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BIAXIAL ORIENTATION OF MULTILAYER LDPE/PET FILMS: STRUCTURE AND PROPERTIES.

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Abstract

In this study, initially thick low density polyethylene / polyethylene terephthalate (LDPE/PET) multilayer blown films were biaxially stretched at different temperatures. The effect of draw temperature and draw speed as well as adhesion between the LDPE and PET layers, through a middle tie layer, on orientation and some properties is studied. The properties of interest are tensile properties, tear, impact strength and haze. The results indicate that the tie layer enhances the properties of stretched films and that toughness is strongly affected by draw ratio and little by draw speed and temperature in the ranges studied.

Introduction

The production of oriented flat films and sheets from thermoplastic materials represents a large segment of the polymer industry. In fact, orientation of polymers enhances many of their properties, particularly mechanical, impact, barrier and optical. Biaxial orientation has the added advantage of allowing this enhancement in two directions. Biaxial orientation is particularly important in films, where it allows to produce thinner films having superior mechanical, optical and barrier properties, and if required, the ability to shrink when reheated. The most widely used biaxial orientation processes for films are the standard film blowing process (such as for PE), tubular double-bubble film blowing (such as for PP) and cast film biaxial orientation or tentering (PP, PS, PET etc.). The latter process is used for many applications, particularly in packaging. High levels of orientation in semi-crystalline polymers are however known to somewhat lead to deterioration of toughness and impact properties, which is particularly the case for biaxially oriented PET at relatively high orientation levels. One of the solutions to overcome this shortfall is through multilayer films, by sandwiching the PET layer using a tougher polymer such as polyethylene.

Experimental

Extrusion grade PET was obtained from Dupont (Selar PT 7086), its molecular weight determined by GPC were Mn = 28,800 and Mw=54,600. Novapol LF-Y819-A LDPE film resin with density of 0.92 g/cc and melt flow index of 0.75 g/10min obtained from Nova Chemicals was used. Ethylene-co-glycidyl methacrylate (EGMA) random copolymer (Lotader AX8840) with 8 wt% GMA contents and melt index of 5g/10min obtained from ElfAtochem was used as a tie layer to enhance the interfacial adhesion between PET and LDPE. Thick multilayer films were prepared by extrusion blowing process and the conditions used are presented in Table-1. More details on films preparations, structure and properties were published elsewhere (1-2). The PET layer was completely amorphous in these initial films.

Table-1: Multilayer blown films process conditions

Sample	BUR	TUR	Thick.	FLH
Ref	2.5	2.7	157	70 ± 10 cm
Tie	2.6	2.7	157	70 ± 10 cm

The thick films were stretched in a biaxial stretching machine (Karo IV biaxial stretcher from Bruckner company) under different conditions. Draw temperatures were 90, 100 and 110 C, draw rates from 1 to 30 m/min and draw mode simultaneous. Draw ratios from 1 to 3.5 in both directions were performed. The initial samples before stretching were 10 cm squares.

Both equibiax and non-equibiax samples were prepared. Samples for both structure and performance characterization were cut from the stretched films.

The crystalline morphology of the films was observed on samples without etching using a Field emission scanning electron microscope (FE-SEM). The films without a tie layer could be delaminated and observed separately.

Orientation of the films, for both LDPE and PET layers, was characterized using FTIR spectroscopy. The measurements were carried out on a Nicolet 170SX FTIR at a resolution of 2 cm^{-1} with an accumulation of 128 scans. Polarization of the beam was performed by using a zinc selenide wire grid polarizer from Spectra-Tech. Tilted configuration was used in order to determine the biaxial orientation factors. More details about this method were reported in previous studies [1-3].

A standard test for tear resistance of plastic film based on ASTM D1922 was used for MD and TD tear resistance. The tensile tests were performed according to ASTM D 882--97, a standard test method for tensile properties of thin plastic sheeting. A crosshead speed of 50 mm/min and a 0.1 kN cell with rubber clamps were used. A video extensometer with 50 mm specimen gauge length and 50 mm grip separation distance was employed. Haze also was measured for most of the films as a function of stretching conditions.

Results & Discussions

Biaxial Stretching of LLDPE/PET Films:

The effect of stretching conditions on the true stress-strain curves is presented on figures 1 (a and b) and 2 (a and b). In Figure 1a, the results obtained for the transverse direction (TD) true stress as a function of strain, on films without a tie layer, drawn at 90 C and 10 m/min to different draw ratios are presented. It is clearly seen that the curves superimpose within experimental error. Figure 1b shows the same results but for films with tie layer. The same observation as above can be made, but the general levels of stresses are significantly higher than for films without a tie layer. This is an indication that the tie layer improves the adhesion between the LDPE and PET phases and that an additional stress is needed to deform the interface created between the two. No strain hardening is observed up to a draw ratio of 2x2, indicating that strain induced crystallization in PET occurs at higher draw ratios.

Figures 2a shows the effect of draw temperature on the true stress-strain curves. Strain hardening is observed for all temperatures, but is more significant at low drawing temperatures, which is expected. Finally, Figure 2b shows the effect of drawing speed for the same temperature and draw ratio. Strain hardening is observed to occur a little earlier for higher speeds. This can be due to the compensating effects between draw rate and stress relaxation: less relaxation occurs at high speeds, which favours earlier crystallization.

Morphology and Orientation:

The initial morphology of the LDPE outside layers was observed using FE-SEM and is shown in Figure 3a and b for films without tie and with tie layer respectively. It consists in a lamellar crystalline structure as observed by others (4-5), which is subsequently rearranged upon drawing as clearly seen on Figures 3c and d. For the PET layer, it was initially amorphous, and upon drawing, a draw ratio of 1.5x1.5 shows no crystalline lamellae as observed in Fig. 3e. For a draw ratio of 3.5x3.5, clear crystalline lamellae are observed in the PET layer, as illustrated in Fig. 3f. Differential scanning calorimetry results (not shown here) also confirmed that the PET layer draw to 3.5x3.5 was crystalline, whereas those drawn up to 2x2 were not.

Orientation of the LDPE and PET layers was determined using FTIR spectroscopy. The results are shown on Figures 4a and b. The orientation of PET trans conformers increased steadily with draw ratio as well as the fraction of trans conformers. The gauche conformer had a negligible orientation. For the LDPE layer, crystalline c axis oriented towards the machine and transverse directions. The orientation of both the PET and LDPE layers was not perfectly equibiax because of some residual initial orientation and a shift in the biaxial stretching machine between the M and T directions (a little more M orientation than T).

Performance:

The performance of the films was determined in terms of tensile mechanical properties, tear strength and haze. The results on tensile strength and elongation at break are shown on figures 5a and 5b respectively. It is clearly seen that strength increases and the elongation at break decreases with draw ratio as expected. No significant differences can be observed between the films with and without a tie layer. The area under the stress-strain tensile curve was also evaluated and called toughness here and is shown on figure 5c. It also decreases with draw ratio, but to a less extent that the elongation at break, which is due to the increase in tensile strength. The speed also seem to decrease somewhat this toughness for constant draw ratio (Figure 6), which may be due as discussed above to higher orientation for higher speeds.

Finally, tear strength and Haze seem to be affected significantly by draw ratio as illustrated on figure 7 and 8.

Conclusions

This study allow us to conclude the following:

- Performance and structure (crystalline morphology and orientation) are strongly dependent on process parameters.
- A tie layer increases the stress levels needed for deformation, but don't affect significantly the final properties
- Toughness is strongly affected by draw ratio and little by draw speed and temperature.

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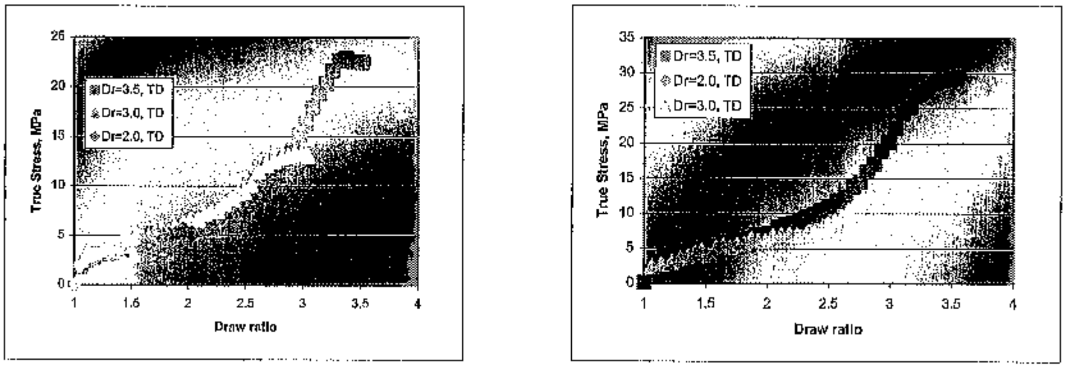
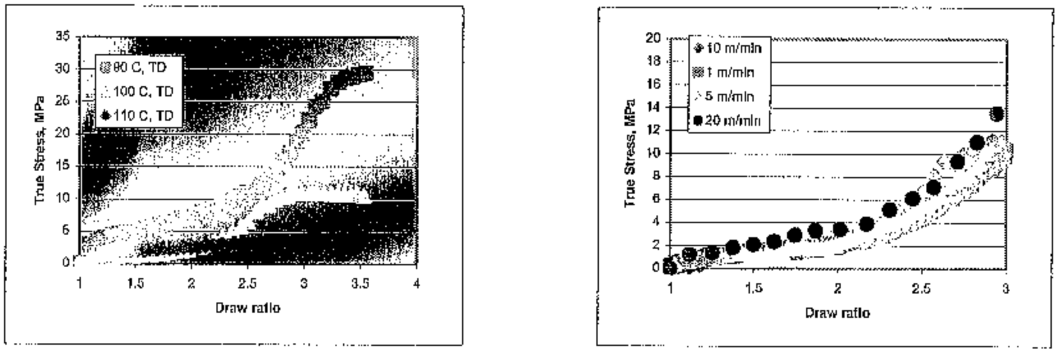


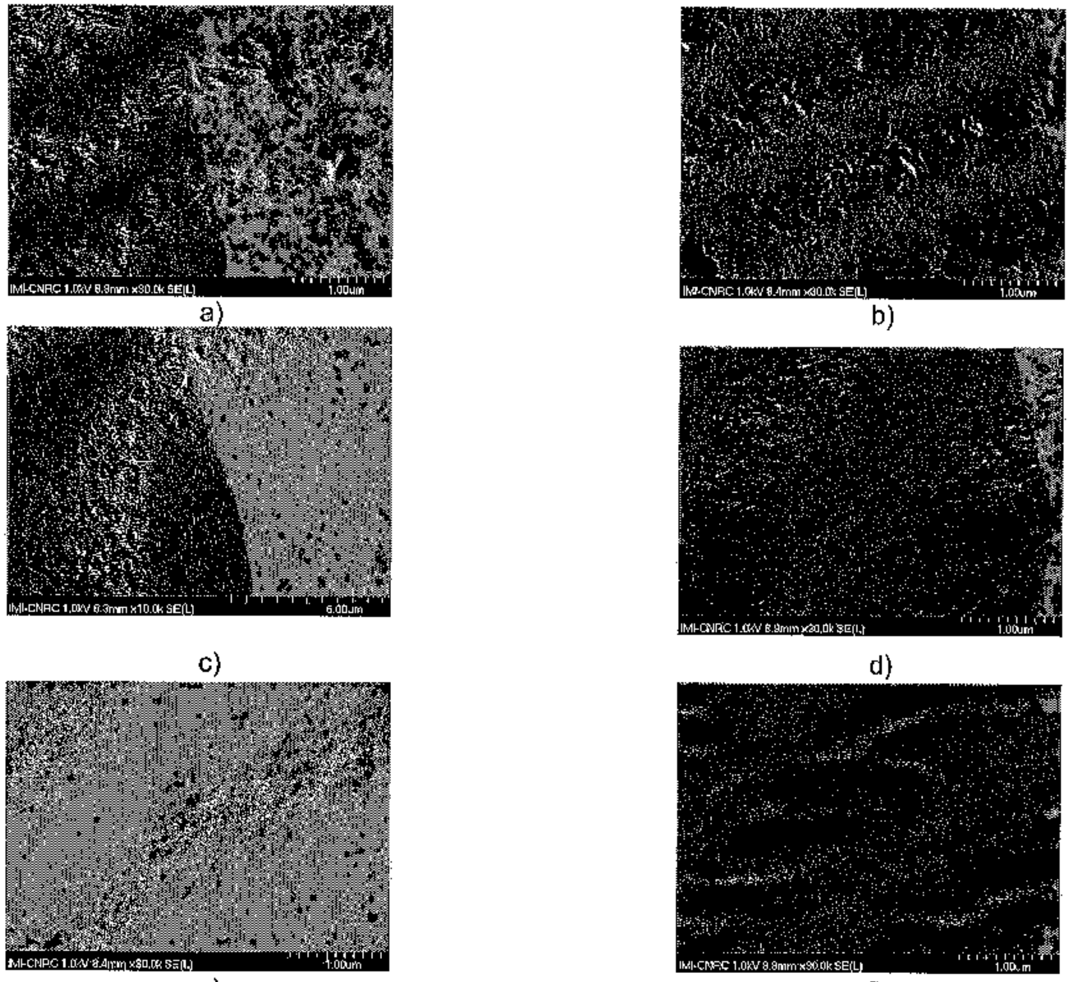
a) b)

Figure 1: True stress-strain curves for films drawn at 90 C and 10 m/min simultaneously, a) without tie layer and b) with tie layer.



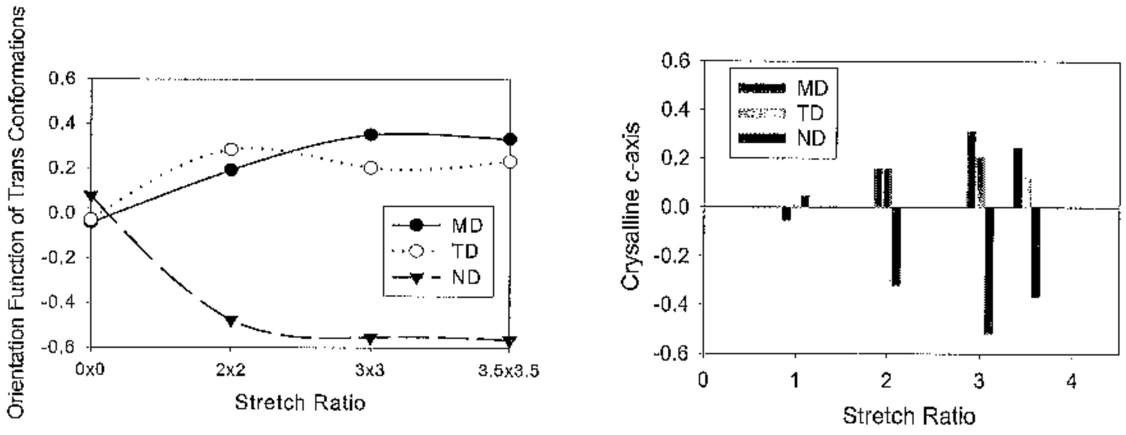
a) b)

Figure 2: True stress-strain curves for films with tie layer drawn to a draw ratio of 3.5x3.5 simultaneously; a) effect of draw temperature for a draw speed of 10 m/min and b) effect of draw speed at 100 C.



a) b) c) d) e) f)

Figure 3: SEM micrographs for a) initial film (LDPE outer surface) without tie layer; b) initial film (LDPE outer surface) with tie layer; c) LDPE layer (outside) drawn to 1.5x1.5; d) LDPE layer (outside) drawn to 3.5x3.5; e) PET layer (outside) drawn to 1.5x1.5; f) PET layer (outside) drawn to 3.5x3.5



a) b)

Figure 4: Orientation of a) PET trans conformers and b) LDPE crystalline c axis.

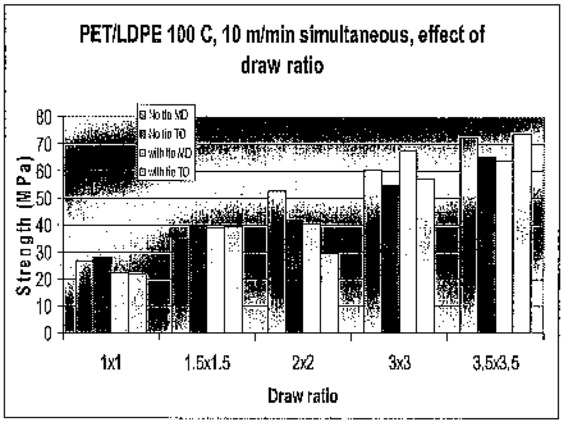


Figure 5a: Tensile strength of the drawn films.

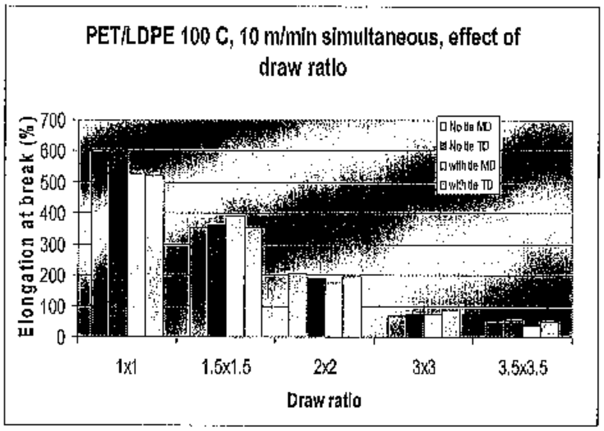


Figure 5b: Elongation at break of the drawn films

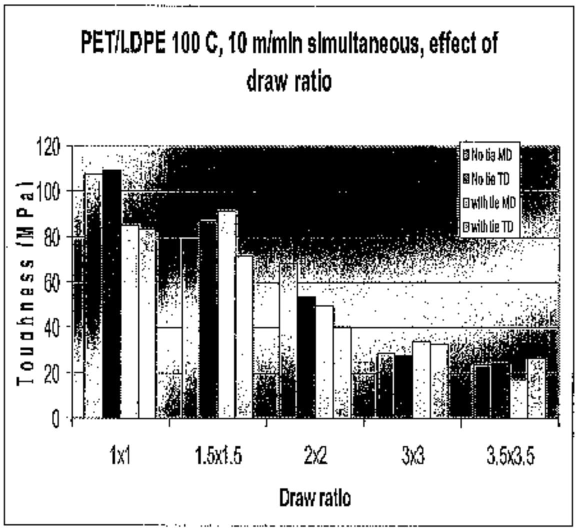


Figure 5c: Toughness vs draw ratio

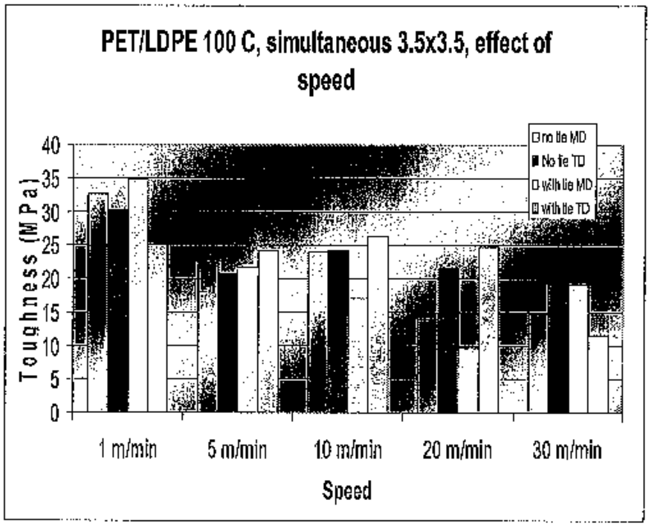


Figure 6: Toughness vs stretch speed

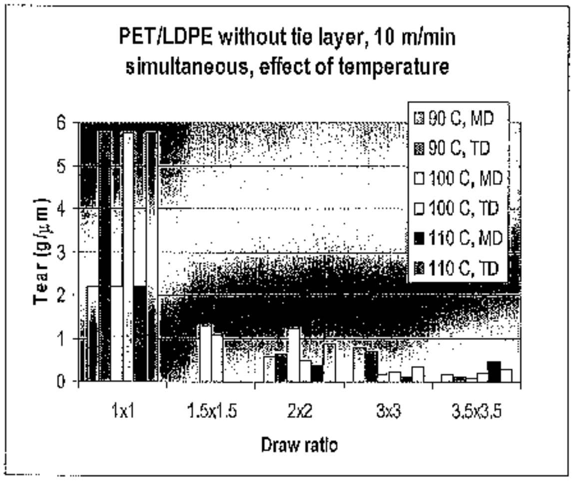


Figure 7: Tear strength vs draw ratio

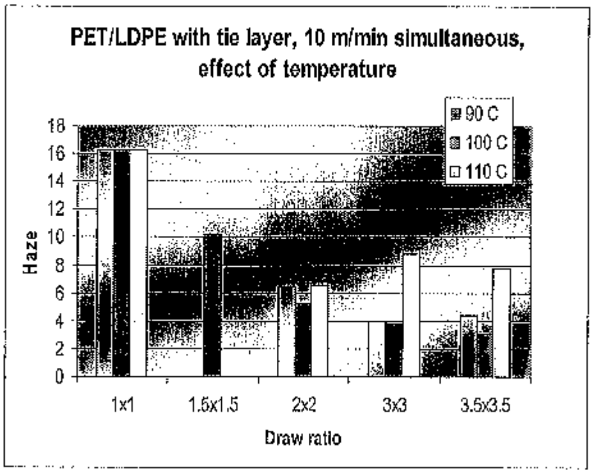


Figure 8: Haze vs draw ratio.