## The Structure and Properties of Drawn Blends of Polyethylene and Polypropylene

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#### **SYNOPSIS**

A series of highly oriented tapes has been prepared from a blend consisting of equal proportions of polyethylene and polypropylene. The mechanical properties and the structure and morphology of the samples have been investigated using DSC, optical microscopy, and wide angle and small angle diffraction, including measurements of crystal strain on samples under stress. It has been confirmed that the blend is incompatible, and a structural model has been proposed which is consistent with the observation that the polyethylene and polypropylene components act essentially independently in their response to external macroscopic stress.

## INTRODUCTION

During recent years there has been a substantial number of publications (e.g., Refs. 1-10) concerned with blends of polyethylene (PE) and polypropylene (PP), many of which have discussed the detailed structure of isotropic samples, and a few oriented samples. The present paper describes the preparation, structure, and mechanical properties of highly drawn tapes prepared from a 50/50 blend of PE and PP. The processing conditions follow closely these developed at Leeds University<sup>11</sup> to produce high modulus PE and PP. We have also followed the methodology developed in our laboratory to establish morphological models for these homopolymers.<sup>12-14</sup>

A key structural technique is to investigate the strain response of the crystalline regions of the material when an external extensional load is applied to a drawn fiber or tape. This has proved valuable in studies of highly drawn polyethylene,<sup>12</sup> polyoxymethylene,<sup>13</sup> poly(ethylene terephthalate)<sup>14</sup> homopolymers and has again shown its usefulness in the present study.

A relatively simple model is shown to account for the observed properties of the blended tapes very satisfactorily and it seems that, although quite intimately blended, the two components of the incompatible blend act, to a certain extent, independently, although they are mechanically linked to one another.

## **EXPERIMENTAL**

## **Preparation of the Materials**

The commercial polymer grades used in this work are listed in Table I, together with the value of weight average molecular weight and melt flow index (MFI). The PE/PP blend is known to be incompatible and so, in order to produce as coherent a blend as possible, grades of PE and PP of similar MFI were used.<sup>15</sup> To produce a good pellet mix, equal masses of each component were premixed for 1 h in square drum rotating at 40 rpm. This mixture was then fed into a 25 mm single-screw extruder with a metered screw. The barrel temperature was monitored and controlled by three thermostats, the temperature rising as the blend proceeded along the extruder. The die temperature was also controlled by a thermostat and was adjusted, together with the barrel temperatures, to yield as uniform an output as possible. Barrel temperatures of 160, 190, and 260°C were used with a die temperature of 240°C. The extrudates, produced in the form of a 1 mm diameter monofilament from a die pack consisting

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Polymer	Grade	Supplier	$ar{M}_w/10^3$	MFI
РР	HM 61	Shell	425ª	1.5
PE	Rigidex 006-60	BP Chemicals Ltd.	126	0.7

Table I Weight Average Molecular Weights and Melt Flow Indices of the Homopolymers Used

<sup>a</sup> Value supplied by RAPRA Technology.

of spinneret and fine gauze, were cooled in air. The monofilament, produced using a screw speed of around 10 rpm, was uniform and opaque.

Compression-molded sheets were prepared by pressing cut segments between brass plates at 200°C. A pressure of 2 MPa was applied for 5 min to allow the blend to melt and spread out between the plates. This was then increased to 15 MPa for a further 5 min, after which time the pressure was removed and the sheets quenched in water at room temperature.

To facilitate comparison of the blend properties with those of its components, sheets of the PE and PP homopolymers were produced using the same procedure and conditions, but pressed directly from the pellets.

#### **Characterization of the Isotropic Blends**

The first step in characterizing any polymer mixture is to determine whether the two homopolymers have formed a compatible blend. Compatible blends result when two homopolymers mix on a segmental level, giving rise to a material with a single melting point. A Perkin Elmer DSC-2 was used to investigate the thermal behavior of the homopolymers and of the as-extruded and melt-pressed samples of the blend over the temperature range +50-210 °C at a scanning rate of 10 °C/min. Typical thermograms are shown in Figure 1.

The PE and PP homopolymers have melting points of 135 and 162°C, respectively, which are retained in the blend. It is therefore clear that the blend thermogram is simply a superposition of the two homopolymer traces. This indicates that the PE and PP crystallize separately in the blend, i.e., no cocrystallization has taken place. This result is consistent with the reported immiscibility<sup>15</sup> of the polymers and the attendant incompatibility of the system. The same conclusion applies to the as-extruded blend, which produced a thermogram identical to that of the melt-pressed sample.

#### X-Ray Characterization

Wide-angle X-ray scattering patterns for the blend and its component homopolymers are shown in Figure 2. It can be seen that the WAXS photographic pattern for the blend is equivalent to a superposition of the individual homopolymer characteristic reflection patterns. To the extent that the two components have crystallized separately, this confirms that the blend is incompatible, although there could still be some degree of mixing in the amorphous component.

The homopolymers involved in this work usually crystallize as spherulites with a lamellar morphology so small angle X-ray studies were carried out using a Franks-type camera to investigate the morphology of the blend. A small-angle X-ray photograph of the isotropic blend is shown in Figure 3. The broad halo suggests that a lamellar structure may be present, but it is not possible, at this stage, to differentiate between the homopolymer contributions.

## **Hot-Stage Microscopy**

Hot-stage microscopy was used to determine the degree of mixing present in the melt-pressed blends. We have seen that PE and PP are incompatible in



Figure 1 DSC thermograms showing the melting endotherms of isotropic samples of: (a) PE; (b) PP; (c) PE/PP.



Figure 2 WAXS photographs of isotropic samples of: (a) PE; (b) PP; (c) PE/PP.



Figure 3 SAXS photograph of the isotropic PE/PP blend.

the blend but have no indication of the size of the homopolymer domains. The investigation was carried out using a Leitz-Wetzlar (SM-LUX-POL) polarizing microscope fitted with a Leitz-Wetzlar 350 "hot-stage." A small sliver of the material to be studied was taken and placed between a glass slide and slip on the hot-stage, set at 200°C. In order to produce a thin transparent film, suitable for optical microscopy, the polymer, once molten, was pressed between the glass slide and slip.

The blend sample was cooled to room temperature to allow crystallinity to develop. From the DSC thermograms it is apparent that determination of the PP domain sizes can be obtained by heating the blend up to 150°C, thus melting the PE crystallites. The temperature of the hot stage was therefore set to 150°C and left for 30 min. When viewed through crossed polarizers, the characteristic Maltese Cross patterns of spherulitic structures were observed, and good mixing of the two components was evident, the average size of the PP spherulites being about 25  $\mu$ m. On cooling to room temperature, the space between the PP spherulites was taken up by smaller PE spherulites.

#### **Preparation of Drawn Samples**

Dumbbell-shaped samples, 5 mm wide  $\times$  20 mm long, were stamped out of the isotropic blend sheets and drawn at a crosshead speed of 100 mm/min, a strain rate which has proved suitable for the successful drawing of both homopolymers.<sup>16,17</sup>

The best drawing temperatures for the individual homopolymers have been found to be  $75^{\circ}$ C for PE<sup>16</sup> and 110°C for PP.<sup>17</sup> Hence, the blend was drawn at both 75 and 110°C as well as at an intermediate

temperature, 90°C. By far the best drawing results were obtained at  $110^{\circ}$ C and this temperature was subsequently used for all drawing operations. Oriented samples were then produced with draw ratios ranging from the natural draw ratio up to the maximum draw ratio attainable. To aid comparison with the homopolymers, a series of dumbbell samples of each homopolymer, obtained from the melt-pressed sheets was drawn under the same conditions as the blend to a similar range of draw ratios.

Upon drawing, the samples necked with a natural draw ratio of  $\times$  7 which compares with  $\times$  6 for PE and  $\times$  5 for PP under similar conditions. Each of the drawn samples was transparent with no sign of the stress whitening, caused by voiding, evident in the PP homopolymer drawn beyond  $\times 10$ . The loadextension curves for PE, PP, and the blend are plotted in Figure 4, the formation of a neck for all three materials being indicated by the distinctive yield drops observed in each curve. The blend curve lies between those of its components and an analysis of the nominal yield stress of the samples investigated reveals that the yield stress of the blend (7.07 MPa) is halfway between those of PE (5.4 MPa) and PP (8.5 MPa). Thus, during drawing, the blend appears to behave as a simple combination of the two constituent homopolymers. The maximum PE/PP blend draw ratio obtained was  $\times$  16.5, which, again, was intermediate between those of PE ( $\times$  19) and PP ( $\times$  15.5) achieved under the same conditions. All the highly drawn samples possessed a fibrillar texture. At 75°C the PE homopolymer was drawn to much higher draw ratios ( $\times 25$ ); but PP has poor drawing properties at this temperature and a maximum blend draw ratio of  $\times$  8 was obtained. This



Figure 4 Load-extension curves for: (a) PP; (b) PE/ PP; (c) PE.

implies that the drawability of this particular blend under given conditions is determined by the drawability of the PP component and suggests that the drawing properties of any binary blend might be determined by the properties of the least drawable of the two components.

In some respects it is surprising that this incompatible blend can be drawn to such high draw ratios. Previous attempts<sup>15</sup> at producing highly drawn PE/ PP used draw temperatures approaching the melting point of PE and hence sacrificed ultimate axial stiffness for high draw ratios.

## Extensional Young's Moduli of the Oriented Samples

The 10-s isochronal creep moduli of the oriented blend samples were measured using a standard deadload creep apparatus,<sup>18</sup> and the usual conditioning procedures, i.e., the maximum load to be used in the modulus determination (the load required to produce approximately 0.4% extension), was applied in a cyclic fashion—10 s on with 100 s recovery time, this being repeated until the extension obtained was reproducible. The stress-strain curves of the drawn samples were linear up to approximately 0.2%, and Young's moduli were evaluated at 0.1% strain.



Figure 5 Ten-second isochronal creep modulus vs. draw ratio for PE, PP, and PE/PP.



**Figure 6** WAXS photograph of PE/PP drawn  $\times$  7 at 110°C.

The measured moduli of the homopolymers and the blends are shown in Figure 5, and it can be seen that, as is usually the case, the modulus increases with draw ratio, the moduli of the blend samples lying between those of its components at similar draw ratios. This indicates that the PE and PP in the blend are both contributing significantly, if not equally, to its axial stiffness.

#### X-Ray Characterization of the Drawn Blends

WAXS was used to investigate, qualitatively, the relative orientations of the two components in the blend. The formation of a neck on drawing ensures that the randomly oriented isotropic material very rapidly becomes highly oriented in the necked region. Figure 6 shows an X-ray photograph of a sample with a draw ratio of 7 and shows that both components appear to be oriented to a similar extent implying simultaneous orientation on drawing. The more highly drawn samples show increasing orientation with draw ratio.

SAXS photographs of drawn samples were recorded with the sample aligned with the draw direction perpendicular to the beam. Figure 7 shows the SAXS pattern for the draw ratio 7 sample and is seen to be a simple two-point pattern (the original negative is more distinct), indicative of an oriented lamellar structure. As the draw ratio increases, the pattern becomes less distinct, which has been noted before in polyethylene.<sup>19,20</sup> A study of drawn polypropylene by Duxbury<sup>21</sup> failed to observe any distinct small angle pattern. Hence the blend pattern is probably produced solely by the PE component,



Figure 7 SAXS photograph of PE/PP drawn  $\times$  7 at 110°C.

a conclusion supported by the absence of any sign of a double peak on microdensitometer scans. The observed long period was found to be  $280 \pm 30$  Å in agreement with values obtained by Clements et al.<sup>22</sup> for PE homopolymer drawn at  $115^{\circ}$ C—the long period being highly dependent on draw temperature.

The morphology of the samples was investigated further by estimating crystallite sizes from the breadths of the related X-ray diffraction profiles. The integral breadth of an X-ray line arising from size broadening is related to crystallite size by the Scherrer equation:

$$\Delta(2\theta) = k\lambda/L(hkl)\cos(\theta)$$

where L(hkl) is the average crystallite size in the direction perpendicular to the hkl plane and  $\theta$  is the Bragg angle for that reflection. k is the shape factor, which has been shown by Stokes and Wilson<sup>23</sup> to vary from 0.9 to 1.3, depending on crystallite shape and the reflection being studied. In the absence of any further information and because crystallite shapes in such systems are very ill defined, a value of unity was chosen.

The size-broadening profile was obtained from the observed profile by deconvoluting the latter from the instrumental profile, which was determined using a specially prepared copper foil standard sample having a carefully controlled range of crystallite sizes. The deconvolution process was carried out using the method of Stokes.<sup>24</sup>

The size-broadening profile is not solely dependent on crystallite size but is also affected by crystal disorder effects, which can be evaluated reasonably simply if more than one order of the reflection under analysis can be investigated. In the present case, however, as in the polyethylene homopolymer, only one reflection is available for the c-axis direction so that no simple method is available for separating size and disorder effects.

It has already been inferred that the long period pattern observed on small-angle photographs of drawn samples of the blend may be attributed to the PE component only. Previous studies of highly drawn PE have shown that the mean crystallite size can be greater than the long period in the draw direction, and this has led to be establishment of the crystal bridge model, which was very successful in accounting for the mechanical properties of the materials.<sup>25</sup>

Crystallite sizes for PE in the drawn blend were determined using a scanning diffractometer with entrance and exit slits set to give an instrumental breadth (full width half maximum) of about 0.1°  $(2\theta)$ . The observed integral breadths of the polyethylene (002) reflection were never less than about 1° so that the deconvolution was quite reliable. The assumption has to be made, as it was in the polyethylene case, that disorder contributed negligibly to the experimentally determined line widths. Thus the Scherrer equation was used directly and the mean crystallite lengths in the *c*-axis direction were determined to be 220, 250, and 270 Å for the  $\times$  9,  $\times$  12, and  $\times$  15 samples, respectively.

It is seen that, as for the homopolymer, the crystallite sizes increase with draw ratio and they approach the observed long period of 280 Å. Although they do not exceed the latter, the close correspondence between the figures does hint that crystal bridges may be formed upon drawing the blend, which is another indication that the drawn blend may be considered equivalent to a simple combination of the two drawn homopolymers, each component possessing the structure of the similarly drawn homopolymer.

The orientation of the two components of the drawn blend was measured to ascertain whether there was any difference between them. The measurement of PE crystallite orientation is straightforward. The [002] reflection was observed in the diffractometer with the sample held normal to the diffractometer axis and the sample was then rotated around that axis and the reflected intensity recorded. After corrections for background and variation in volume illumination,<sup>18</sup> the intensity variation was analyzed to determine the orientation functions,  $\langle P_2(\cos \theta) \rangle$  and  $\langle P_4(\cos \theta) \rangle$ .

Polypropylene, as a result of its monoclinic crystal structure, does not exhibit a simple meridional re-

flection. The nearest, and hence most suitable, candidate is the  $(\bar{1}13)$  reflection which occurs approximately 11° off the meridian. In order to determine the *c*-axis orientations from measurements upon the  $(\bar{1}13)$  reflection, use was made of the orthogonal relationship between Legendre functions. Analysis of measurements of the  $(\bar{1}13)$  reflection (after correction) yield raw values of  $P_2$  and  $P_4$ . The values of  $P_2$  and  $P_4$  for the *c*-axis are found from the expression

$$\langle P_n(\cos \phi_c) \rangle = \langle P_n(\cos \phi_{113}) \rangle / P_n(\cos \delta_{113})$$

where  $\delta_{113}$  is the angle between the ( $\overline{1}13$ ) plane normal and the crystallite *c*-axis and  $\phi_c$  is the angle between the crystallite *c*-axis and the draw direction. Values of  $P_2$  and  $P_4$  for both PE and PP in the drawn blend are shown in Table II. These show that there is a very high degree of orientation, which only increases slightly with draw ratio. It is also clear that both components are oriented to similar extents within the blend.

#### Crystal Strain Measurements and Determination of Apparent Crystal Moduli

So far we have seen that two components of the blends are phase-separated in both the isotropic and drawn states. Also, Young's modulus measurements for the drawn blend were compared with results for similarly drawn homopolymers and the blend moduli were found to lie approximately halfway between those of the components at comparable draw ratios.

Further insight into the structure of the blends can be obtained by making measurements of crystal strain, as have been made in previous work on homopolymers. In this case it is necessary to make measurements on both components of the blend to determine the extent to which the applied stress is distributed between the components. To determine crystal strains accurately, a detection system is required which can determine changes in the Bragg

Table IIOrientation Moment Averages for thePE and PP Components in the Drawn Blend

	$\langle P_2(\mathbf{c}$	$\langle P_2(\cos  heta)  angle$		$\langle P_4(\cos  heta)  angle$	
Draw Ratio	PE	PP	PE	PP	
	±0.	005	±0	.01	
	0.986	0.983	0.955	0.942	
12	0.988	0.989	0.962	0.964	
15	0.993	0.991	0.977	0.970	

Table IIIRoom Temperature Apparent CrystalModuli Results for the Drawn Blend andfor the Two Homopolymers

		$E_c(app)$ (GPa)		
Sample	Draw Ratio	PP Component	PE Component	
Blend	9	$24 \pm 2$	$240 \pm 20$	
Blend	12	$22 \pm 2$	$245 \pm 25$	
Blend	15	$23 \pm 2$	$235 \pm 20$	
PE	12	_	$190 \pm 30$	
PP	12	$28 \pm 2$	_	

angles of X-ray reflections to within a few thousandths of a degree. In our measurements we used a linear position sensitive detector, with which such measurements can be made relatively straightforwardly.<sup>14</sup>

Analysis of the PSD data was carried out using a program which fits a combined Gaussian and Lorentzian profile with constant background to the data. Measurements on the polyethylene component were straightforward since the intense meridional (002) reflection at  $2\theta = 74.4^{\circ}$  was used.

For the polypropylene component, the  $(\bar{1}13)$  reflection was used and a correction was applied to take account of its nonmeridional nature. In addition to measurements at room temperature, the variation of the apparent crystal modulus,  $E_c(app)$ , with temperature for both components was investigated but only for the draw ratio 12 sample since all three samples gave similar room temperature results. Values of  $E_c(app)$  were measured at 0, -30, -60, and -100 °C for both components of the blend as well as for the PP homopolymer for comparison.

The room temperature results for the apparent crystal moduli of PE and PP in the three drawn blend samples are shown in Table III. For each component the apparent crystal modulus remains constant with draw ratio, as expected. What is surprising is the actual values of the moduli. The average value for the polyethylene component is around 245 GPa, which is consistent with the predicted theoretical modulus and, at first glance, appears to be an unexceptional result. However, previous work carried out by Clements on the crystal strain of polyethylene<sup>12</sup> produced values of 150 GPa for unannealed drawn samples at room temperature. The tapes in that study had been drawn at 75°C. Analysis of the PE homopolymer produced in a similar fashion to the blend gave a room temperature value of apparent crystal modulus of 190 GPa, which



Figure 8 Apparent crystal modulus vs. temperature for the PE component in the PE/PP blend drawn to  $\times$  12.

is still well below that obtained in the blend. The PP component behaved in the opposite fashion, the blend gave a value of 23 GPa, wheras the homopolymer value was measured to be 28 GPa. Figures 8 and 9 show the variation with temperature of  $E_c(app)$  for both components. The results for PE are quite unexpected and a complete reversal of what is generally seen in the homopolymer. In PE homopolymer,  $E_c(app)$  rises from its value at room temperature to a value approaching that of the theoretical crystal modulus as the temperature is *reduced*. This arises because of the stiffening up of the amorphous component.

The PP component appears to behave as one would expect. However, measurements carried out on the temperature dependence of the PP  $E_c(app)$  for the oriented homopolymer gave the results shown in Figure 10. These show a small rise from 28 to 35 GPa in the temperature range concerned compared with a rise from 22 to 42 GPa observed in the blend. Thus the temperature-dependent behavior of the polypropylene component in the blend cannot be simply explained by direct reference to the homopolymer.

It seems therefore that a change of stress distribution is taking place between the constituents as the temperature is reduced. At room temperature there is stress concentration on the PP component, but with decreasing temperature the PE takes up more of the applied stress until at  $-30^{\circ}$ C the stress is concentrated on the PE crystallites and reduced on the PP component.





Figure 9 Apparent crystal modulus vs. temperature for the PP component in the PE/PP blend drawn to  $\times$  12.

Figure 10 Apparent crystal modulus vs. temperature for PP homopolymer drawn to  $\times$  12.

## DISCUSSION

# Development of a Mechanical Model for the Blend

In order to devise a mechanical model for the material, we require values both of the Young's modulus and of the apparent crystal modulus over a range of temperatures. A problem arises at room temperature and at  $0^{\circ}$ C because of the nonlinear nature of the polyethylene stress-strain curve (Fig. 11), the consequence of which is that the distribution of either stress or strain (depending on the structure of the blend) between the constituents may vary with the load which is applied to a blend sample.

Strains of up to 1% had to be used in the crystal strain experiment at room temperature and a value for the polyethylene modulus was used in the modeling, which was the average of the tangent moduli at 0.1 and 1.0% strains. At low temperatures the maximum strains were smaller and the 0.1% value was used. There was no problem with the polypropylene component because the stress-strain relationship was found to be linear over the range considered.

The temperature dependence of the Young's moduli for the PE and PP homopolymers using these modified figures is shown in Figure 12. Comparison of these data with those for  $E_c(app)$  for the blend show up a definite relationship, with the crossover



Figure 11 Thirty-second isochronal creep response at room temperature for PE homopolymer drawn to  $\times$  12.

Ф-Ф РЕ а-а РР G-0 РЕ/РР



Figure 12 Variation of isochronal creep modulus with temperature for drawn  $\times$  12: PE (30 s), PP (10 s) and PE/PP (10 s).

point in the homopolymer moduli occurring in the same temperature range (between 0 and  $-30^{\circ}$ C) as the largest changes in  $E_c(app)$  for the blend components (Figs. 8 and 9).

It is clear that the blend, owing to its incompatible nature, has four separate phases, two crystalline and two amorphous, so that the mechanical modeling is, in principle, very complicated. However, for the sake of simplicity, we start with a model which is constructed using PE and PP as the basic blocks, ignoring, for the moment, the inherent two-phase structure of each. Such an approach is justified since the components retain their separate crystalline identities and orient equally in the blend.

The two simplest models are the series and the parallel models. The series model predicts a value of  $E_c(app)$ , which is always equal to the theoretical value and since measurements show this not to be the case we dismiss this possibility.

In the parallel model the Young's modulus,  $E_p$ , of the blend is given by

$$E_p = v_1 E_1 + v_2 E_2 \tag{1}$$

where  $v_{1,2}$  and  $E_{1,2}$  are the volume fraction and Young's moduli of the respective components.

Temp (°C)	1	Measured Values (GPa)			Predicted Values (GPa)	
	E(PP)	E(PE)	E(Blend)	Series Model	Parallel Model	
21	13	12	14	$12.5 \pm 1.3$	$12.5\pm1.0$	
0	16.2	19	18.3	$17.4 \pm 1.8$	$17.6 \pm 1.6$	
-30	18.1	28	24	$21.8\pm2.3$	$22.9\pm2.0$	
-60	19.8	31	26	$24.0 \pm 3.0$	$25.2\pm2.5$	
-90	21	30.2	27.5	$24.6 \pm 3.3$	$25.5 \pm 3.0$	

 Table IV
 Comparison of Measured Young's Moduli for the Homopolymers and the Blend, and Predicted

 Values for the Blend from the Series and Parallel Models, as a Function of Temperature<sup>a</sup>

<sup>a</sup> All were drawn to a draw ratio of 12.

Table IV shows that  $E_p$  is predicted quite satisfactorily by this model, but it is on consideration of the apparent crystal modulus results that the advantages of the parallel model over the series become most obvious. At room temperature PE is less stiff than PP, resulting in stress concentration on the PP, giving rise to an increased PP crystal strain and a correspondingly decreased PE crystal strain. As the temperature decreases PE stiffens up more quickly than PP causing a change in the stress distribution within the sample, the stress now being concentrated on the PE component and diminished in the PP producing the switch over in crystal strains. This corresponds exactly with the observed variations in  $E_c(app)$  with temperature.

To calculate predicted values of  $E_c(app)$  of the constituents in the blend, account must be taken of both the inhomogeneous stress distribution and different volume fractions of components present. First consider the blend. The  $E_c(app)$  of either component is given by

$$E_c(\text{app}) = \sigma_m / e_c \tag{2}$$

where  $\sigma_m$  is the macroscopic stress and  $e_c$  is the crystal strain. For comparison with the  $E_c(app)$  homopolymer values,  $\sigma_m$  must be replaced by the stress actually applied to the component. Now

$$\sigma_m = v_1 \sigma_1 + v_2 \sigma_2 \tag{3}$$

and

$$\sigma_1/\sigma_2 = E_1/E_2 \tag{4}$$

In the case of constituent 1, if  $\sigma_m$  is replaced by  $\sigma_1$ , then the resultant  $E_c(app)$  should be equivalent to that observed in the homopolymer. Thus

$$\frac{E_c(\text{app}) \text{[blend]}}{E_c(\text{app}) \text{[bnoopolymer]}} = \frac{v_1 E_1 + v_2 E_2}{E_h} \quad (5)$$

where  $E_h$  is the Young's modulus of the homopolymer.

This argument ignores the two-phase nature of each component and assumes that the morphology of the individual components is precisely maintained in the blend.  $\sigma_1$  (or  $\sigma_2$ ) represents the stress acting on component 1 (or 2) but is generally not equal to the stress acting on the crystalline fraction of the component. Thus the values obtained for the modified  $E_c$  (app) should agree with those observed in the homopolymer under the same conditions but will not necessarily equal the true crystal modulus. This is especially true in systems where the compliance of the amorphous component changes significantly with temperature.

Now the measured apparent crystal modulus in both homopolymers changes over the temperature range of interest.  $E_c$  (app) for PP rises from 28 GPa at room temperature to 35 GPa at  $-100^{\circ}$ C and, over the same range, the PE  $E_c$  (app) rises from 190 to 240 GPa. At  $-90^{\circ}$ C the Young's moduli for PP and PE are 21 and 30.2 GPa, respectively.

Thus, a stress applied to the parallel combination would be divided in the ratio 21/51.2:30.2/51.2, i.e., 42% of the macroscopic stress will be taken up by the PP and 58% by the PE. Hence, the PE  $E_c(app)$  will be reduced and the PP  $E_c(app)$  enhanced in comparison with the homopolymer. At 21°C the situation is reversed: PP is stiffer than PE, which produces an enhanced PE  $E_c(app)$  and diminished PP  $E_c(app)$ .

Table V shows the values obtained for the blend crystal moduli when the inhomogeneous stress distribution between the components has been accommodated using eq. (5). The results obtained fit the

Table V	Comparison of the Apparent Crystal
Moduli of	the Components of the Drawn Blend
Measured	at –100 and 21°C and Those
Predicted	by the Parallel Model

	$E_c(app)$ (GPa)			
Temp (°C)		Blend		
	Homopolymer (Measured)	(Measured)	(Predicted)	
-100 PE	250 (Ref. 17)	$185 \pm 15$	$210 \pm 30$	
-100 PP	35	$42 \pm 3$	$42 \pm 4$	
21 PE	190	$245\pm20$	$245 \pm 40$	
21 PP	28	$22\pm2$	$22 \pm 4$	

homopolymer measurements well within the experimental errors, indicating that the parallel model is a good representation of the mechanical structure of this particular drawn blend. It can also be concluded that the structure of each component is close to that of its similarly drawn homopolymer.

## **CONCLUSIONS**

We have investigated the structure and properties of a particular blend of two specific grades of polyethylene and polypropylene. The study has indicated that the properties of the oriented blend can be well described by a simple parallel model in which the two components, although mechanically linked, act more or less independently. This suggests that it should be possible to manufacture a material, with properties tailored to a desired specification, by choosing suitable grades of homopolymer and blending them in predetermined proportions.

#### REFERENCES

- R. E. Robertson and D. R. Paul, J. Appl. Eng. Sci., 17, 2579 (1973).
- O. F. Noel and J. F. Carley, Polym. Eng. Sci., 15, 117 (1975).

- W. Wenig and K. Meyer, Coll. Polym. Sci., 258, 1009 (1980).
- A. K. Gupta, V. B. Gupta, R. H. Peters, W. G. Harland, and J. P. Berry, J. Appl. Polym. Sci., 27, 4669 (1982).
- E. Martuscelli, M. Pracella, G. D. Volpe, and P. Greco, Makromol. Chem., 185, 1041 (1984).
- M. Kojima and H. Satake, J. Polym. Sci. Polym. Phys. Ed., 22, 285 (1984).
- 7. E. S. Shermann, J. Mater. Sci., 19, 4014 (1984).
- B. Lotz and J. C. Wittman, J. Polym. Sci. Polym. Phys. Ed., 25, 1079 (1987).
- C. Sawataria, S. Shimogiri, and M. Matsao, *Macromol.*, 20, 1033 (1987).
- 10. G. Rizzo and G. Sparado, Eur. Polym. J., 24, 303 (1988).
- 11. G. Capaccio and I. M. Ward, Polymer, 15, 233 (1974).
- J. Clements, R. Jakeways, and I. M. Ward, *Polymer*, 19, 639 (1978).
- S. Jungitz, R. Jakeways, and I. M. Ward, *Polymer*, 27, 1651 (1986).
- 14. T. Thistlethwaite, R. Jakeways, and I. M. Ward, *Polymer*, **29**, 61 (1988).
- S. Danesi, in *Polymer Blends: Processing, Morphology* and *Properties*, M. Kryszewski, A. Galeski, and E. Martuscelli, Eds., Plenum Press, New York, London, 1979, Vol. 2, p. 35.
- G. Capaccio, T. A. Crompton, and I. M. Ward, J. Polym. Sci. Polym. Phys. Ed., 14, 1641 (1976).
- 17. A. J. Wills, G. Capaccio, and I. M. Ward, J. Polym. Sci., 18, 493 (1980).
- C. J. Morgan and I. M. Ward, J. Mech. Phys. Solids, 19, 165 (1971).
- A. Peterlin and G. Meinel, Appl. Polym. Symp., 2, 85 (1966).
- F. M. Willmouth, A. Keller, I. M. Ward, and T. Williams, J. Polym. Sci. A-2, 6, 1627 (1962).
- 21. J. Duxbury, Ph.D. thesis, The University of Leeds, 1986.
- J. Clements, Ph.D. thesis, The University of Leeds, 1978.
- A. R. Stokes and J. C. Wilson, Proc. Camb. Phil. Soc., 38, 313 (1942).
- 24. A. R. Stokes, Proc. Phys. Soc. (London), 61, 382 (1948).
- J. Clements, R. Jakeways, and I. M. Ward, *Polymer*, 19, 639 (1978).

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