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Mechanical Properties, Stress-Relaxation, and Orientation of Double Bubble Biaxially Oriented Polyethylene Films

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ABSTRACT: Biaxially oriented linear low density polyethylene (LLDPE) films were produced using the double bubble process with different machine direction (MD) orientation levels and the same transverse direction (TD) blow-up ratio. Their mechanical behavior was characterized in terms of the tensile strength and tear resistance. The viscoelastic behavior of oriented films was studied using dynamic-mechanical thermal analysis (DMTA). The microstructure and orientation were characterized using microscopy, X-ray diffraction pole figures, and birefringence. The results indicate that MD ultimate tensile strength increases and the TD one decreases with MD stretching ratio. Tear propagation resistance, in general, remained mainly con-

stant in TD and decreased in MD, as the draw ratio was increased. The morphology analyses exhibit a typical biaxial lamellar structure for all samples with different lamellar dimensions. Orientation of *c*-axis in crystalline phase, molecular chain in amorphous phase along MD increased with draw ratio. In most crystals, *a*-axis was located in the normal direction (ND) and the *b*-axis in the ND-TD plane. A good correlation was observed between *c*-axis orientation factor and MD mechanical properties. © 2006 Wiley Periodicals, Inc. *J Appl Polym Sci* 100: 3545–3553, 2006

Key words: morphology; oriented films; mechanical properties

INTRODUCTION

Polyethylene films are widely used products in packaging industry. These films are extensively used in flexible packaging of a wide spectrum of products. They provide a very good combination of physical and mechanical properties, which meet the demands and technical requirements of the packaging industry. One of the most used packaging techniques is the shrinkage method. In this method, the film wraps the packaged item, producing the bag. This bag is introduced into a heated tunnel. If the film is oriented it shrinks and wraps the item smoothly. There are several processes for the production of these oriented films. One of the best known is biaxial orientation by the double bubble process or tubular orientation process.

In this process, the primary extruded tube is quenched, reheated to a temperature below the melting point, and then oriented in both machine direction (MD) and transverse direction (TD), simultaneously. Although stretching occurs simultaneously in MD and TD, the forces for each are controlled separately. The

MD force is applied by the differential speed between the two sets of nip rolls that hold the tube. TD stress is applied through the air pressure introduced into the bubble.

The advantage of this technique is the balanced properties of the film in both directions.¹ Biaxially oriented films also present exceptional clarity, superior tensile properties, good combination of flexibility and toughness, unique heat-shrink property, and higher gas-barrier properties than unoriented film. Molecular orientation during stretching takes place in the following manner: below their melting point (or glass-transition temperature in the case of amorphous polymers), polymer chains are rigid. At higher temperatures, they become more flexible and are able to unfold as stress is applied. If a mass of randomly coiled and entangled chains is at a temperature high enough when stress is applied, as in biaxial stretching, the polymer chains disentangle, unfold and straighten, and slip past their nearest neighbor.²

The main difference of the double bubble process from the usual blown process is the blowing of the film below the melting point of the polymer. This provides the possibility to achieve films with lower thickness, better strength and optical properties, and increased thermal shrinkage.

The orientation and properties of polyethylene films produced by the blown-film technique have been

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studied in depth.³⁻⁷ There are also some literature data on orientation of blown polypropylene^{8,9} and polylactic acid (PLA) films.¹⁰ Many other papers investigated the morphology of blown polyethylene.^{6,7,11-17} However, only few authors investigated the morphology, orientation, and properties of films produced by double bubble process. In these studies, the orientation has been mostly planar. Orientation and morphology of oriented polyethylene (mostly blown film) has been studied in depth as well.¹⁸⁻²³

The stress-relaxation and creep behavior in a number of oriented structures have also been studied. For example, mechanical relaxation in uniaxially oriented linear low density polyethylene (LLDPE) was studied by Chong et al.²⁴ Chow and Van Laeken²⁵ and Hawthorne²⁶ explained stress relaxation in oriented poly(ethylene terephthalate) (PET) films. These studies dealt with relatively thick films (76 and 33 μm , respectively), which have been oriented above the glass transition temperature. Shinozaki and Sargent²⁷ examined the relaxation behavior of uniaxially hot-drawn polypropylene. They investigated the effect of the relaxation temperature. DTMA (dynamic-mechanical thermal analysis) study of polyethylene naphthalate (PEN) film relaxation was carried out by Gillmor and Greener²⁸ as well.

Stress-relaxation behavior of simultaneously oriented LLDPE films was studied from the viewpoint of relaxation time spectrum.²⁹⁻³¹ The effects of different layers' structure and crosslinking on relaxation time spectrum were presented.

Nevertheless, the morphology of films produced by simultaneous biaxial orientation during double bubble process has not been studied thoroughly. There is a lack of information on the relationship between the orientation level and properties of biaxially oriented shrink films. Hence, the main goal of the present investigation is to find correlations between orientation, morphology, and properties of biaxially oriented LLDPE films produced using different orientation ratios.

EXPERIMENTAL

LLDPE monolayer films with density 0.920 g/cm³ and thickness of 15 μm were tested. All the films were produced at an industrial double bubble line (Prandi) with a constant orientation ratio (5.2) in the transverse direction (TD) and different orientation ratios in machine direction (MD). The orientation temperature was 109°C. Unstretched primary extruded tube was taken as a reference sample.

Mechanical properties of the films were tested in both MD and TD using LRX tensile tester (Lloyds) according to ASTM-D882 method. The tear propagation resistance of the film was measured by Elmendorf tear tester using Elmendorf standard.

Dynamic-mechanical tests were conducted using a dynamic mechanical and thermal analyzer (DMTA, model MKII, by Polymer Laboratories Ltd, Loughborough, UK). The samples were tested at a frequency of 1 Hz, and at temperature range from -50 to 130°C, at a heating rate of 0.5°C/min, under a constant tensile load of 0.2 N for all samples. The gauge length of the specimen was 20 mm and the width 5 mm. Samples for DMTA tests have been tested in the tension mode longitudinally (along the extrusion direction).

For orientation determination, birefringence and wide angle X-ray diffraction (WAXD) pole figures were used. The birefringence in the MD-ND and TD-ND planes was measured by an incident multi-wavelength double beam and photodiode array assembly, combined with an in-house developed software. Details of the measurements technique can be found in a previous publication.⁶ These measurements allowed the determination of the average total orientation, using an intrinsic birefringence $\Delta^\circ = 0.058$ for polyethylene. Fourier Transform Infra Red spectroscopy (FTIR) was used as well for orientation study.

For crystalline axes orientations, pole figures of (110) and (200) reflections were obtained using a Bruker AXS X-rays goniometer equipped with a Hi-STAR two-dimensional area detector. The generator was set up at 40 kV and 40 mA and the Cu K α radiation ($\lambda = 1.542 \text{ \AA}$) was selected using a graphite crystal monochromator. Sample to detector distance was fixed at 8 cm. Prior to measurement, careful sample preparation was required to get the maximum diffracted intensity. This preparation consisted of stacking several layers (about 180 layers) to obtain the optimum total thickness. An order of magnitude of this thickness is obtained by taking the reciprocal of the linear X-ray absorption coefficient, μ , of the material of interest. As μ is generally between 3.5 and 3.9 cm⁻¹ for polyethylene, this leads to sample thickness of about 2.5-3.0 mm.³²

Hermans orientation functions were derived from (110) and (200) pole figures using the Bruker analytical system software. The (200) Hermans orientation functions give directly the orientation of *a*-axis. By contrast, *b*-axis orientation functions were calculated following Desper et al.'s method.³³ Finally, the *c*-axis orientation function was derived from the *a*- and *b*-axes using the orthogonality condition of the orthorhombic crystallographic structure.

The biaxial orientation factors defined by White and Spruiell, f_M^B and f_T^B measure the level of molecular orientation corresponding to machine and transverse directions, respectively. Those factors for an amorphous polymer (or the average for a crystalline polymer) can be obtained from birefringence measurements as:³⁴

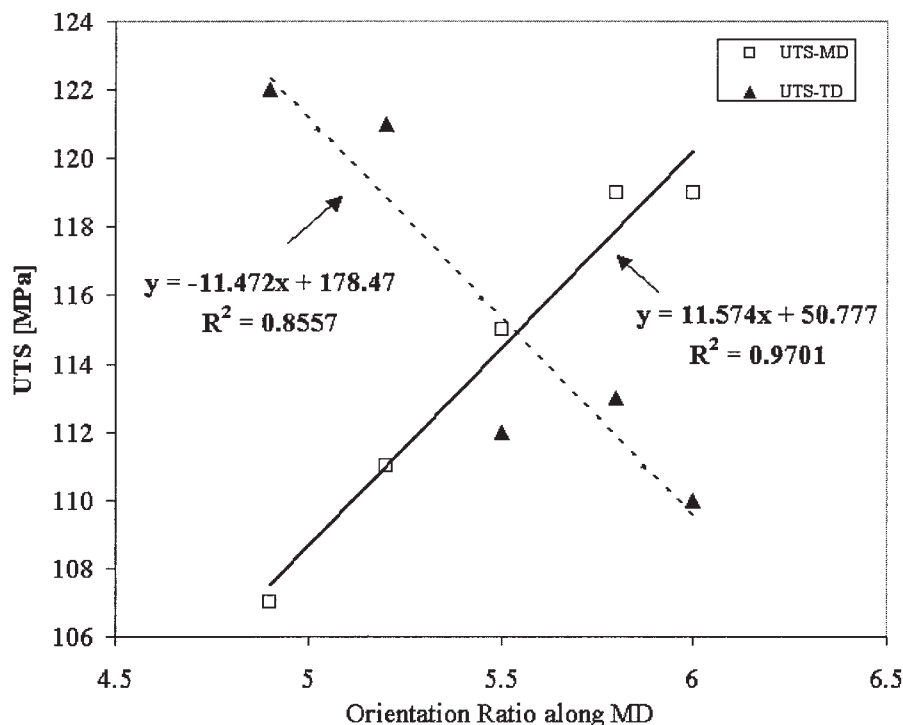


Figure 1 Ultimate tensile stress as a function of orientation ratio.

$$f_M^B = \frac{\Delta n_{MN}}{\Delta^\circ} \quad \text{and} \quad f_T^B = \frac{\Delta n_{TN}}{\Delta^\circ} \quad (1)$$

the Herman's orientation factors defined as:

$$f_{jM} = \frac{3\langle \cos^2 \theta_{jM} \rangle - 1}{2} \quad (2)$$

for an axis j with respect to a direction M are used in this paper.

The lamellar morphology was observed on samples etched for 20 min. using a Hitachi field emission scanning electron microscope.

RESULTS AND DISCUSSION

Figure 1 shows the ultimate tensile stress (UTS) versus MD orientation ratio. The UTS in MD increases linearly as the draw ratio increases while the UTS in TD decreases as the draw ratio increases. The increase of the stress in MD can be explained by increase of orientation with orientation ratio. So one can assume that the decreasing of the stress in TD occurs because of the decreasing of the TD-orientation. Therefore, based on the tensile stress results, we can assume that the increasing of the orientation ratio in the MD causes the increasing of the orientation in this direction accompanied by the decreasing of orientation in TD. The results presented in Figures 2 and 3 approved the assumption of the orientation in MD. While both,

elongation and tear propagation results do not show any significant tendency of the mechanical properties in TD, both tests show the linear dependence of elongation (Fig. 2) and tear propagation resistance (Fig. 3) on orientation ratio.

Figure 4 (a–c) depicts the results of DMTA experiments. We start the presentation of the data from -60°C . T_g or γ -relaxation that takes place usually at the temperatures much below and that is the characteristic of the main amorphous phase of polyethylene was not observed neither by DMTA nor by DSC (differential scanning calorimetry) method. This can be explained by the very low mobility of the oriented molecules in amorphous phase.

Figure 4(a) presents the difference of the β -relaxation on orientation ratio. The clear peak of this relaxation in the unstretched sample changes its form and widens when the film is stretched. We assume that β -relaxation is the characteristic of the amorphous phase that is close to the crystalline and that is why the temperature of β -relaxation is higher than the temperature of γ -relaxation or T_g . So we can propose that the ordering occurring in the amorphous phase during orientation bring about to the lower mobility of some molecule fractions. That causes the widening of the relaxation peak. This orientation can be also the reason of $\tan \delta$ peak widening in the samples stretched with 1 : 5.2 and 1 : 5.5 orientation ratios. When the material is stretched to the ratio of 1:6 most of the molecules are aligned and the order increases. That causes the nar-

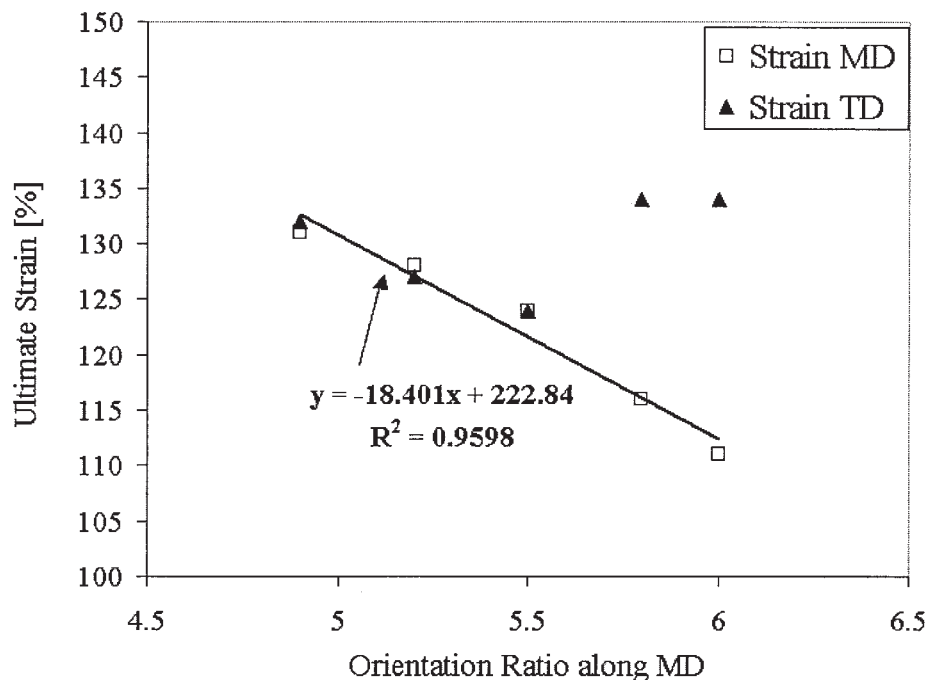


Figure 2 Ultimate strain as a function of orientation ratio.

row relaxation peak that can be detected at higher temperatures than the β -relaxation peak at the unstretched LLDPE. The dependence of the storage modulus on orientation ratio is presented in Figure 4(b). One can see that increase of the orientation ratio

causes the increase of the storage modulus that can be detected at the temperatures below 0°C. When the temperature increases, the differences of the storage modulus of the films stretched at different orientation ratios are less significant. The assumption made above

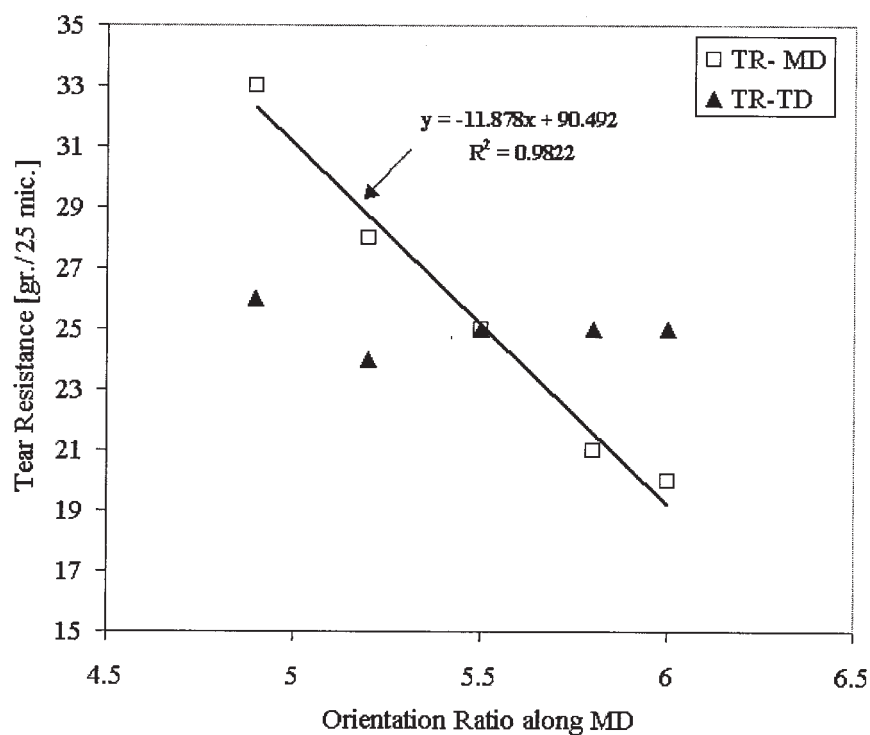


Figure 3 Tear propagation resistance as a function of orientation ratio.

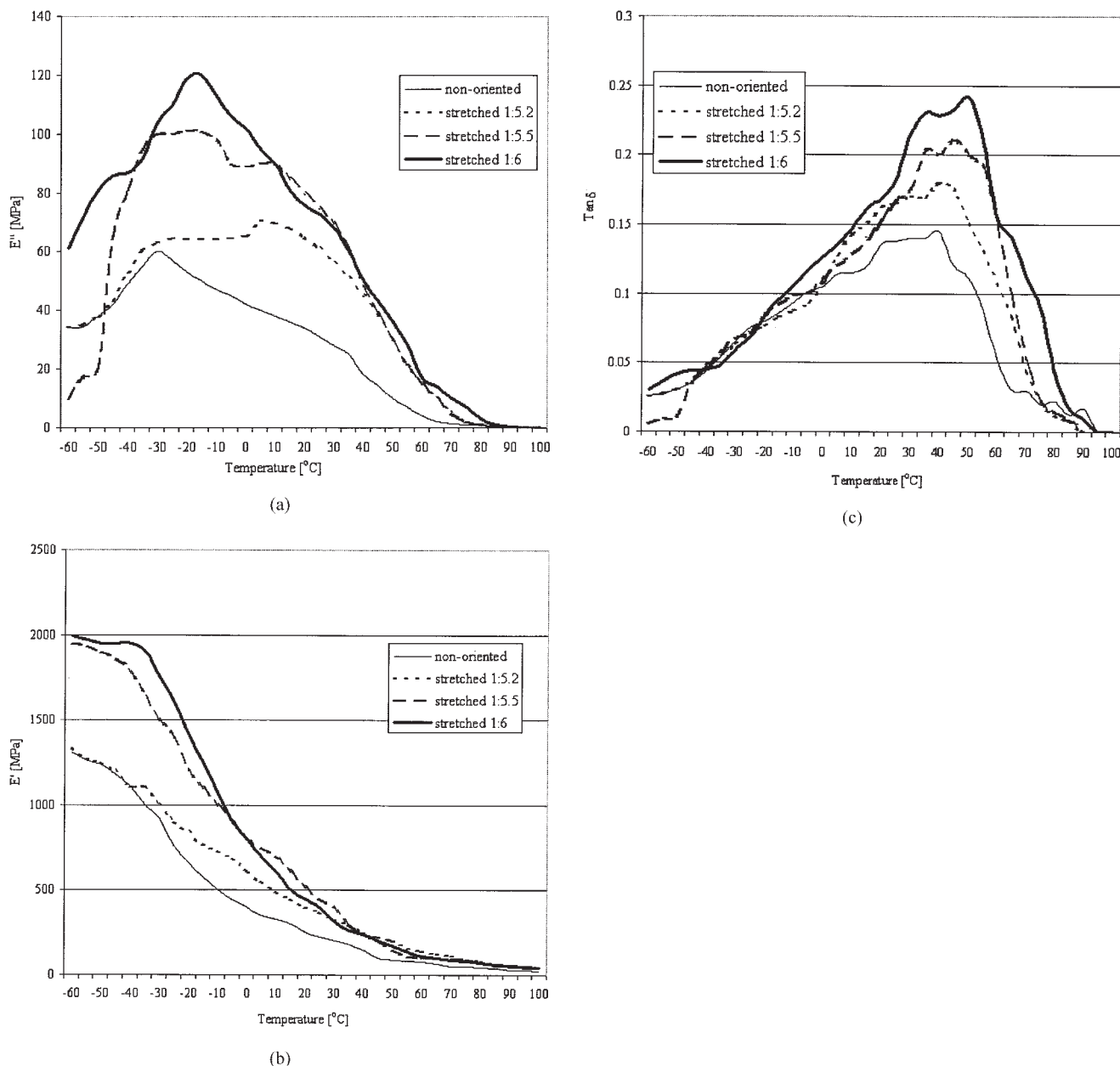


Figure 4 (a) β -transition (E'') in LLDPE film as a function of orientation ratio, (b) Storage modulus (E') as a function of orientation ratio, and (c) $\tan \delta$ in LLDPE film as a function of orientation ratio.

about the lower mobility of the oriented molecules can be approved by $\tan \delta$ [Fig. 4(c)]. One can see the slight increase of $\tan \delta$ temperature. This phenomenon can be definitely caused by limited molecules mobility.

Morphology and orientation were probed using SEM, WAXD pole figures, and birefringence. Figure 5 shows micrographs obtained on selected etched films. The initial morphology (unstretched film) had a random lamellar structure typical of blown LLDPE films. As draw ratio increases, the lamellae thicken and get more aligned. As mentioned in the experimental section, the blow-up ratio (TD) was about 5.2. For the sample drawn in the MD to 5.5, the lamellae are

observed to thicken more and oriented at about the diagonal (45°), because the orientation ratios in machine and transverse directions are close to balanced orientation. For a film drawn to a draw ratio of 6 in MD, the lamellae are even thicker, longer, and oriented perpendicular to MD, which is also typical for a polyethylene oriented in MD as reported in other cases.^{6,7,19} On the other hand, note that TEM observation made in previous study²⁹ showed decreasing of lamellar thickness after biaxial stretching by double bubble process. This phenomenon was accompanied by the decrease of the polymer melting temperature observed by DSC. Therefore one can conclude about

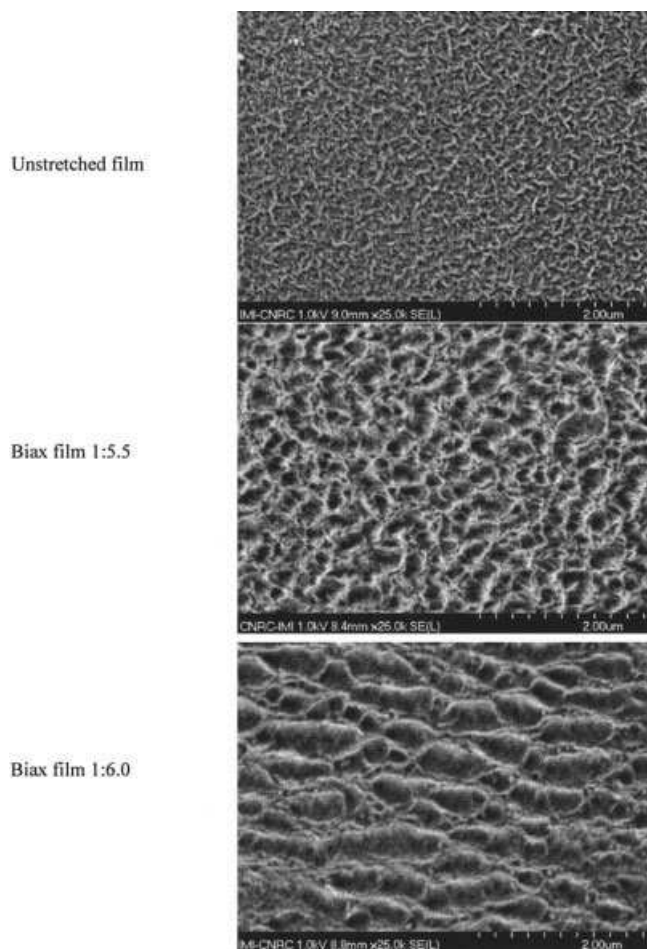


Figure 5 SEM micrographs of etched films with orientation ratios as indicated (MD direction is vertical).

the presence of a highly oriented amorphous phase that cannot be etched because the inability of the oriented molecules. Here we observe the highly oriented unetched amorphous phase around the lamellae structure.

Figure 6 illustrates the WAXD pole figures obtained for the (110) and (200) crystalline reflection of the unstretched and stretched films. One can see that with the increase of orientation ratio the (200) reflection (*a*-axis) is oriented basically towards the normal direction. The (110) reflection, which contains contributions from both, *a*- and *b*-axes, is mostly in the normal and transverse directions. The orientation functions of all the crystalline axes for all the films were determined from their pole figures and are presented at Figures 7 and 8.

In Figure 7(a), the *c*-axis (chain axis) crystalline orientation function in the machine and transverse directions are presented, as a function of MD draw ratio. As can be expected, as orientation proceeds in the MD, the *c*-axis orientation function increases in MD and decreases in TD. The global (average) orientation function, determined from birefringence, in MD and TD as

a function of MD draw ratio is presented on Figure 7(a). The global TD orientation decreases slightly except for the highest draw ratio, whereas the MD one is initially almost constant, but increases for the highest draw ratios. This behavior is not clear yet and further elucidation using FTIR spectroscopy is underway.

The amorphous phase orientation was determined from a combination of the orientation functions of the crystalline phase and overall using a crystalline content of 32% and neglecting crystalline *a*- and *b*-axes and form contributions. A value of 0.058 was taken for the intrinsic birefringence of polyethylene. The results are presented on Figure 7(b) for MD and TD orientation functions. It was observed that the TD orientation factor initially decreased, and then increased for the highest draw ratio, as already seen above for the global orientation. For MD, a similar tendency was observed as well. The orientation of the amorphous phase is thought to come essentially from the tie molecules in the area between the pure crystalline phase and disordered amorphous phase. Since lamellae are rearranging as a result of drawing, a clear trend in the evolution of the orientation factors is not obvious, until very high orientation levels are achieved (fibrillar morphology). A comparison with FTIR results will also be performed to better assess these results.

The orientation functions obtained for the *a*- and *b*-axes are presented in Figure 8. For the *a*-axis [Fig. 8(b)], which is in the normal direction as shown by the pole figure example discussed above, both MD and TD orientation factors are negative, which verify the observations. This orientation in the normal direction increases with MD draw ratio. For the *b*-axis, the MD orientation factor is negative and the TD one is positive. Taking into account the fact that (110) pole shows increasing orientation in ND–TD plane and (200) pole shows increased *a*-axis orientation in ND with the increasing of draw ratio, one can conclude that the *b*-axis is closely located to the TD–ND plane. These observations are in agreement with the observed lamellar morphology discussed above.

Finally, in an attempt to correlate measured properties with structural features, tensile strength in MD and TD as well as MD tear resistance are plotted as a function of different orientation factors in Figures 9 and 10 respectively. For tensile strength, presented in Figure 9, a good correlation is observed with the orientation of the *c*-crystalline axis for the MD direction. With the increase of chain axis orientation in MD, one expects the tensile strength of the film to increase, which indeed is observed. For TD, the correlation is not as good, but fairly acceptable. Apparently it does not coincide with the MD results because of the non-equibaxial orientation nature of the films. These results will also be compared with FTIR spectra for verification.

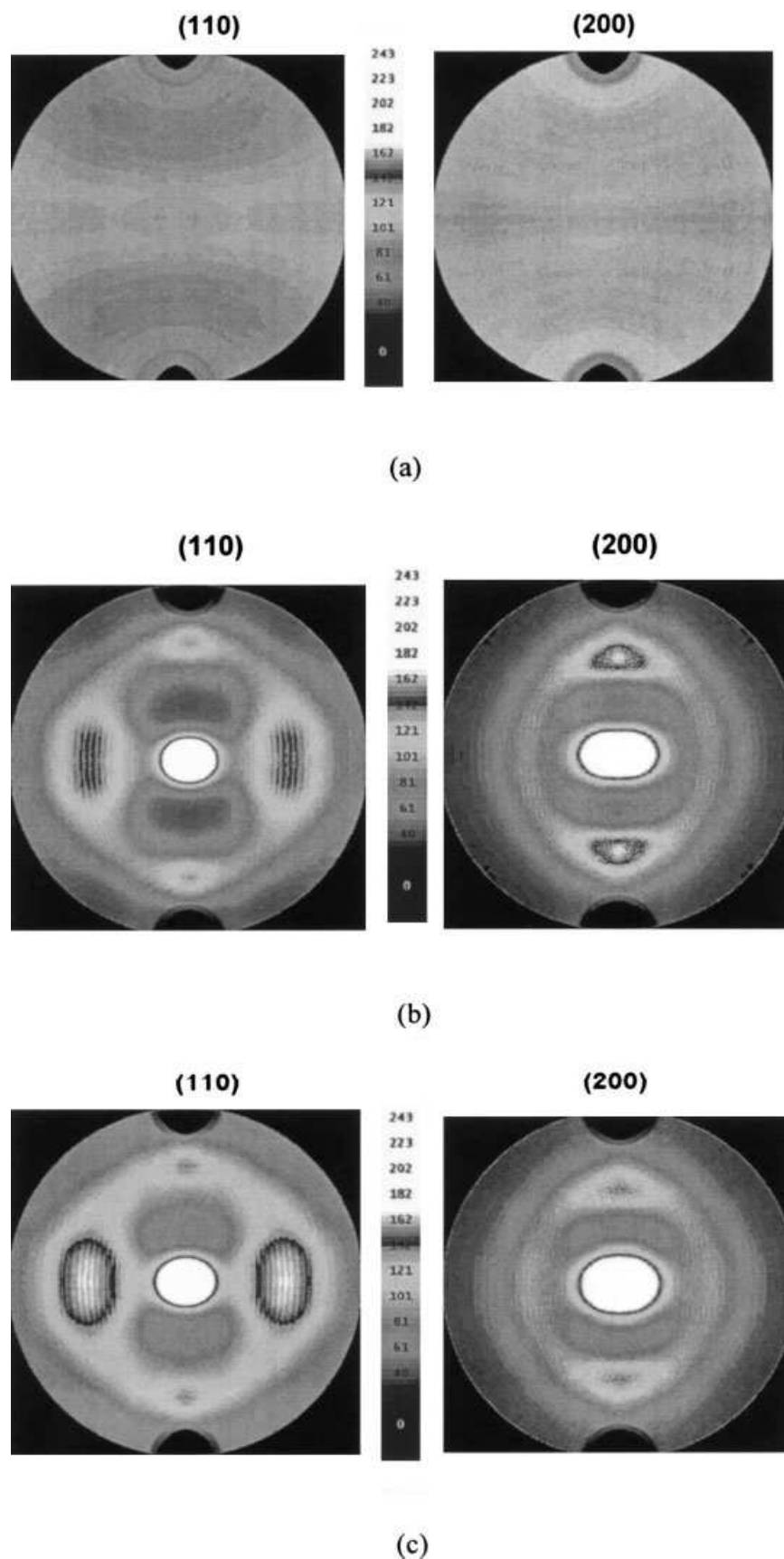


Figure 6 Typical pole figures obtained for the (110) and (200) poles for the: (a) Unoriented polymer (b) film with an orientation ratio of 1 : 5.5 (MD is vertical and TD horizontal) (c) film with an orientation ratio of 1 : 5.8 (MD is vertical and TD horizontal).

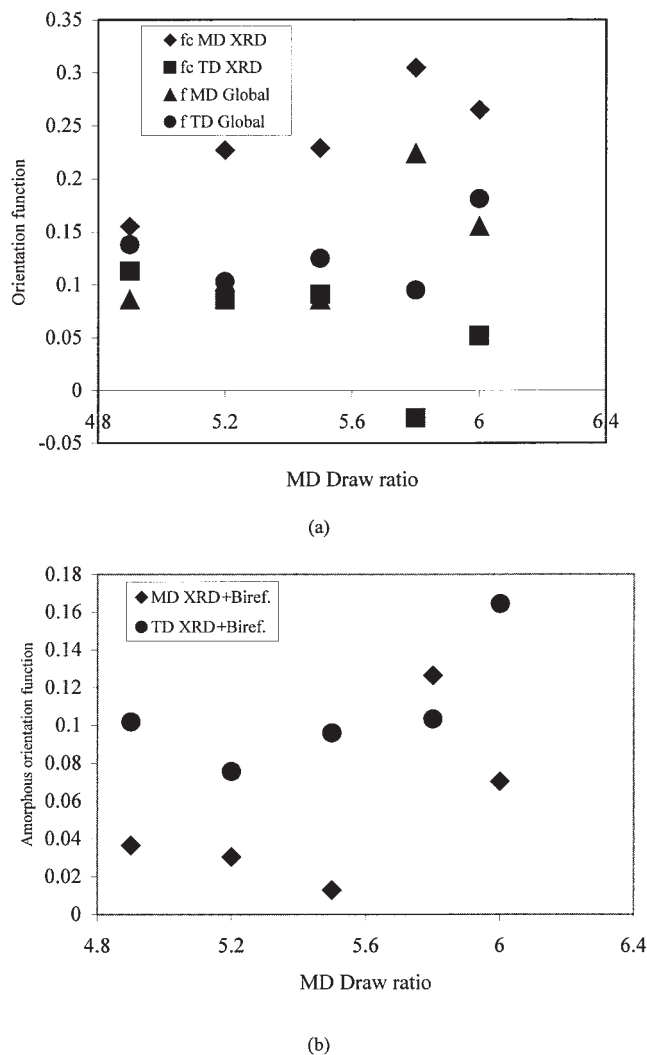


Figure 7 MD and TD orientation functions from X-ray pole figures and birefringence for (a) *c*-crystalline axis and average, and (b) amorphous phase.

For tear resistance, only the property in MD was considered because the TD one did not change significantly with MD draw ratio. The correlation between MD tear resistance and different MD orientation factors is presented in Figure 10. A very good correlation is also observed with the crystalline *c*-axis orientation in MD, and an acceptable one is seen with the global and amorphous orientations as well. This basically indicates that as the orientation in the MD increases (crystalline and amorphous), MD tear resistance decreases almost in a linear relationship. As mentioned above, increasing MD draw ratio rearranges lamellae perpendicular to MD, a structure that gets closer to that of an HDPE with a row structure, for which a low MD tear resistance was also observed,⁷ an effect that becomes more and more prominent with draw ratio.

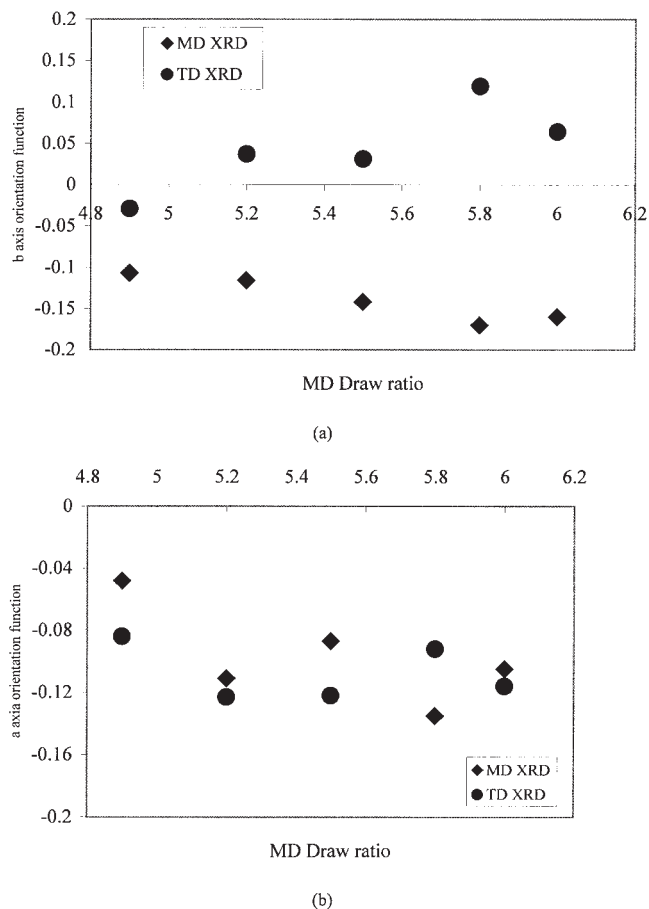


Figure 8 MD and TD orientation functions from X-ray pole figures for (a) *b*-axis and (b) *a*-axis.

CONCLUSIONS

Biaxially oriented films produced by the double bubble technique, with different MD orientation levels, showed that MD ultimate strength increases with MD stretch ratio and the TD one decreases. Tear resistance remained mainly constant in TD and decreased in MD

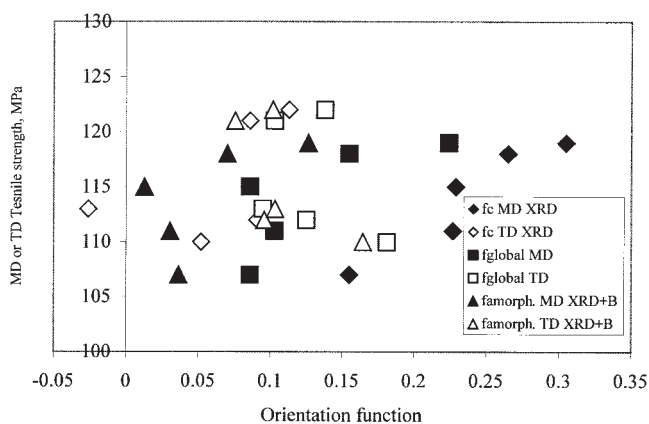


Figure 9 Correlation of tensile strength with different orientation functions.

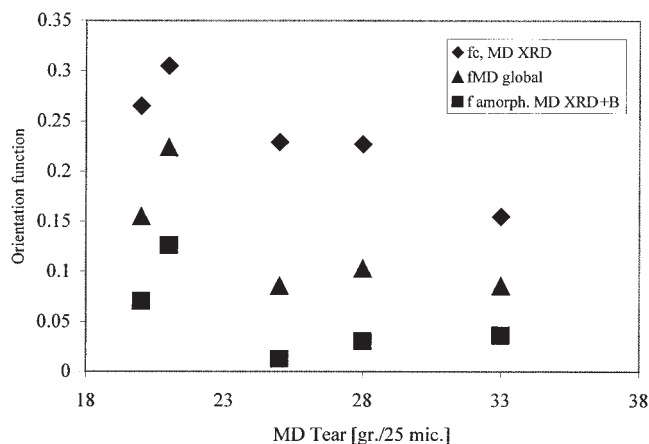


Figure 10 Correlation of tear resistance with different orientation functions.

with draw ratio because of the morphology changes that showed a typical biaxial lamellar structure with increasing orthogonality to MD with MD draw ratio. Orientation in MD increased with draw ratio for c -axis, the amorphous phase and the average, a -axis was located in the normal direction and the b -axis in the ND–TD plane. A good correlation was observed between c -axis and average orientation on one hand and MD tear resistance and tensile strength on the other hand. Good correlation between c -axis orientation and mechanical properties has been found. The DMTA data obtained suggested that increasing of orientation ratio leads to the decreasing of the molecular mobility.

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