

Correlations Between Processing Parameters, Morphology, and Properties of Blown Films of LLDPE/LDPE Blends, Part 2: Crystalline and Amorphous Biaxial Orientation by WAXD Pole Figures

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Received 9 May 2005; accepted 10 February 2006

DOI 10.1002/app.24403

Published online in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The biaxial molecular orientation of blown films made of blends of linear low density polyethylene (LLDPE) with low density polyethylene (LDPE) was characterized by two different methods: complete pole figures obtained by wide angle X-rays diffraction (WAXD) and polarized infrared spectroscopy (IR) using the Krishnaswamy approach. The molecular orientation of the blends amorphous phase was also evaluated by polarized IR. The crystallinity of the blown films was determined by WAXD. A good correlation between the X-ray pole figures and the polarized IR results was obtained. At all blends compositions, it was shown that the *a*-axis of the polyethylene orthorhombic cell was preferentially oriented along the machine direction, the orientation degree along this direction increasing with the

increase of the LDPE amount in the blends. The *b*-axis changed its preferential orientation from film thickness in the 100/0 LLDPE/LDPE film to along the transverse direction with increasing LDPE in the blends. The *c*-axis changed its orientation from orthogonal to normal direction in the 100/0 LLDPE/LDPE film to along the film thickness with increasing LDPE in the blends. Polarized IR characterization showed a negligible orientation of the amorphous phase. The amount of crystallinity was dependent on blend composition decreasing with the increase of LDPE content in the blends. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 102: 2760–2767, 2006

Key words: molecular orientation; pole figure; dichroism; LLDPE; LDPE

INTRODUCTION

Blends of linear low density polyethylene (LLDPE) and low density polyethylene (LDPE) in the form of blown films are extensively used in different packaging applications. In general, LLDPE has better thermal and mechanical properties than LDPE due to its linearity, with low amount of short chain branching; however, it is more difficult to process and has worse optical properties. Therefore, blends of LLDPE/LDPE can provide optimal properties that would improve the general performance of these films.¹

It is well known that the final properties of blown films are determined by a complex correlation involving materials properties, processing conditions, structure, crystallization, and morphology development. The molecular orientation of the crystalline and the amorphous phases control the final structure and performance of blown films. Thus, it is important to be able to characterize, qualitatively and quantitatively, the

molecular orientation of blown films, which have biaxial nature due to the processing mode.

Recently, Krishnaswamy² introduced a polarized infrared spectroscopy (IR) method that employs a general form of the Beer–Lambert law to estimate the White/Spruiell biaxial orientation factors, f^B , of the crystalline phase in several LLDPE and high molecular weight high density polyethylene (HMW-HDPE). The Krishnaswamy key assumptions and the relevant equations used to estimate the average biaxial orientation of the crystal unit cell axes with respect to the axes of the polyethylene blown films are described in the literature.³ This method allows the identification of the nature of the preferential orientation of the *a*, *b*, and *c* axes and quantify the degree of the crystalline orientation along the machine (MD) and transverse (TD) directions of all PE blown films. Nevertheless, the calculation of the orientation factor for blends is not common. The calculation of the orientation by wide angle X-rays diffraction (WAXD) pole figures is the more accurate of all the known techniques, being also helpful as a parameter for others techniques. In this method the diffracted intensity of Bragg's planes is measured in the transmission and reflection modes, since neither of the two modes covers all the space by itself. The intensity of the diffracted beam is proportional to the pole density in

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Contract grant sponsor: FAPESP.

the specimen. The pole figure is prepared by the projection of this variation of pole density with pole orientation for a selected crystalline plane. By this method it is possible to get qualitative information about the preferential orientation of the molecule axes and quantitative information of the degree of the crystalline orientation along the MD, TD, and normal (ND) direction of all PE blown films.⁴

In this report, the biaxial molecular orientation of the crystalline phase of blown films of LLDPE/LDPE blends was characterized by WAXD pole figures and polarized IR techniques. The aim of this work was to compare both methods to validate the use of the Krishnaswamy approach in LLDPE/LDPE blends films. The orientation of the blends amorphous phase was evaluated by polarized IR. The crystallinity of the blends films was determined by WAXD.

EXPERIMENTAL

Materials

The LLDPE and the LDPE employed in this work were both kindly supplied by Braskem S.A. from Brazil. The LLDPE number-average (M_n) and weight-average (M_w) molecular weights were 8.500 and 90.000 g/mol, respectively, and its density was 0.920 g/cm³. The LLDPE comonomer was butyl and its melt flow index (MFI) was 1.0 g/10 min at 190°C and 2.16 kg. The M_n and M_w of LDPE were 7.000 and 90.000 g/mol, respectively.³ These materials were dried for 8 h at 70°C before extrusion. Both polymers had no additives.

Blown films processing

The blown films of the LLDPE/LDPE blends at 100/0, 90/10, and 80/20 compositions, in weight percent, were produced in a tubular extrusion line from Carnovali, model CLD-75. The processing conditions were described in details in the first part of this work.³ The blends films studied in this work were produced with a blow-up ratio (ratio of final diameter of bubble to the die diameter) of 3/1.

Index of crystallinity

The index of crystallinity, I_c (WAXD), was measured by WAXD, using a Siemens diffractometer, model D5000, with Fe-filtered CoK α radiation, operating at 33 kV and 50 mA. The I_c (WAXD) of the blends was calculated as the ratio between the total area of the crystalline peaks and the total area of the diffractogram. A separate amount of crystallinity for each polymer could not be calculated because both polymers have the same unit cell parameters, even if recent results⁵ have found an increase of the unit cell

parameters with the increase of the length of the branching. To obtain the crystalline and amorphous areas, peaks deconvolution of the diffractograms was performed using Gaussian functions.

Calculation of the molecular orientation factors

By WAXD pole figures

The biaxial molecular orientation factor (f^B) and the representation of the orientation of the PE blown films crystalline plane were determined by the WAXD pole figure technique.

The X-ray incident beam was collimated by a 0.6 mm vertical diffraction slit coupled with a 0.6 mm horizontal divergent Schultz slit. Pole figures were obtained using a closed Eulerian cradle by rotating the film samples with scanning angles of $0 \leq \phi \leq 360^\circ$. Two experimental techniques were required to measure the intensities at a given Bragg angle: the Schultz reflection method was used to tilt the sample from $0^\circ \leq \chi \leq 65^\circ$, while the Decker transmission method was employed for the remaining χ values up to and including 90° .⁴ An angle step of 5° for ϕ and χ was used during the measurements. Exposition times were adjusted in agreement with intensities. Data collection was done by using the software provided by Siemens. Correction of background, absorption effects, and the connection of both experimental methods were performed by the Polo2003 software before plotting the pole figures.

From the pole figure intensities of a (hkl) crystalline plane it is possible to calculate the average square cosine values of the White/Spruiell biaxial orientation factor (f^B)⁶:

$$f_{j,MD}^B = 2 \langle \cos^2 \phi_{j,MD} \rangle + \langle \cos^2 \phi_{j,TD} \rangle - 1 \quad (1)$$

$$f_{j,TD}^B = 2 \langle \cos^2 \phi_{j,TD} \rangle + \langle \cos^2 \phi_{j,MD} \rangle - 1 \quad (2)$$

where j corresponds to the crystallographic axes ($j = a, b, \text{ or } c$) and $\phi_{j,MD}$ and $\phi_{j,TD}$ are the angles between the j crystallographic axis and the machine and transverse directions, respectively.

For orthorhombic crystal systems, when the orientation behavior of two of the crystallographic axes is determined in a chosen reference direction, the orientation of the third axis can be deduced, for example in the MD direction, because

$$f_{a,MD}^B + f_{b,MD}^B + f_{c,MD}^B = 0 \quad (3)$$

To calculate the crystalline f^B in the MD and TD directions, the experimental pole figures were reconstructed by the Polo2003 software, which is based on spherical harmonics for texture analysis.

By polarized IR spectroscopy

The crystalline and amorphous phase molecular orientation of the LLDPE/LDPE blends blown films were measured in a Fourier transform infrared spectrometer, model Spectrum 1000, equipped with a AgBr wire grid polarizer, both from Perkin-Elmer. All absorption spectra were recorded with the incident IR radiation polarized parallel to and perpendicular to the machine direction and at room temperature. Each spectrum was an average of 128 scans, between 400 and 2200 cm^{-1} with a resolution of 4 cm^{-1} .

The infrared absorption bands used for determination of the molecular orientation of PE were the bands at 719 and 730 cm^{-1} . The polymer chains of PE lie parallel to the *c*-axis, with strong covalent bonds along the chain and weaker van der Waals bonds between the chain segments.⁷ The band at 730 cm^{-1} arises from the crystalline phase and is assigned to "in-phase" CH_2 rocking modes. This band has a transition moment parallel to the crystalline *a*-axis. The band at 719 cm^{-1} has been assigned to "out-of-phase" CH_2 rocking modes and contains components due to both crystalline and amorphous phases. The crystalline component of the absorption band at 719 cm^{-1} has a transition moment parallel to the crystalline *b*-axis. The amorphous phase shows a broad band centered at 722 cm^{-1} and has the transition moment perpendicular to the chains axis.^{2,8}

In this work the characterization of the crystalline biaxial molecular orientation of LLDPE/LDPE blends blown films is based on Krishnaswamy's approach.² In our recent publication³ we demonstrated that the use of the Krishnaswamy's method to characterize the biaxial orientation features of PE blown films is appropriate. Thus, calculation of f^B by this approach will not be reported here.

Nevertheless, the Krishnaswamy method is quite sensitive to the film thickness.² In view of the known variation of orientation with film thickness⁹ and of the experimental evidence that it is very difficult to produce a blown film with constant thickness along the extension,³ the samples for the IR measurements were cut along the transverse section of the blown films, where the variation of the film thickness was very low. The pure LLDPE film had an average thickness of 60 μm and the blends specimens had approximately a thickness of 30 μm .

To apply the Krishnaswamy method to the LLDPE/LDPE blends, two assumptions were made: (i) the LLDPE and LDPE crystals were immiscible and (ii) both polymers have "row nucleated" type morphology.³

The orientation behavior of the amorphous phase was evaluated by the Herman's uniaxial orientation factor (f_{am}) from IR dichroism using the amorphous band at 1368 cm^{-1} . This band has been assigned to the molecular segments involving gauche conformers present only in amorphous phases. Since the transition

moment associated with the 1368 cm^{-1} band is perpendicular to the chains axis, the orientation of the amorphous phase can be determined by¹⁰⁻¹²:

$$f_{\text{am}} = \frac{(\text{DR}_{1368} - 1)}{(\text{DR}_{1368} + 2)} \quad (4)$$

where DR_{1368} is the dichroic ratio of the band at 1368 cm^{-1} , and is defined as the ratio of the absorbances measured with the infrared radiation polarized parallel (A_{\parallel}) and perpendicular (A_{\perp}) to the chosen reference direction, as given by the equation:

$$\text{DR} = \frac{A_{\parallel}}{A_{\perp}} \quad (5)$$

The absorption intensities of all bands were obtained by integrating the area under the absorption bands. The film thickness normalization was done using the area of the PE thickness band at 1976–2107 cm^{-1} .¹³ A minimum of four measurements was made on each blend sample to obtain a significant statistical average of the data.

RESULTS AND DISCUSSION

Amount of crystallinity

Figure 1 shows the X-ray diffractogram of the anisotropic LLDPE films obtained in the angle range $10^{\circ} \leq 2\theta \leq 50^{\circ}$ using a very small interval (0.003°). The diffraction patterns were indexed using the orthorhombic unit cell of PE, with $\alpha = \beta = \gamma = 90^{\circ}$ and lattice parameters $a = 0.741$ nm, $b = 0.495$ nm, and $c = 0.255$ nm.⁷

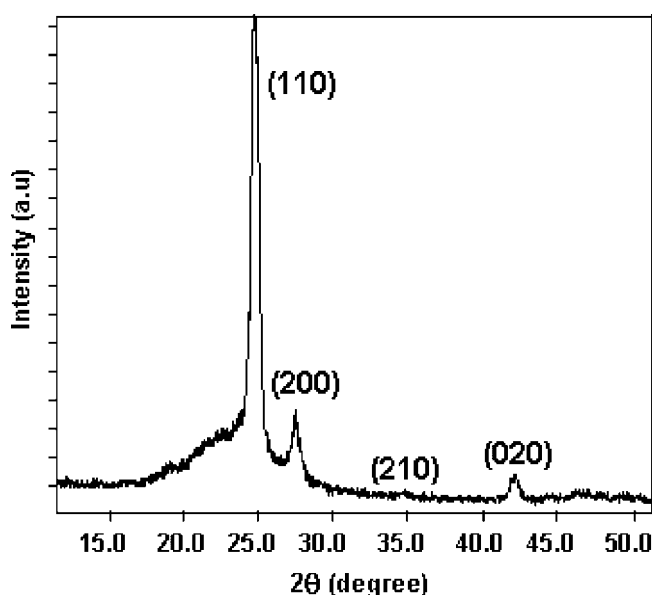


Figure 1 X-ray diffractogram of anisotropic LLDPE film.

TABLE I
Index of Crystallinity, I_c , of the Blends

LLDPE/LDPE	I_c (WAXD)	$A_{(amorp.)}$	$A_{(cryst.)}$
100/0	0.51	790.78	810.22
90/10	0.39	1275.47	805.23
80/20	0.32	1329.01	620.12

I_c , index of crystallinity determined by WAXD; $A_{(amorp.)}$, area of the amorphous peak; and $A_{(cryst.)}$, area of the crystalline peaks.

Two strong peaks at $2\theta = 24.8$ and 27.5° corresponding to the (110) and (200) diffraction planes are observed. In comparison with the (110) reflection, the (020) diffraction plane displays a weak, but visible reflection at $2\theta = 42.3^\circ$, that was suitable for the *b*-axis orientation measurement.

On the basis of these observations, we can conclude that the molecular orientation of the *a*-axis and *b*-axis of the PE crystallites can be characterized by the study of the pole figures of the (200) and (020) diffraction planes. However, because of the very weak (002) reflection at $2\theta = 89.5^\circ$, the *c*-axis pole figure cannot be obtained directly. Instead, it can be indirectly evaluated by making use of the strong (110) reflection. Therefore, the *c*-axis orientation was calculated from the *a*- and *b*-axis orientation by using eq. (3).

The I_c ($WAXD$) of the LLDPE/LDPE blends is summarized in Table I. Figure 2 shows an example of the region of the 90/10 LLDPE/LDPE diffractogram used to obtain the I_c ($WAXD$). In this figure the deconvolution curves of the peaks can be seen. The areas of the crystalline and amorphous peaks resulting from the peaks deconvolution are also listed in Table I.

The results pointed out that the index of crystallinity decreased with the increase of LDPE concentration in

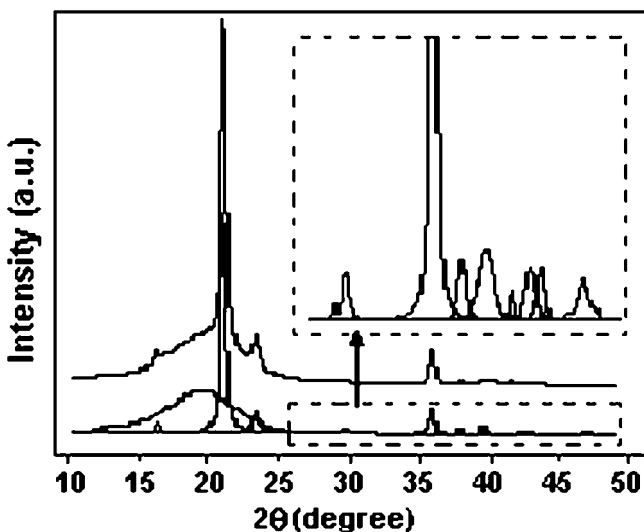


Figure 2 Deconvoluted X-ray diffractogram of 90/10 LLDPE/LDPE film.

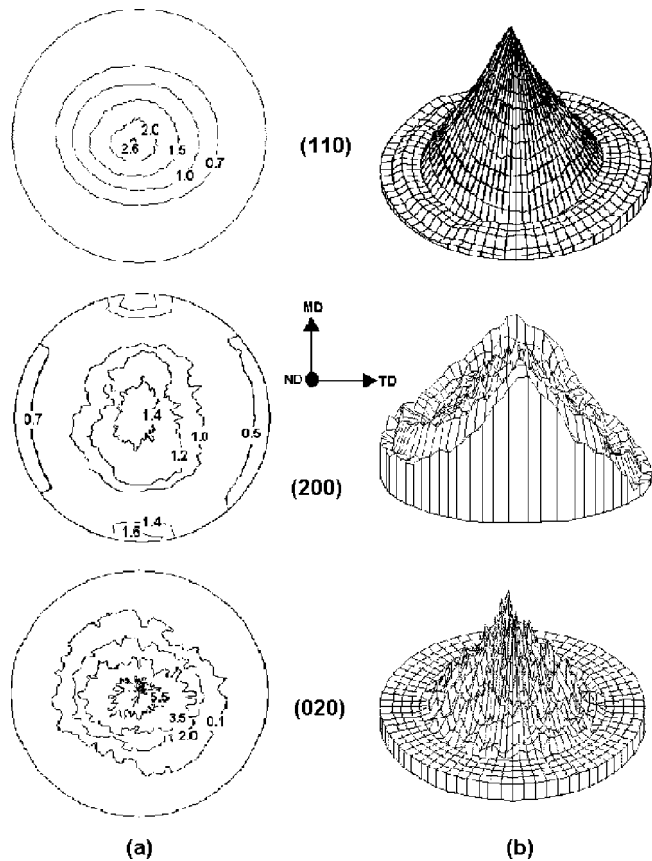


Figure 3 (a) Contour and (b) 3D projections of the X-rays pole figures for the (110), (200), and (020) planes of the 100/0 LLDPE/LDPE film.

the blends. Data from the literature¹⁴ on LLDPE crystallinity amount calculated by DSC are in the range of 22 and 55%, confirming our results.

Molecular orientation by WAXD pole figures

The X-ray pole figures of the (110), (200), and (020) planes for the pure LLDPE film are shown in Figure 3. In this work, the pole figures were plotted in a contour and a three-dimensional (3D) projection with the MD and TD in the plane and normal direction (ND) perpendicular to the MD/TD plane. The maximum pole density was indicated in the isolines of the contour projection. The 3D projection allows a more detailed interpretation in terms of the disposition of the *a*, *b*, and *c* axes with respect to the MD, TD, and ND.

The pure LLDPE blown film has a light texture as seen in Figure 3. By the interpretation of the (110) pole figure it can be concluded that the *c*-axis of the pure LLDPE is perpendicular to the ND since the maximum pole density of the (110) pole figure is in the normal direction. The (200) pole figure shows two maxima. The more pronounced of these maxima is along MD and the less one along ND. This result evidences that the *a*-axis is distributed along the MD/ND plane with the main

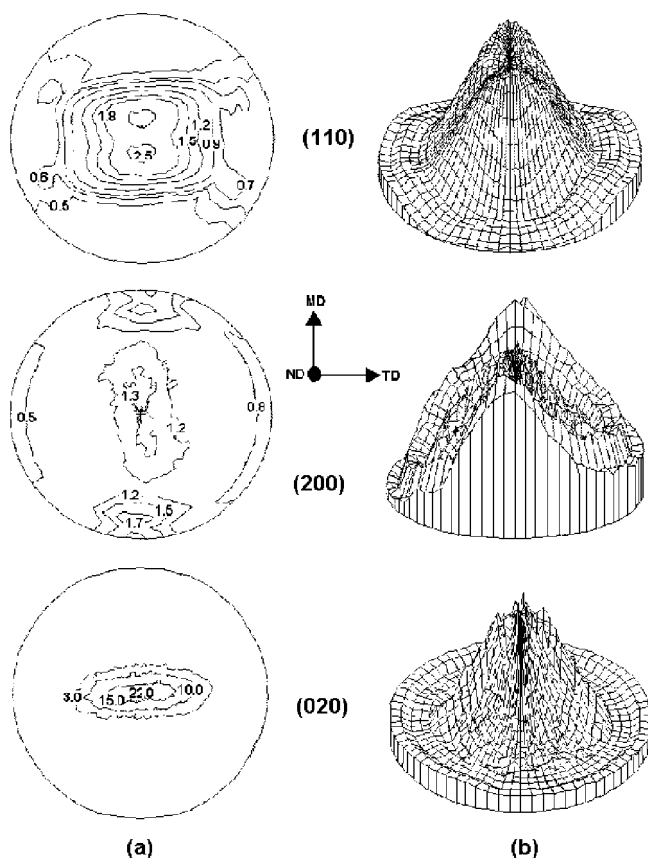


Figure 4 (a) Contour and (b) 3D projections of the X-rays pole figures for the (110), (200) and (020) planes of the 90/10 LLDPE/LDPE film.

maxima disposed about MD. The (020) pole figure for the pure LLDPE film shows a very well defined maximum in ND, indicating that the *b*-axis of the orthorhombic unit cell is preferentially oriented along the thickness of the film.

The X-ray pole figures of the 90/10 and 80/20 LLDPE/LDPE blends films are shown in Figures 4 and 5, respectively.

Regarding the *a*-axis, it can be concluded from the analysis of the pole figures of the (200) plane that this axis is also distributed along the MD/ND plane but preferentially oriented in the MD. The *a*-axis orientation degree along MD is higher when the amount of LDPE in the blends is higher. By looking at the (020) pole figures in Figures 4 and 5, it is evident that a maximum pole density in the ND occurred. The *b*-axis is oriented along the film thickness (ND) in the 100/0 film but oriented in the TD in the 90/10 and 80/20 LLDPE/LDPE films. However, a decrease in the ND and an increase in the TD orientation magnitude with the increase of LDPE in the blends were observed. The (110) pole figures of the blends exhibited two maximum groups. One, in the pole figure center, exhibited a significant amount of (110) planes, with its maxima along MD

forming an angle of 19° and considerably oriented in the TD. The other maximum had its normal in the TD forming an angle of 77° with the MD. These observations indicated orientation of the *c*-axis perpendicular to the MD/TD plane in these films, that is, along the film thickness (ND).

To quantify the molecular orientation of the PE axes (*j*), values for $f_{j,MD}^B$ and $f_{j,TD}^B$ were calculated from the experimental pole figures for the films of the blends and were plot in the form of the White/Spruiell orientation diagram, as shown in Figure 6.⁶ The dashed line represents equibiaxial orientation along the plane of the film. Standard deviations of these values are of the order of ± 0.05 . A comparison between the values reveals again the preferential orientation of the *a*-axis along MD for all blends composition and the change of the *b*- and *c*-axes orientation in the blends films. The *b*-axis changes its preferential orientation from ND in the pure LLDPE to along TD in the blends. The *c*-axis changes its orientation from equibiaxial MD/TD in the pure LLDPE film to along the film thickness in the blends.

After leaving the die extruder, the LLDPE and LDPE molecules will crystallize under the influence of biaxial tensions. The resultant morphology will depend, besides

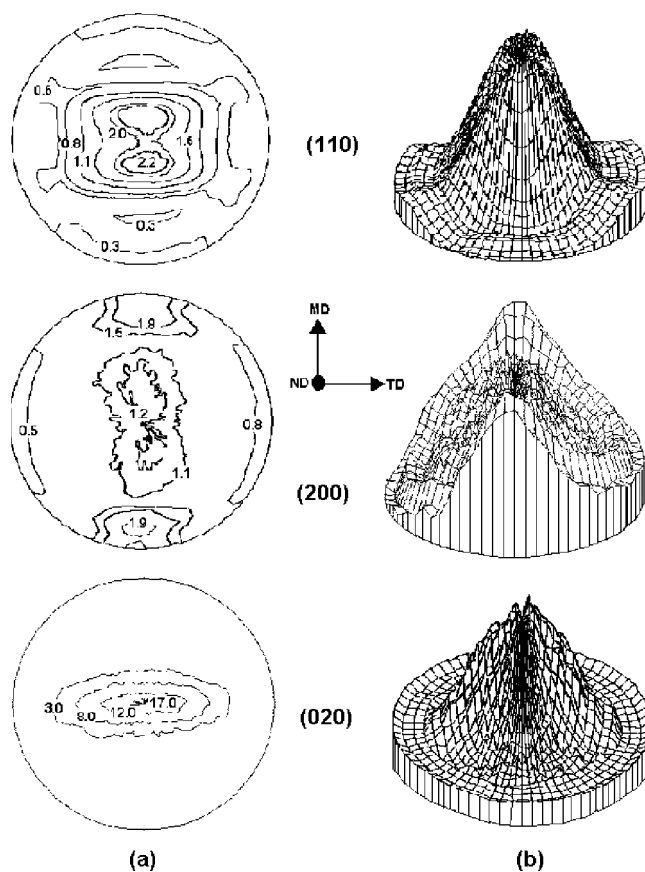


Figure 5 (a) Contour and (b) 3D projections of the X-rays pole figures for the (110), (200) and (020) planes of the 80/20 LLDPE/LDPE film.

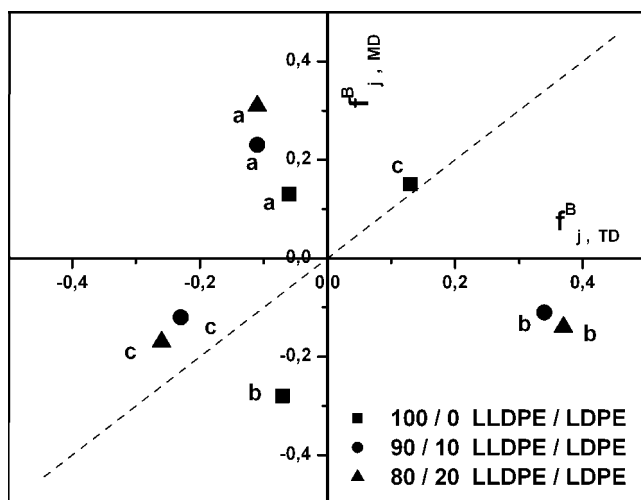


Figure 6 Biallaxial orientation factors of the unit cell axes of the LLDPE/LDPE blends films as calculated by X-ray pole figure; $j = a, b, \text{ or } c$.

M_n , M_w , M_n/M_w , long, and short chain branching distribution, on the elastic recovery during crystallization. Linear molecules will crystallize first than branched ones¹⁴ and, all the structural parameters being similar, linear molecules will have higher relaxation times than branched ones. Therefore, during the blown film operation, LLDPE will crystallize first but will relax later than LDPE.

As a pure material, the LLDPE will form, during blowing, "row nucleated structures,"¹⁴ the so called cylindrites, highly organized, with stacked lamellae of higher dimensions than the LDPE lamellae. Its texture will be the *a*-texture (Keller-Machin I) with the lamellae *a*-axis oriented along MD and the *b*-axis (lamellae growth direction) oriented along TD. On the other hand, as a pure material, the LDPE will form, during blowing, very small spherulites, with many tie molecules and many intra and interspherulitic amorphous phase. The thin lamellae will have the *c*-axis oriented perpendicular to the spherulites radius and the *b*-axis will be parallel to this radius.

We have found¹⁵ that both polymers are miscible in the melt state, up to a 20 wt % LDPE concentration, and after crystallization, they will form two separate crystals populations.³ Thus, we are assuming that during the blends blown film operation, the LLDPE, because it crystallizes first but relax later than the LDPE molecules, will form cylindrites. However, because of the presence of the LDPE, the LLDPE row nuclei will be widely apart and its lamellar growth will be tilted, changing from along ND (pure LLDPE) to along TD (therefore assuming the Keller-Machin I texture). At the same time, the LDPE has already relaxed and it will begin to crystallize later forming very thin lamellae that will follow the same crystal habitat than the LLDPE lamellae. One

question remains unanswered: the LLDPE small lamellae will form separate small spherulites or will stack between the LLDPE lamellae? There was a reduction of the index of crystallinity in the blends; both morphologies can promote this reduction.

Molecular orientation by polarized IR

In this study, the orientation behavior of the crystalline phase in the LLDPE/LDPE blends blown films determined by polarized IR are also described in terms of the White/Spruiell biaxial orientation factors (f^B).^{2,16-18} Details regarding the f^B calculation from the IR absorbencies were documented previously.^{2,3} It must be pointed out that the crystallinity indexes used to calculate f^B were the ones measured by differential scanning calorimetry (DSC).³

A typical parallel (full line) and perpendicular (dashed line) to the machine direction IR absorption spectra for the LLDPE/LDPE blend blown films is shown in Figure 7. The spectra shown in this figure was recorded for the 90/10 LLDPE/LDPE blend.

The calculated f^B values were smaller than 0.370. This is an indicative that the average biaxial crystalline orientation of the films was small. Typical standard deviations of these values are of the order of ± 0.010 . Branciforti et al.³ analyzed the orientation of the same films along the height, and it was observed that the standard deviations of the measurements were very high. The measurement error deviation was attributed to the high variation of the film thickness along the film height.

In Figure 8, the f^B of the crystalline phase (*a*, *b*, and *c* unit cell axes) in the LLDPE/LDPE blends blown films are plotted in the form of the White/

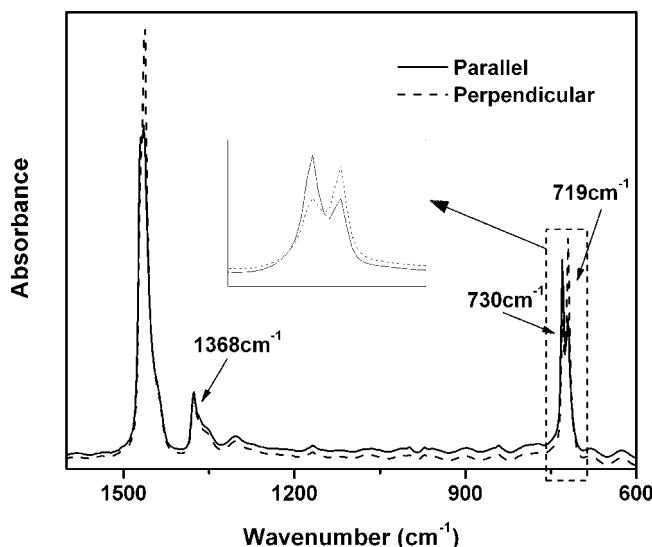


Figure 7 IR spectra of 90/10 LLDPE/LDPE blend using polarized light parallel and perpendicular to the MD.

Spruiell orientation diagram.⁶ This diagram confirms the conclusions drawn from the pole figures.

The graphical representation in Figure 8 clearly shows that the pure LLDPE film display preferential orientation of the *b*-axis in the crystalline phase along ND. The *a*-axis displays orientation between MD and ND planes, but closer to MD. The *c*-axis is not as strongly oriented and displays close to equibiaxial orientation in the plane of the film. In the 90/10 and 80/20 LLDPE/LDPE blends, the *a*-axis has significant orientation along MD. The *c*-axis is preferentially oriented along the film thickness (ND). However, the *b*-axis appears to be located largely in the ND/TD plane with a strong orientation along the TD. The orientation along TD increases with the increase of LDPE content. This behavior suggests that the crystallographic *b*- and *c*-axes were reoriented upon increasing the LDPE amount in the blends. These results are largely consistent with prior orientation studies of crystalline phase in LLDPE and LDPE blown films by WAXD and polarized IR^{2,16,17-26} and are typically interpreted in terms of the Keller-Machin I "row nucleated" structure.^{27,28}

Molecular orientation of the blends amorphous phase

Many reports^{8,11,12,29} have indicated that the amorphous orientation plays an important role on the final properties of the blowing films. In this work, we present an attempt to measure the orientation of the amorphous phase. This is achieved by dichroism IR measurements of the amorphous band at 1368 cm⁻¹ (see Fig. 7). The Herman's orientation factors of the chain segments of the amorphous phase in the blends blown films were estimated by eq. (4).

The values of the dichroic ratios (DR₁₃₆₈) and of the Herman's orientation factors (*f*_{am}) of the amor-

TABLE II
Dichroic Ratios (DR) and Herman's Orientation Factors (*f*_{am}) of the Amorphous Phase in the LLDPE/LDPE Blends as Calculated by Polarized IR and Measured in MD and TD

Blend LLDPE/LDPE	DR _{1368, MD}	<i>f</i> _{am, MD}	DR _{1368, TD}	<i>f</i> _{am, TD}
100/0	1.185	0.058	1.178	0.056
90/10	1.190	0.060	1.190	0.060
80/20	1.187	0.059	1.158	0.050

phous phase calculated from IR spectra measured relative to MD and TD are listed in Table II. The anisotropic effect increases as the DR for the specific transition moment deviates from 1. The measurement error for orientation factors from dichroism in this work was found to be below ± 0.010.

Note that the Herman's molecular orientation factors of the amorphous phase determined by dichroism IR are uniaxial. They only give the average orientation of the amorphous phase with respect to a reference direction. Examination of the data of Table II indicates that the transition moment corresponding to the 1368 cm⁻¹ band is almost randomly distributed in the blends blown films. The *f*_{am} values are very low, showing that there is small orientation of the amorphous phase and it is independent on blend composition.

CONCLUSIONS

A texture analysis of blown films of LLDPE/LDPE blends was carried out. Detailed qualitative and quantitative information relating to the molecular orientation of the crystalline phase was done by using pole figures from WAXD data. Using polarized IR spectroscopy measurements, the orientation of the crystalline and amorphous phases was also obtained. The films crystallinity was calculated by WAXD measurements. The texture of the LLDPE/LDPE blown films can be described as an arrangement of lamellae with their *a*-axis along the MD, their *b*-axis along TD, and *c*-axis along the ND (Keller-Machin I, a-texture). No orientation in the amorphous phase was observed. Regarding the crystallinity of the blends, it was observed that it decreased with the increase of LDPE content. From these results, it can be concluded that the orientation factors obtained by polarized IR using the Krishnaswamy approach have been confirmed by the orientation factors calculated by WAXD pole figures; however, the homogeneity of the film thickness is an important feature for the accuracy of this calculation.

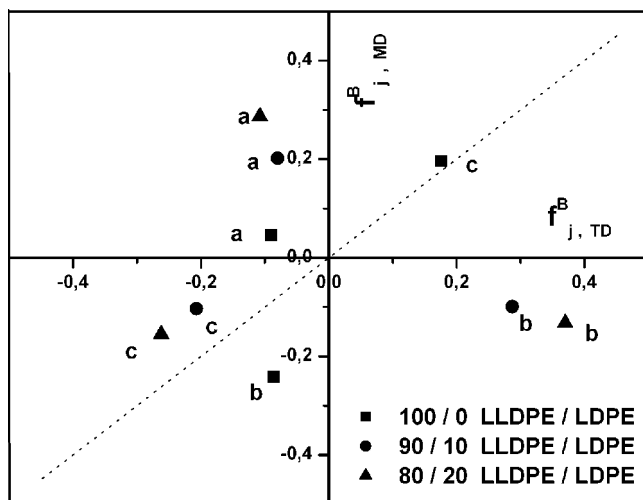


Figure 8 Biaxial orientation factors of the crystal unit cell axes of LLDPE/LDPE blends films plotted in a White/Spruiell diagram as calculated by polarized IR; *j* = *a*, *b*, or *c*.

The authors thank Braskem S.A. of Brazil and R. Pimentel for the donation of the samples.

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