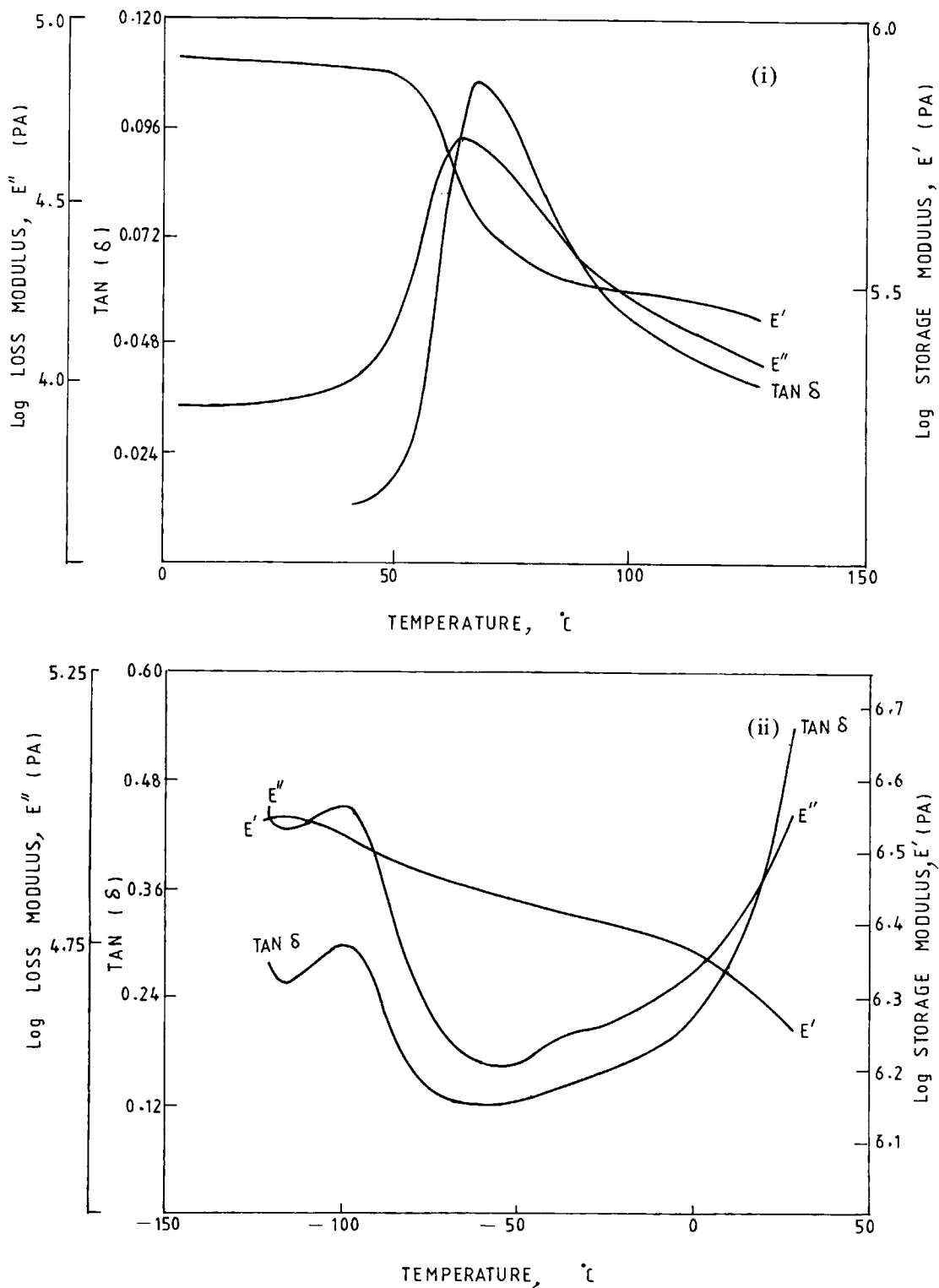


Figure 4 Dynamic Mechanical Thermal Analysis (DMTA). (i) Variation of the storage modulus (E'), loss modulus (E'') and the loss Tangent ($\tan \delta$) with temperature for pure PBT. (ii) Variation of E' , E'' and $\tan \delta$ with temperature for pure HDPE. (iii) Variation of E'' with temperature for (A) PBT/HDPE; (B) PBT/HDPE/ionomer (4%). (C) PBT/HDPE/ionomer (8%). (iv) Variation of E' with temperature for (A) PBT/HDPE; (B) PBT/HDPE/ionomer (4%); (C) PBT/HDPE/ionomer (8%). (v) Variation of $\tan \delta$ with temperature for (A) PBT/HDPE (B) PBT/HDPE/ionomer (4%); (C) PBT/HDPE/ionomer (8%).

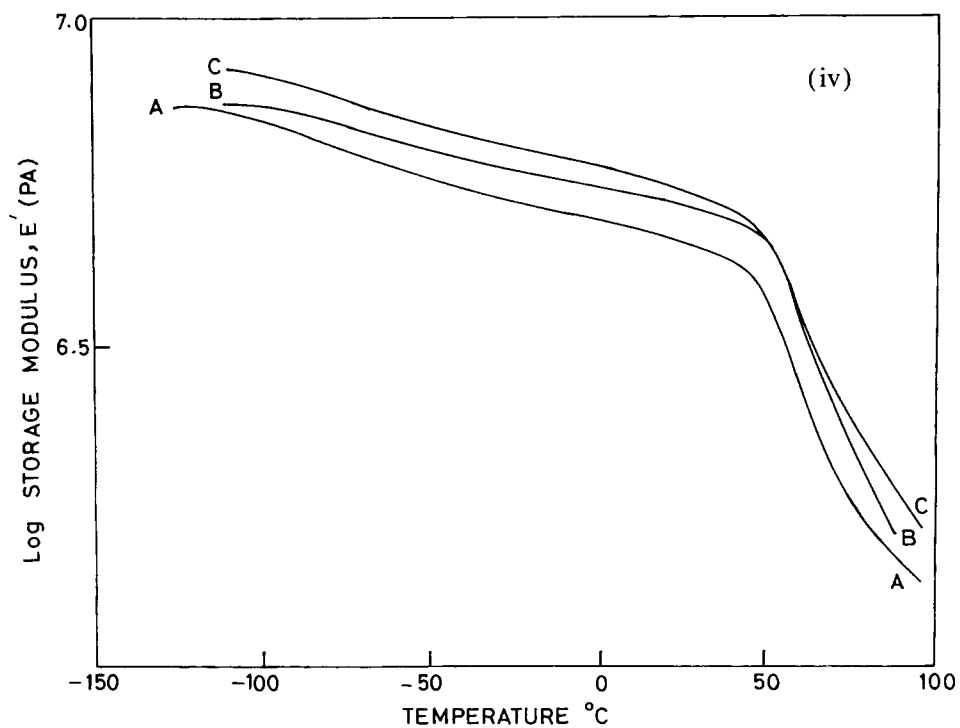
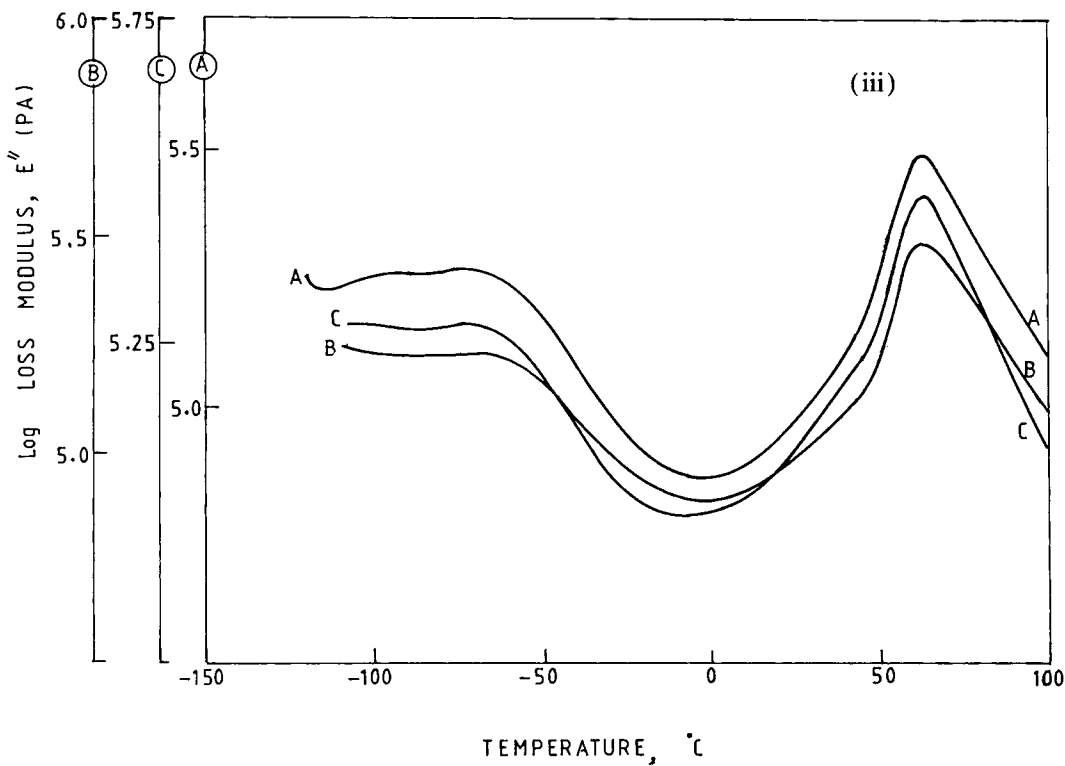


Figure 4 (Continued from the previous page)

the presence of PBT in the mixed phase rich in HDPE. However, because of the presence of PBT, β -relaxation has been recorded. β -Relaxation is virtually absent in linear and highly crystalline HDPE;

hence, it is not recorded for pure HDPE samples. But in the blend, it has been detected and the temperature decreases on addition of ionomer, which indicates that the HDPE amorphous region is more

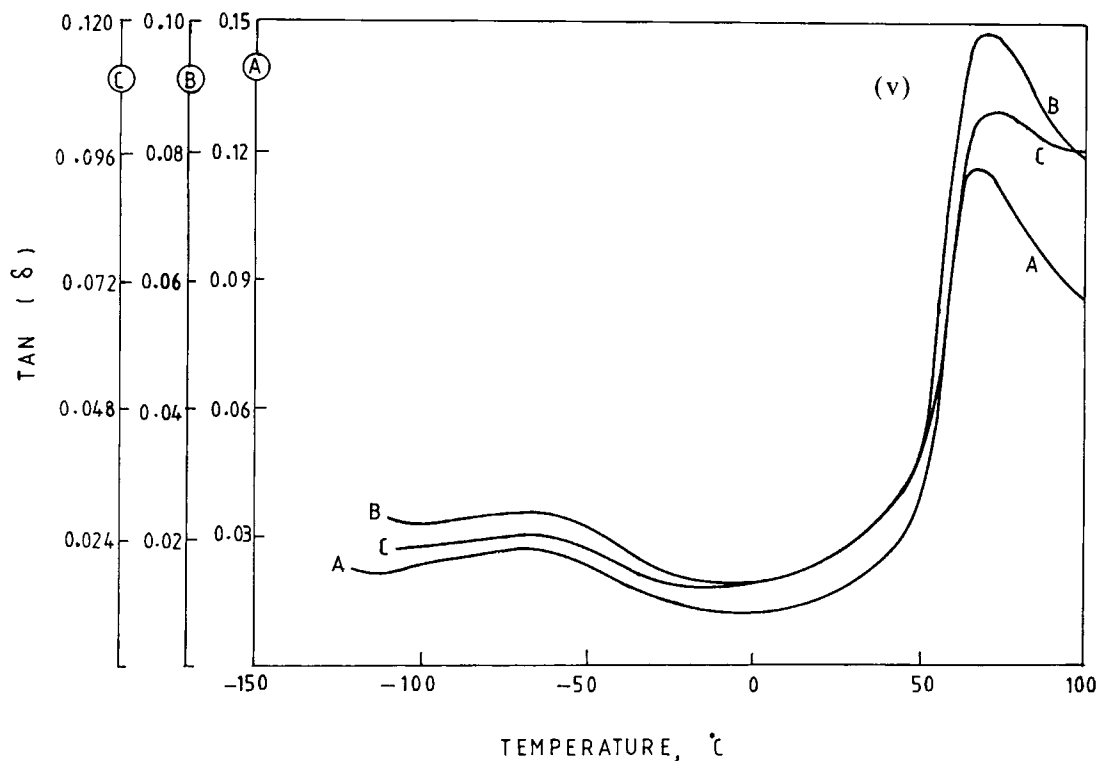


Figure 4 (Continued from the previous page)

branched, due to presence of ionomer (Surlyn) in the compatibilized blends. Hence, β -relaxation, which is associated with large-chain segment movement, becomes more conspicuous in the blend samples, especially in those with ionomer.

Morphological Studies

Scanning Electron Microscopy

The SEM photomicrographs of liquid nitrogen-fractured samples are shown in Figure 5. The ternary blends of PBT, HDPE, and Surlyn were made at the processing temperature of the high-melting polyester component, which is much higher than is the melting temperature of HDPE. Therefore, HDPE is believed to form the lower viscosity phase in the melt of the blend PBT/HDPE. As a result, redistribution of phases due to shearing flow should occur during processing when the lower viscous HDPE tends to migrate to the wall of the channel and encapsulates the higher viscosity polyester component. A similar explanation has been given for the PP forming a sheath around the PBT/PC core in ternary blends of PBT/PC/PP.²⁰ Two-phase morphology is therefore expected to be found in the

molded specimens of these kinds of blends, and it is quite probable that the core and the sheath portions of the specimens would have different compositions. The sections of sheath and core of the molded specimens of the blends were studied by SEM.

Figure 5 shows the micrographs of pure polymers PBT, HDPE, and the blends. The blend without an ionomer shows a sheath section that is almost like HDPE, indicating that HDPE forms a sleeve around the polyester component, whereas the core region shows distinct HDPE domains, spherical in shape, dispersed in the PBT matrix. There is no evidence of adhesion between the minor phase and the matrix, since the surfaces of HDPE particles are perfectly clean. The large cavities that can be seen represent the places occupied by HDPE that were taken away as a result of fracture. Since there is no adhesion between the phases, the mechanical properties of this blend have been relatively poor. Important morphological changes occurred on addition of ionomer. The blend having 2% ionomer shows a better dispersion of HDPE phases. The average size of the HDPE domains have also decreased a bit. The blend having 4% ionomer shows an even better dispersion of HDPE domains. The dimensions of the dispersed

HDPE domains have decreased significantly. It is also evident that the presence of ionomer has improved adhesion between the two phases since there appears to be some material coating of the surfaces of the dispersed phase. On increasing the ionomer content to 8%, a homogeneous distribution of HDPE domains in the PBT matrix is seen.

Another important morphological observation is that, as the ionomer content increases, the sheath and core morphology becomes less distinct. Similar observations have also been reported by Willis and Favis for polyamide/polyolefin blends with Surlyn as compatibilizer.²¹ It means that Surlyn is acting as an effective interfacial agent and has more compatibility with the HDPE phase than with PBT. The fracture surface of HDPE/ionomer (4%) blend does not differ much from that of pure HDPE, which again confirms that HDPE and Surlyn are miscible with each other. For 4% ionomer content, the sheath and core sections are, moreover, giving a similar appearance, whereas for 8% ionomer content composition, the sheath appears to be more like the PBT (pure) fracture surface.

SALS

The small-angle light-scattering (SALS) studies were done in the H_v mode of polarization. The patterns obtained are due to the anisotropy present in the crystalline superstructure due to the anisotropic fluctuations. For spherulitic crystalline superstructure, a four-leaf clover pattern is generally observed.²² Figure 6 shows H_v patterns obtained for the homopolymer and the blends at room temperature. In all cases, except where the pattern is sheaf-like, the spherulitic radii have been calculated using Stein's equation²³:

$$R = 1.025 \lambda / \sin (\theta_m / 2) \pi$$

where $\lambda = 6328 \text{ \AA}$ and θ_m is the angle of maximum intensity measured at an azimuthal angle of 45° and are tabulated in Table V.

Pure PBT shows a clear four-leaf clover pattern with the lobes along the polar directions ($0-90^\circ$), which is referred to as the unusual pattern. They exhibit a maximum intensity in each of the lobes, which is characteristic of spherulitic scattering. The spherulitic radius was calculated to be $3.26 \mu\text{m}$. Pure HDPE also shows four-leaf clover pattern characteristics of spherulitic crystalline superstructure with lobes at 45° to the polar directions and is referred to as the usual pattern. The spherulitic radius

for HDPE was calculated to be $4.33 \mu\text{m}$. The PBT/HDPE blend without ionomer also shows a four-leaf clover scattering pattern with lobes along the polar directions. In other words, the PBT superstructure dominates the morphology of the blend. The size of the pattern that is characteristic of spherulitic scattering has also increased, which means that the radius of spherulites has decreased on blending and the radius was found to be $1.35 \mu\text{m}$. On addition of 2% ionomer, it was seen that the pattern becomes more or less like a circular pattern with faint traces of a four-leaf clover pattern with lobes at 45° to the polars. This SALS pattern may be due to superimposition of scattering from PBT, HDPE, and the ionomer crystallites.

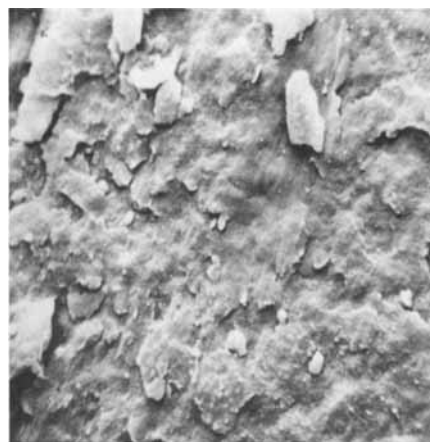
As we increase the ionomer content to 4%, the four-leaf clover pattern becomes clear and the scattering lobes are at 45° to the polars. The maximum in intensity is in the center with intensity decreasing as we go along the lobe. This is characteristic of a sheaflike pattern indicating a two-dimensional disc or platelike growth. For 8% ionomer content, a four-leaf clover pattern characteristic of spherulitic superstructure with lobes along the polars is observed. The size of the pattern has also increased, indicating a reduction in spherulitic size. The spherulitic radius was calculated to be $1.97 \mu\text{m}$. It is known that PBT forms unusual as well as the usual kind of spherulites. The presence of the ionomer is producing a condition that is leading to the formation of the usual kind of spherulites. Generally, such spherulites are formed at a low rate of crystallization; however, the reason for this is not obvious in this case. On the addition of 8% ionomer, the pattern reverts back to the unusual kind, thus returning to the situation favoring the formation of such spherulites. This is

Table V Morphological Parameters of PBT/HDPE/Ionomer Blends Obtained from SALS Study

Sample	Type of Superstructure	Spherulitic Radius (μm)
PBT	Spherulitic	3.26
HDPE	Spherulitic	4.33
PBT/HDPE	Spherulitic	1.35
PBT/HDPE/ionomer (2%)	Mixed/circular	—
PBT/HDPE/ionomer (4%)	Sheaflike	—
PBT/HDPE/ionomer (8%)	Spherulitic	1.97



PBT



HDPE



— 10 μm

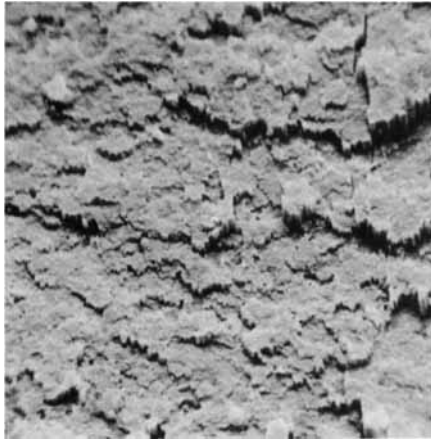
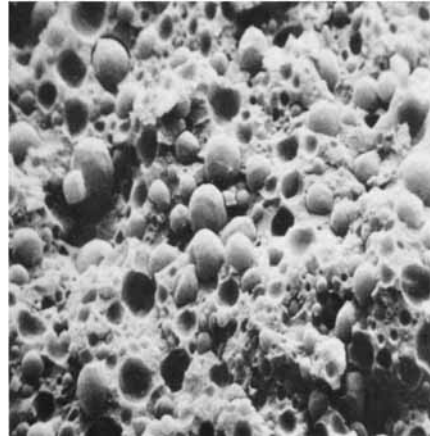
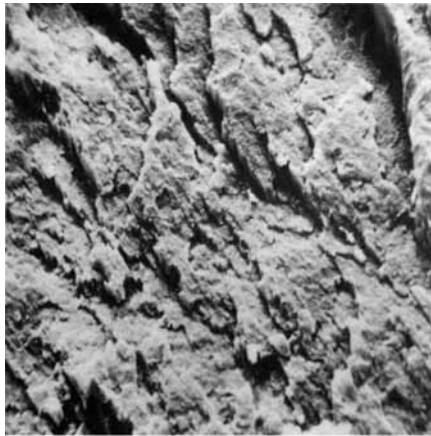
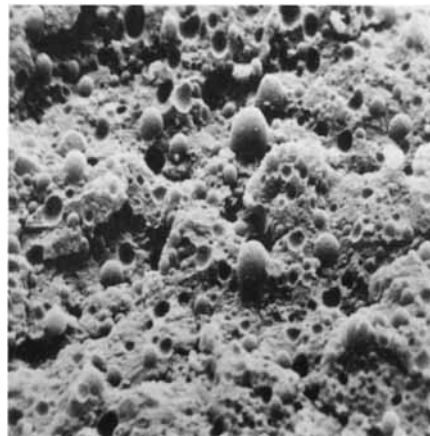
HDPE/IONO(4%)

Figure 5 SEM photomicrographs of liquid nitrogen fractured samples.

perhaps due to an increase in the rate of crystallization of PBT.

These observations indicate that blending of HDPE with PBT affected only the size of the spherulitic radius, decreasing from $3.26 \mu\text{m}$ for PBT to $1.35 \mu\text{m}$ for PBT/HDPE blend. This can be due to the difficulty posed by the HDPE-dispersed phase on PBT in arranging the polymer chains, and, hence, the growth of large spherulites is hindered.

On adding ionomer, we see that not only size but also the nature/type of superstructure formed has changed from spherulitic to sheaflike for 4% ionomer content. This indicates that in the presence of ionomer there is better mixing/adhesion of the two polymers, which has resulted in the significant observed change in morphology of the blend system. Thus, the SALS study also stands as proof of the influence of ionomer on the morphology of the PBT/

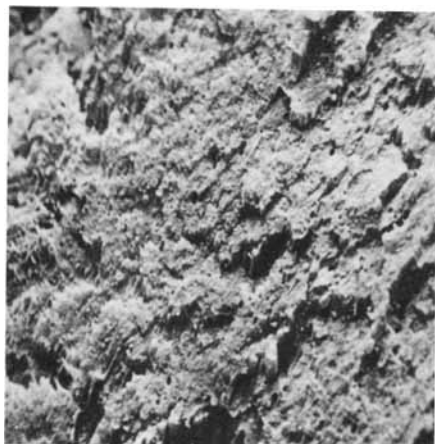
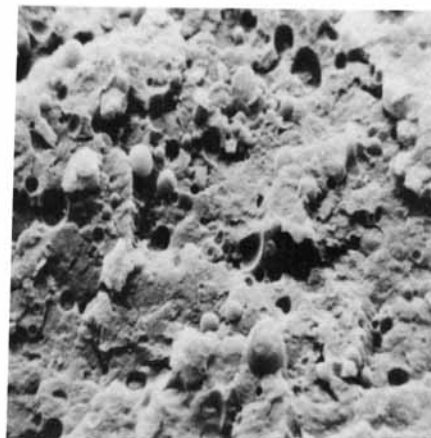
**(Sheath)****(Core)****PBT/HDPE****(Sheath)****(Core)****PBT/HDPE/IONO(2%)****Figure 5** (Continued from the previous page)

HDPE blend system. This is in agreement with the results obtained from DSC and polarizing microscopy.

Polarizing Microscopy

The photomicrographs taken on a polarizing microscope at room temperature using compression-

molded films are shown in Figure 7. PBT and HDPE show clear spherulitic crystalline structure. For the blend containing 80% PBT and 20% HDPE, two distinct regions can be clearly seen due to PBT and HDPE. The distribution of HDPE is not uniform, and large portions of HDPE domains can be seen dispersed in the PBT matrix. On adding 2% ionomer, distribution becomes much more even and the

**(Sheath)****(Core)****PBT/HDPE/IONO(4%)****(Sheath)****(Core)****PBT/HDPE/IONO(8%)****Figure 5** (Continued from the previous page)

region of HDPE domains become smaller. On increasing the ionomer content to 4%, the distribution becomes very good and HDPE domains become still smaller in size and more uniformly distributed.

On adding 8% ionomer, the PBT and HDPE phases completely depart from the matrix-dispersed phase morphology and a homogeneous dispersion is obtained that may be due to all three crystalline

polymers, since the ionomer also has a crystalline superstructure.

To study the dispersion of HDPE phases more clearly, blend samples were heated to 150°C on a hot stage, and photomicrographs taken are shown in Figure 8. At 150°C, HDPE and ionomer melt and the HDPE domains can be seen as black spots in the PBT matrix. The distribution of HDPE phases

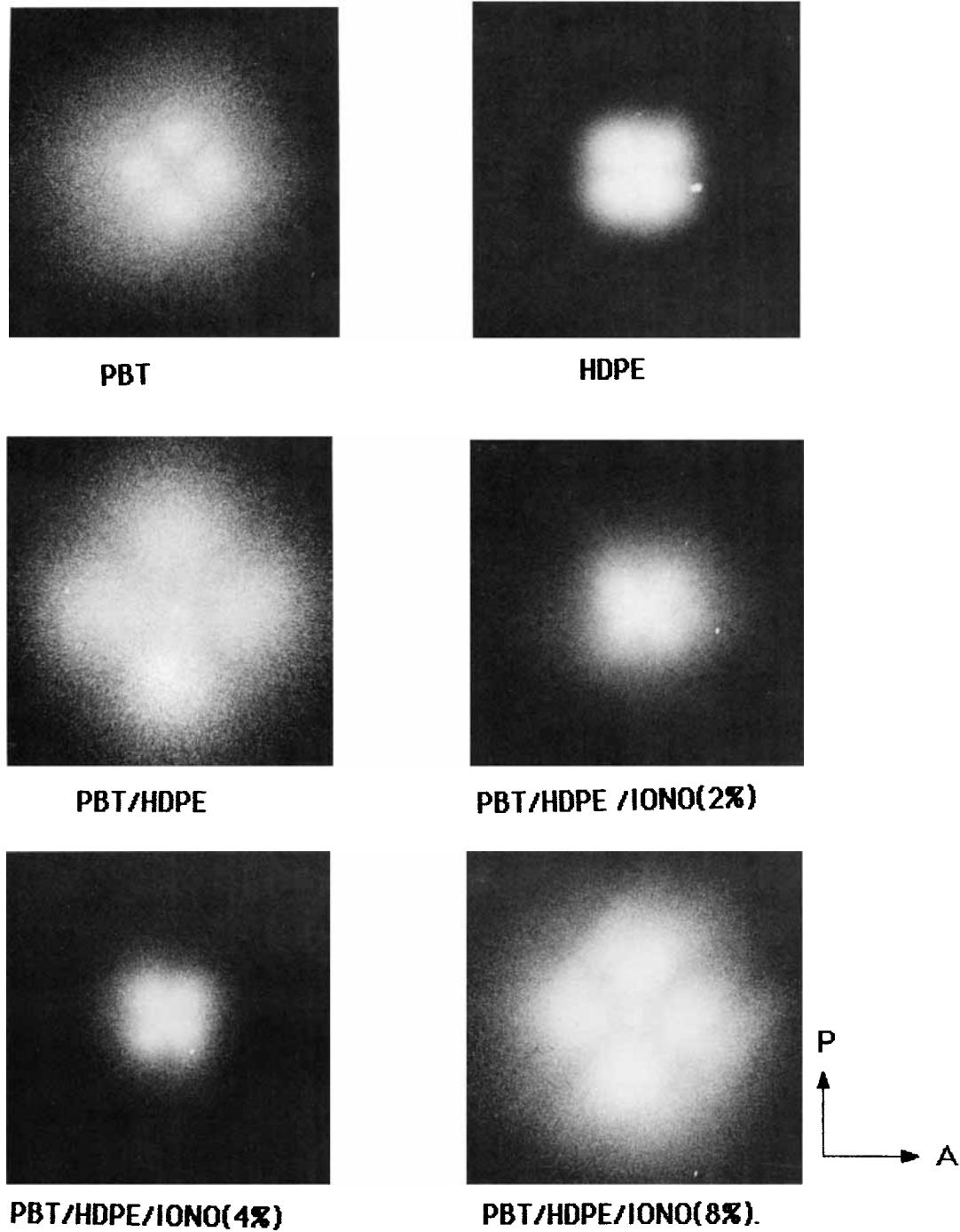


Figure 6 SALS, H , patterns for quenched films. (Sample to film distance is 4.7 cm.)

can be clearly seen. The PBT/HDPE blend without ionomer shows poor dispersion of the HDPE phase in the PBT matrix. Big droplets/domains of molten HDPE can be seen dispersed poorly in the PBT matrix. On addition of ionomer (4%), HDPE domains

become smaller and a little more uniformly dispersed. As the ionomer content is increased to 8%, the HDPE domains ultimately become indistinguishable and a homogeneous dispersion of HDPE domains in PBT is obtained. These results further

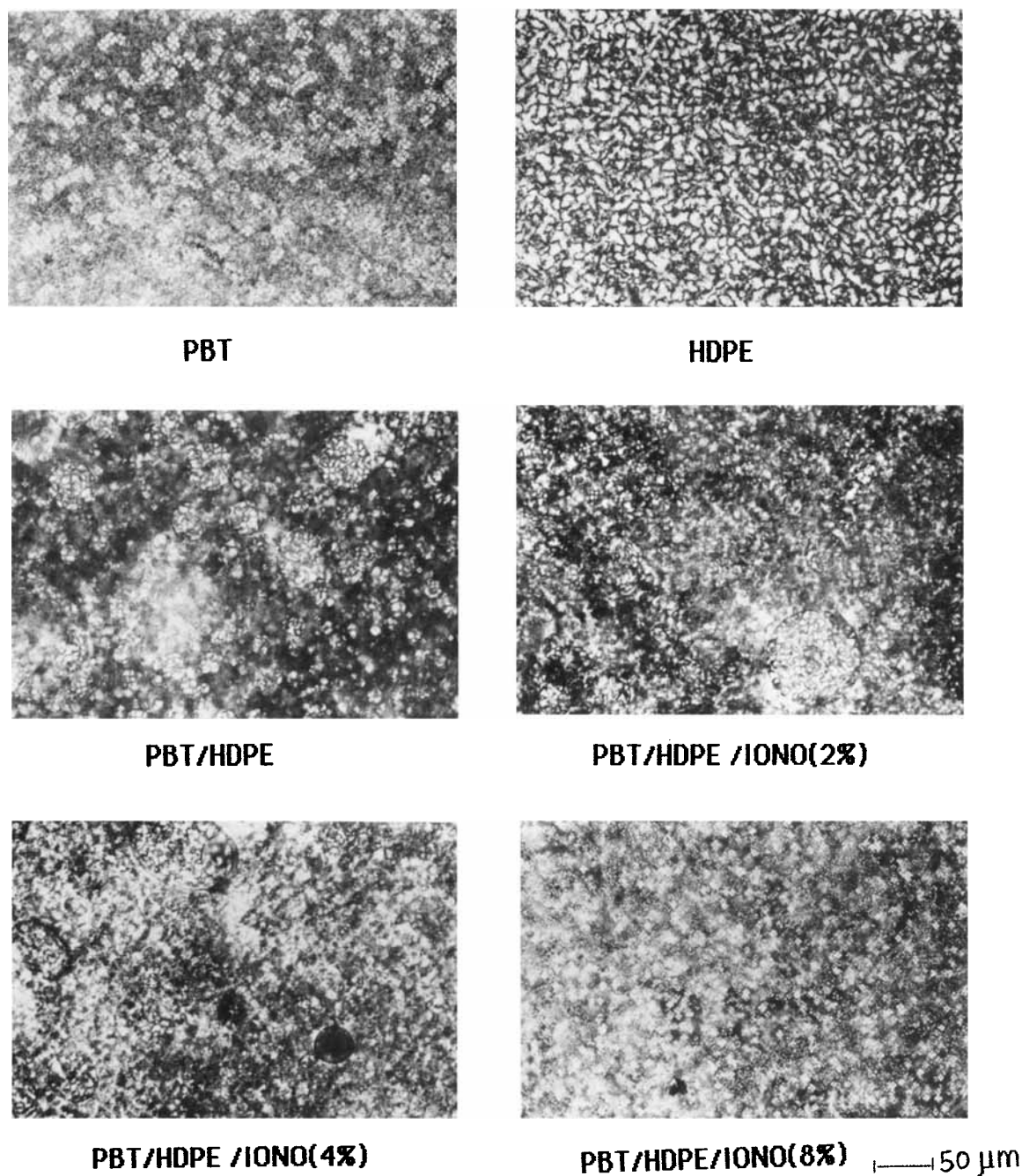


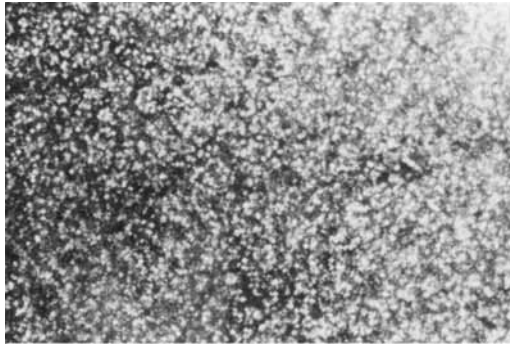
Figure 7 Polarized optical micrographs of quenched films at room temperature.

confirm that the ionomer "Surlyn" acts as an effective interfacial agent for the PBT/HDPE blend system.

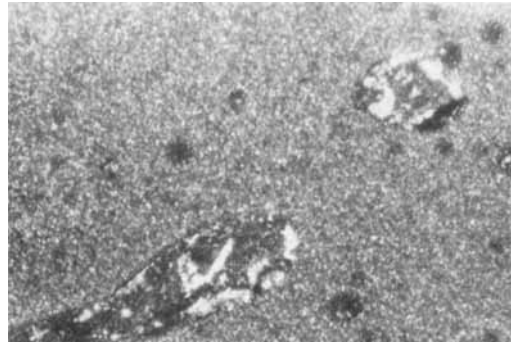
CONCLUSIONS

Based on the observations made, the following inferences have been drawn:

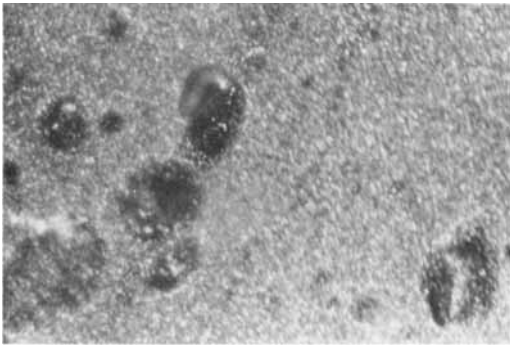
1. PBT and HDPE form a two-phase heterogeneous blend with little or no adhesion between the phases.
2. The ethylene-methacrylic acid copolymer partially neutralized by Na ions (ionomer, Surlyn 8660) used as compatibilizer improves the adhesion between the phases and, hence, the dispersion of HDPE domains in the PBT matrix.



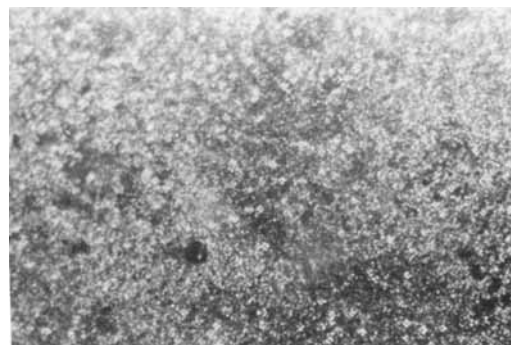
PBT



PBT/HDPE



PBT/HDPE/IONO(4%)



PBT/HDPE/IONO(8%)

Figure 8 Polarized optical micrographs of quenched films at 150°C.

3. DSC results show an increase in rate of crystallization of PBT on adding the ionomer.
4. DMTA results show two distinct T_g 's for both the blends with and without ionomer. One T_g (PBT) is associated with the PBT-rich region, and the other T_g (HDPE) is associated with the HDPE-rich region. On adding ionomer, more PBT is seen to dissolve in the HDPE-rich region, indicating better adhesion between the two polymers.
5. Polarizing micrographs show that HDPE exists as domains in the PBT matrix. Good dispersion of HDPE in PBT is obtained by adding an ionomer.
6. SALS results show that the addition of ionomer influences the type of crystallization of PBT. On adding ionomer, the crystalline superstructure changes from spherulitic to probably a mixture of several different types of superstructures.
7. SEM results also show a decrease in the HDPE domain size on addition of ionomer. The sheath-core morphology also seems to be less distinct in the presence of ionomer,

indicating better adhesion between PBT and HDPE phases.

8. In general, the PBT/HDPE/ionomer blend system can be regarded as a polymer alloy on the basis of above-mentioned properties. An attempt will be made to relate these morphological changes on mechanical properties and also to correlate morphology with rheology in a future publication.

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