

Improving the Softness of BOPP Films: From Laboratory Investigation to Industrial Processing*

Ling-pu Meng, Xiao-wei Chen, Yuan-fei Lin and Liang-bin Li**

National Synchrotron Radiation Lab, Key Laboratory of Soft Matter Chemistry, Chinese Academy of Sciences,
University of Science and Technology of China, Hefei 230026, China

Abstract Young's modulus of biaxially oriented polypropylene (BOPP) films prepared with homemade film stretcher was investigated, which can be used to indicate the softness of films. It was found that the modulus of films was decreased by about 69% as the content of polyethylene (PE) added into polypropylene (PP) reached 30%. Also, increasing draw temperature can induce lower stress level during stretching, which may lead to the formation of crystals with low orientation level and thus decreased modulus of films. Based on laboratory study, BOPP films produced on commercial line were studied by differential scanning calorimetry (DSC), wide and small-angle X-ray scattering (WAXS, SAXS) with varying contents of PE. SAXS results show that the crystals are oriented in both machine direction (MD) and transverse direction (TD), and the crystals are more oriented in TD than MD according to the WAXS results for all films. Also, the orientation parameter of crystal along TD increases from 0.68 to 0.83 as the contents of PE increase from 0% to 25%. Meanwhile, the modulus of films in MD declines with increase of PE contents generally, improving the film softness. Orientation of crystals is thus an effective structure parameter to adjust the film softness. The relationship of processing-structure-property is also established.

Keywords Polymer films; Stretching processing; Processing-structure-property relationship; Crystal orientation

INTRODUCTION

Biaxially oriented polypropylene (BOPP) film is widely used because of its excellent and comprehensive properties, especially its mechanical strength. During processing, the film is stretched to about five times in machine direction (MD) and up to ten times in transition direction (TD) sequentially, forming the crystals with high orientation order. Therefore, the film products possess high modulus, strength as well as stiffness^[1]. Up to now, BOPP films constitute the largest share in biaxially oriented film and the markets become increasingly mature and saturated^[2]. There exists an urgent need to produce films for specialty-type applications on existing equipments^[1, 3], among which BOPP films with high softness are greatly needed. It can replace coverlay films to cover and protect flexible printed circuit board (FPCB) from chemical, environmental and mechanical damage as it is capable of being flexed without loss of the adhesion^[4, 5]. Nevertheless, BOPP films made on present equipments own high stiffness and cannot adhere FPCBs closely. More efforts should be done to improve the softness of BOPP films to increase the additional value of the film products.

On one hand, there are no industrial standards characterizing the softness of polymer films. Generally, softness is indicated by flexural modulus^[6], which cannot be obtained for thin films according to the present testing standard as the film is too soft to stand itself between two pedestals. Furrer *et al.* used the 1% secant modulus to measure the relative softness of the films. In this regard, the lower the calculated value for the secant

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** Corresponding author: Liang-bin Li (李良彬), E-mail: lbli@ustc.edu.cn

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modulus, the softer the films^[7]. Learning from rubber or plastics coated fabrics-determination of flexibility-flat loop method (Standard No. ISO 5979:1982), Harrabi *et al.* evaluated the flexibility of protective gloves^[8]. The testing principle of the standard is that two ends of a piece of film with specified dimension are folded along the given direction to form a closed circle, the height of which is used to characterize the film softness (the smaller value for the measured height, the softer the films)^[9].

On the other hand, no studies have been carried out to adjust and control the softness of BOPP films in previous studies. It is generally recognized that properties of polymer products are mainly affected by the raw material formula as well as the processing conditions. With regard to the influence of materials, efforts have been made to obtain polymer products with comprehensive properties by blending. Studies show that the modulus and tensile strength of PP/PE blends increase with the content of PP, agreeing with a logarithmic mixing rule^[10, 11]. Although the object of study is not film products, it can still provide guidance to adjust the properties of BOPP films. As for the stretching conditions, Chandran *et al.* studied the effects of temperature and extension rate on the orientation level of poly(ethylene terephthalate) (PET) chains. As the temperature increases, the chains of polymer are free to move owing to higher thermal energy and therefore have a greater tendency to relax. This causes a net decrease in the degree of orientation as some of the oriented chains relax back to the random state, showing lower stresses in the stress-strain curves. Increasing the stretch speed has the same effect as lowering the draw temperature. Nevertheless, the studies do not correlate the orientation to the mechanical properties of BOPET films^[12–14]. Still, due to the higher speeds and strain rates on commercial equipment than those on lab scale, how to translate the processing parameters defined on lab stretcher to commercial line is still an unsolved question.

In this work, two property parameters are employed to evaluate the softness of BOPP films: one is the Young's modulus of the film, the other is the height of the closed loop adopted from ISO 5979:1982. Using homemade film stretcher^[15], the influence of stretch temperature and content of PE added into PP matrix on the modulus of stretched films was investigated. Under draw temperature of 143 °C, increasing the content of PE from 0% to 30% can reduce the modulus from 1788 MPa to 560 MPa. As to the effect of draw temperature, the content of PE is set to be 20%. Higher stretch temperature results in lower stress level and decreased modulus of the film. This is due to the fact that the orientation level of crystals or molecular chains is relatively low at higher temperature as the molecular chains relax more easily. On the basis of lab study, films were produced on Brückner 8 meter BOPP lines with PE content of 0% to 25%. The crystal structures of stretched films were analyzed using differential scanning calorimetry (DSC), wide and small-angle X-ray scattering (WAXS and SAXS), respectively. The results show that the crystallinity of films, as measured by DSC, does not change with the content of PE. The SAXS results indicate that the crystals are oriented in both MD and TD and the WAXS results reveal that the crystals are more oriented in TD than MD. Also, the orientation parameter along TD determined by WAXS increases from 0.68 to 0.83 as the content of PE increases from 0% to 25%. The modulus in MD and the height of circle loop decrease with increasing PE content due to the increased orientation level in TD. As a consequence, the softness of BOPP films is enhanced, improving the additional value of the product. By this way, the relationship of processing (PE content), structure (crystallinity, crystal orientation level) and property (softness) is established, beneficial to producing films for specialty-type applications. The influence of draw ratio and stretching temperature (T_s) on crystal structure and mechanical properties of films made on the commercial line will be further explored.

EXPERIMENTAL

Laboratory Study

A commercially available iPP, F03D, was supplied by Sinopec Maoming Company, China. Film grade low density polyethylene (LDPE), 2426H, was supplied by BASF-YPC Company Limited, China. The melting peak points (T_m) of the PP and PE were measured to be 161.6 and 111.7 °C, respectively, by DSC (Q2000, TA Instruments) at a rate of 10 K/min. The polymer blends containing different contents of PP and PE were extruded

in a single screw extruder (die temperature 250 °C) that were subsequently cooled on a chill-roll operating at a temperature of 40 °C. The cast film was about 0.4 mm thick. Samples of about 130 mm × 130 mm were cut from the cast film for biaxial stretching using a homemade film stretcher. The drawing was performed under unequal simultaneous mode^[12] with stretching speed of 20 mm/s in both the directions. Draw ratios of MD and TD were 5 and 8, respectively. The specimen was first stretched to 5 in two directions, and then the stretch was stopped along MD while continued up to 8 along TD. After stretching, the film was heat fixed under the stretch temperature for 5 min in the stretch oven. The blend compositions and stretching temperature for two pairs of experiments are listed in Table 1. The PE contents were increased from 0% to 30% as the stretching temperature was fixed at 143 °C. With respect to the influence of draw temperature, the PE content was set as 20% and the draw temperature was increased from 131 °C to 143 °C with an increment of 4 °C. During stretching, force-elongation curves in two stretching directions were recorded.

Table 1 The recipe and stretching temperature of the laboratory studies for the purpose of improving the film softness

PP content (%)	PE content (%)	T_s (°C)			
100	0	143			
90	10	143			
80	20	143	139	135	131
70	30	143			

Industrial Processing

On the basis of the laboratory study, we translated the composition to commercial line. The experiment was conducted on the Brückner BOPP production line in Anhui Guofeng Plastic Industry Co., Ltd. (Anhui, China). The stretch temperature in MD and TD was 115 and 150 °C, respectively, while the thermoforming temperature after stretching was 155 °C. Draw ratios in the two directions were 5 and 8, respectively. The PE contents of stretched films were increased from 0% to 25% with the increment of 5%. As the PE content was increased to 30%, the BOPP films could not be produced in succession due to the rupture of films on commercial line.

Characterization

The drawn films from lab stretcher and commercial line were cut along MD into strips with 15 mm in width and 150 mm in length for mechanical performance testing, which was performed at room temperature on Universal Testing Machine (Mts System (China) Co., Ltd.). A cross-head speed of 100 mm/min and the initial distance between two clamps of 100 mm were used. The tensile strength, and Young's modulus were recorded. The films obtained from commercial lines were analyzed for crystallinity, morphology and orientation. The melting behavior of the films was analyzed using differential scanning calorimeter (DSCQ2000, TA) under a nitrogen atmosphere. The samples were heated from 0 °C to 200 °C at a heating rate of 10 K/min. The crystallinity of the specimens was calculated from enthalpy change values obtained in the heating curve, supposing that fully crystalline PP has a heat fusion of 209 J/g. When calculating the absolute crystallinity of PP crystals, the content of PP was divided as follows:

$$X_c = \frac{\Delta H_m}{K \Delta H_m^0} \quad (1)$$

where X_c is the absolute crystallinity of PP crystals, ΔH_m is the enthalpy change, ΔH_m^0 is the heat fusion of fully crystalline PP and K is the content of PP in the composition.

Wide-angle X-ray scattering (WAXS) measurements were performed using an in-house micro X-ray source (Incoatec, 1 μs) with Cu K α radiation ($\lambda = 0.154$ nm). The scattering intensity was collected by Mar 345 image plate with a resolution of 3072 × 3072 pixels (pixel size of 150 μm). Small-angle X-ray scattering (SAXS) measurements were carried out at the BL16B beamline in Shanghai Synchrotron Radiation Facility (SSRF). The X-ray wavelength was 0.124 nm and SAXS patterns were collected with a Mar165 CCD detector (2048 × 2048 pixels with pixel size of 80 μm). In order to obtain X-ray scattering through the thickness of stretched

films, 16 layers of thin films were stacked together. The exposure time for WAXD and SAXS measurements was 300 and 20 s, respectively.

Orientation parameter f of the film is obtained from full width at half-maximum (FWHM) of the azimuthal distribution of diffraction intensity of (040) according to Hemans orientation function defined as:

$$f = \frac{3\langle \cos^2 \varphi \rangle - 1}{2} \quad (2)$$

where φ is the reference direction (MD or TD) and the normal of lattice planes. Thus, f equals 1 when all the (040) planes are perpendicular to the reference direction, while 0 for no preferred orientation state.

RESULTS AND DISCUSSION

Influence of Formula and Stretching Temperature on Mechanical Property of Film Stretched by Lab Stretcher

Figure 1(a) shows the photograph of films stretched to 5×8 times on lab stretcher and the MD and TD stretching processes on commercial line are shown as Figs. 1(c) and 1(d), respectively. Figure 1(b) presents the schematic of ISO 5979:1982: film with 250 mm in MD and 100 mm in TD folded to form a closed circle, the height of which is used to represent the film softness. The larger value the height is, the lower level of the film softness can be obtained.

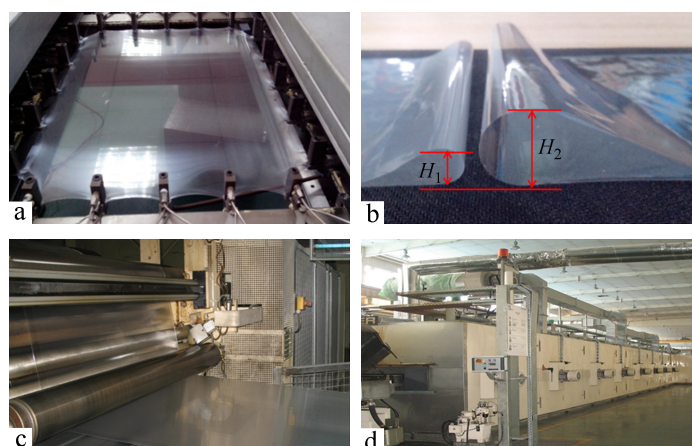


Fig. 1 (a) Photograph of BOPP film after unequal simultaneous biaxial stretching with draw ratio of 5×8 ; (b) A method to characterize the softness of polymer films (Two sides of a piece of film with specified size are folded to form a closed loop. The distances between the top of the loop and the base level (H_1 or H_2) are used to represent the softness of different films.); (c) MD stretching and (d) TD stretching of the Brückner BOPP lines during processing in the manufacturing shop (The online version is colorful.)

Figure 2 presents the stress-strain curves in TD under the stretching temperature of 131 and 139 °C, respectively. Under 139 °C, the stress increases almost linearly with draw ratio and then reaches a stress maximum. Beyond the yield point, the stress evolution undergoes a weak strain softening and then strain hardening process. However, for stretch under 131 °C, the stress evolution experiences the strain hardening period after yield point, not undergoing the strain softening region. Also, the force level is higher than that under 139 °C.

Figure 3(a) presents the Young's modulus of films containing different contents of PE, which were drawn by lab stretcher under 143 °C. As the PE content increases from 0% to 20%, the modulus of BOPP film decreases from 1788 MPa to 451 MPa, with a decline proportion of 75%. Further increase of the PE content to 30% has little influence on the film modulus. With respect to the influence of stretch temperature, the modulus of films stretched at different temperatures are shown in Fig. 3(b). The PE content is set as 20% for all experiments. The modulus declines from 878 MPa to 451 MPa as the draw temperature increases from 131 °C to

143 °C, showing a decline amplitude of 49%.

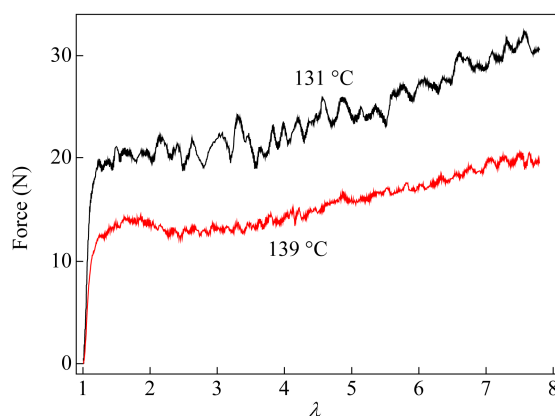


Fig. 2 Stress-strain curves in TD during stretching at 131 and 139 °C, respectively

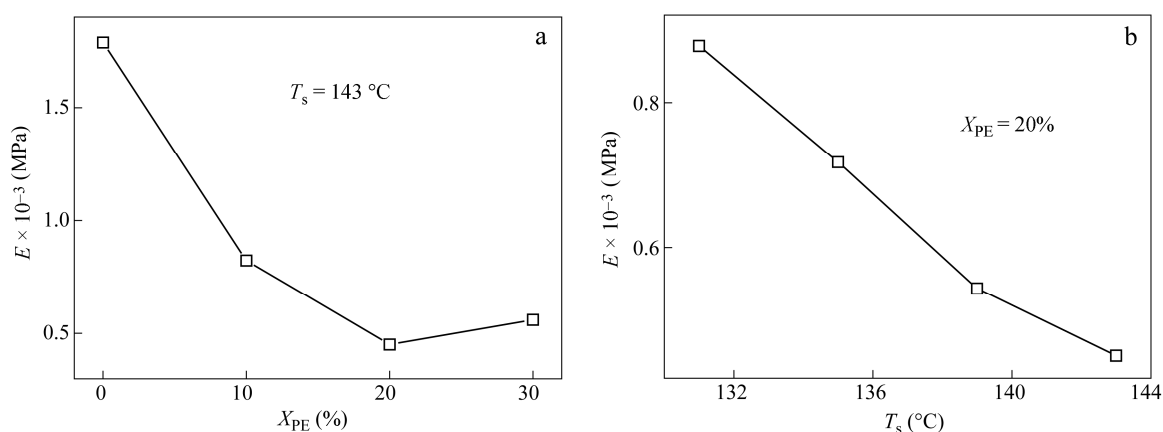


Fig. 3 Young's modulus of films (a) containing different contents of PE after stretching at 143 °C and (b) stretched at different temperatures with PE content of 20%, respectively

Effect of Composition on Structure and Mechanical Property of Films Produced on Commercial Line

Figure 4(a) shows the thermograms of different films produced on commercial line. The curves are shifted vertically to give a better view. As can be seen from the melting curves, there exist weak melting peaks at around 118 °C (as indicated by black dash line) for the films containing PE. These weak peaks originate from the melting of PE crystals during heating. A broad shoulder near 158 °C (as indicated by the red dash line) and a sharp melting peak at about 165 °C are observed for the BOPP films. The quantitative results calculated from the thermograms are presented in Fig. 4(b). The enthalpy of PP (black open square) in the first heating run decreases from 100.6 J/g to 74.4 J/g as the PE content increases from 0% to 25%, showing a 26% descend range. Interestingly, the descend range is in accordance with the amplification of PE content. The absolute crystallinity of PP crystals has nothing to do with the PE contents, remaining the same for all BOPP films. It indicates that PP/PE blend is an immiscible system, where PE has no effect on the crystallization behavior of PP^[16, 17]. This may be the reason why PP remains the biaxial stretchability in spite of the addition of PE^[18–20].

Figure 5(a) presents the two-dimensional (2D) WAXS patterns of different BOPP films. The PE contents are marked at the top right corner of each scattering pattern and the two stretching directions are indicated by red arrows. As can be seen from the scattering patterns, molecular chains for all films are more oriented along TD. The one-dimensional (1D) integrated WAXS curves are shown in Fig. 5(b). The intensity of scattering peak at around 21.6°, which corresponds to the crystallographic planes (131)/(041) of PP and crystallographic plane

(110) of PE, increases as the PE content increases. The peak intensity at around 24° , corresponding to crystallographic plane (200) of PE, also increases with the PE contents. Based on the azimuthal intensity distribution of scattering from crystallographic plane (040) of PP (not shown here), we calculated orientation parameters along TD of molecular chains, which are plotted in Fig. 5(c) versus PE contents. With increasing PE contents from 0% to 25%, the orientation level increases from 0.68 to 0.83.

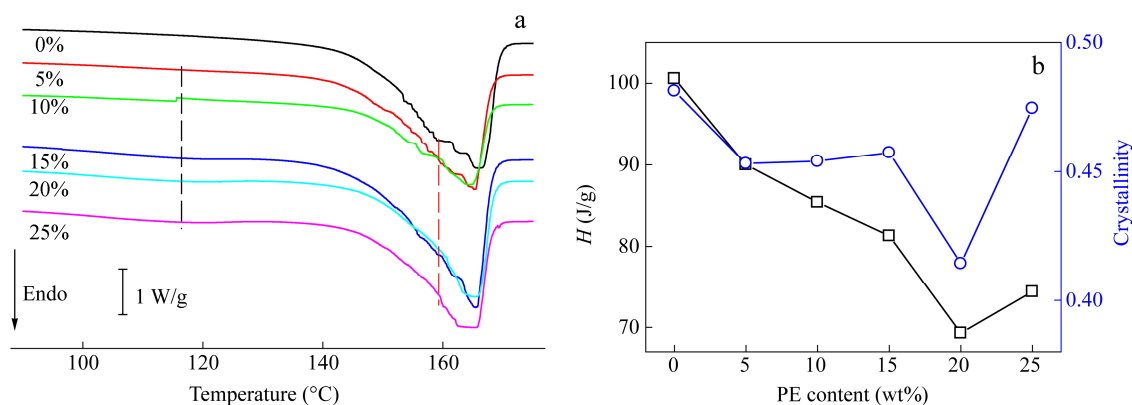


Fig. 4 (a) Differential scanning calorimetry thermograms of different BOPP films produced on commercial line; (b) The corresponding enthalpies of PP (black open square) in the first heating run and crystallinity of PP (blue open circle) calculated from enthalpy change values obtained in the heating curve (Supposing fully crystalline PP has a heat fusion of 209 J/g.) (The online version is colorful.)

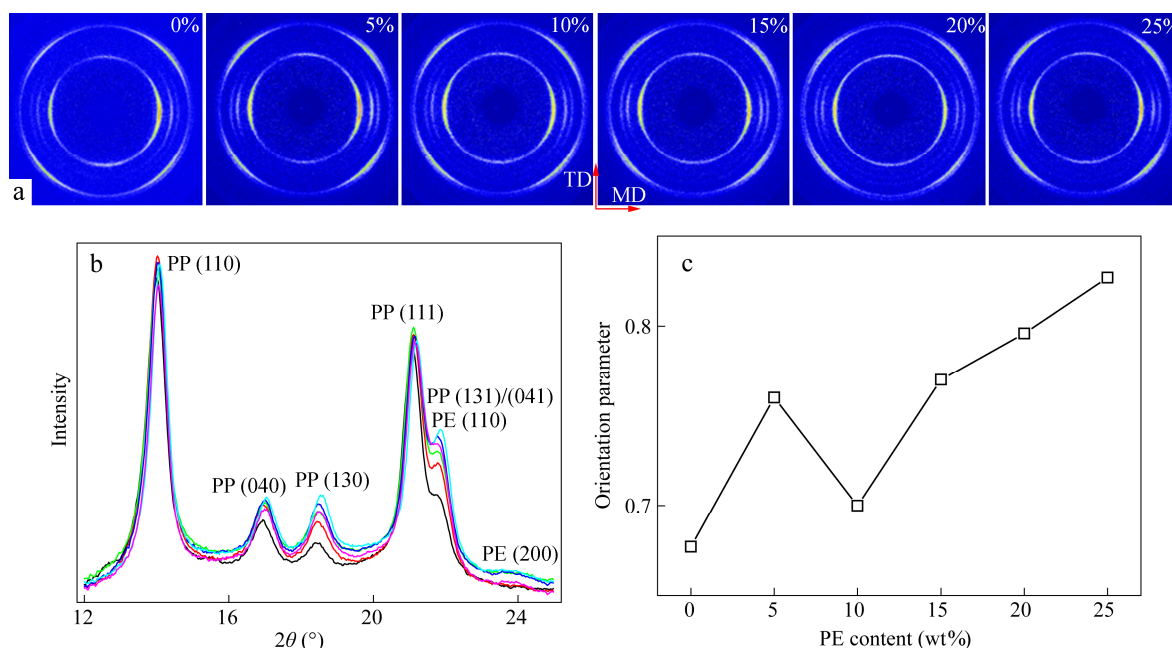


Fig. 5 (a) 2D WAXS patterns of the BOPP films with different contents of PE (The two stretching directions are indicated by the red arrows.); (b) 1D integrated WAXS curves; (c) Orientation parameter along TD for different BOPP films (The online version is colorful.)

Figure 6(a) shows the 2D SAXS patterns of different BOPP films, at the top right corner of which, the PE contents are labeled. The two stretching directions are indicated by the red arrows. Two clear scattering maxima due to the periodicity of lamellae appear in both stretching directions, showing that the crystals are oriented in both MD and TD. Also, the scattering maxima in TD are closer to the beamstop, indicating that the long period

in TD is larger than that in MD. Except for the scattering maximum, a faint elliptic isointensity contour with its long axis parallel to MD appears as PE is added to PP. The faint contour may originate from the scattering of deformed crystal of PE due to the different draw ratios in two directions^[21]. The corresponding azimuthal-integrated intensity distribution of the 2D SAXS patterns are presented in Fig. 6(b). The azimuthal angle of integrated regions is from 0° to 180° as shown by the insert in Fig. 6(b), where 0° represents the horizontal direction. On the basis of 1D integrated curves along in Fig. 6(b), the full width at half maximum (FWHM) of each curve versus PE content can be obtained, as indicated by Fig. 6(c). Generally, the FWHM shows a downtrend with increasing PE content. It indicates that the orientation level of crystals along TD becomes larger^[22, 23] as the PE content increases, which is in accordance with the WAXS results. For the films without PE, the FWHM of azimuthal-integrated curves is out of the trend. The reason for this is that no scattering signal from PE interferes with the scattering maximum of PP, resulting in the lower level of FWHM.

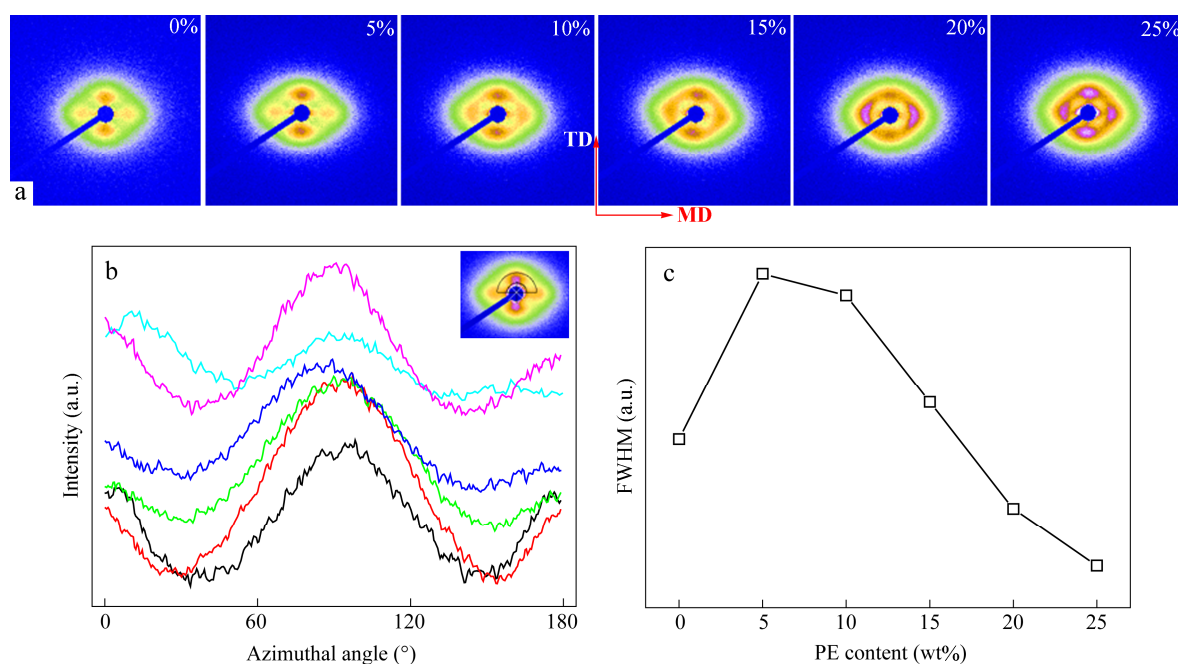


Fig. 6 (a) 2D SAXS patterns of the BOPP films with different contents of PE (The two stretching directions are indicated by the red arrows.); (b) Corresponding azimuthal-integrated intensity distribution of the 2D SAXS patterns, where 0° represents the horizontal direction (The inset shows the integrated region of scattering intensity along azimuthal angle.); (c) The full width at half maximum of the corresponding azimuthal-integrated intensity distribution curves to represent the orientation level of crystals along TD (The online version is colorful.)

The modulus along MD as well as tensile strength in both MD and TD of BOPP films with different compositions are shown in Fig. 7. As the content of PE added into PP matrix increases from 0% to 25%, the modulus of films in MD decreases obviously from 1800 MPa to 1262 MPa. With regard to the tensile strength, it shows a downward trend in both MD (σ_{MD}) and TD (σ_{TD}) with the increase of PE content. The tensile strength decrease from 152.5 MPa to 116 MPa and from 311.9 MPa to 245.2 MPa for MD and TD with PE content of 25%, showing about 24% and 21% decrement, respectively. It should be noted that the modulus and tensile strength of films are much larger than those of the raw materials. According to the technical data provided by raw material supplier, the moduli of PP and LDPE used in this work are 1100 and 260 MPa. And also, the tensile strength are 35 and 26 MPa for PP and PE, respectively. The reason is that during biaxial stretching process, molecular chains and crystals are highly oriented, resulting in the substantial enhancement of mechanical properties compared to the raw materials. In this work, we analyzed the variation of mechanical properties from the aspects of chain or crystal orientation.

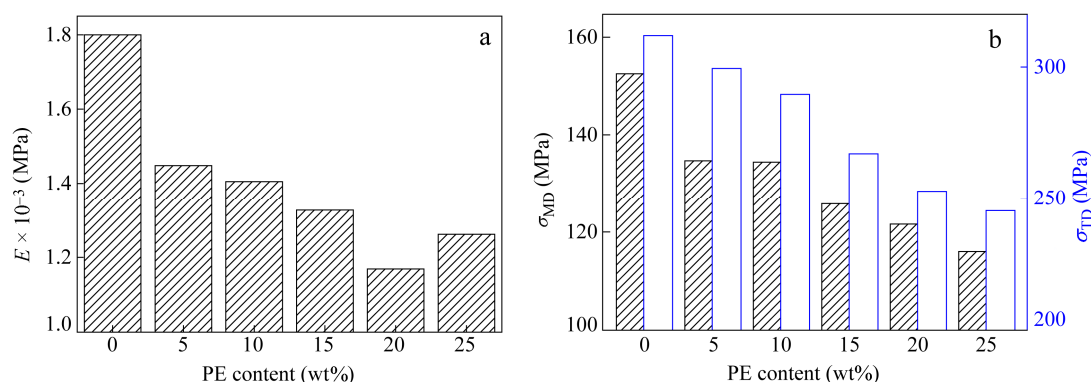


Fig. 7 (a) The Young's modulus along MD and (b) tensile strength in both MD and TD of BOPP films versus PE content (The online version is colorful.)

Figure 8 presents the variation trend of height of closed circle (black open square) and the Young's modulus (blue open circle) of films with orientation parameter, used to indicate the softness of BOPP film. The value of closed circle height decreases from 15 mm to 12.5 mm as the orientation parameter increases from 0.68 to 0.83 with the increase of PE contents. As for the Young's modulus, it declines from 1800 MPa to 1262 MPa along the orientation level, showing a 30% decrement.

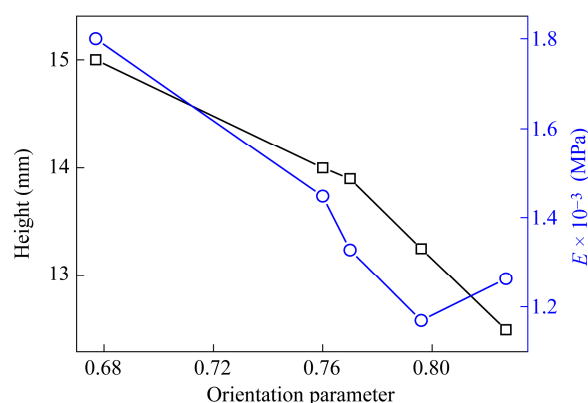


Fig. 8 The change of height of closed circle (black open square) and the Young's modulus (blue open circle) of films with orientation parameter along TD

Establishment of the Relationship of Processing, Structure and Properties

The above results lead to several interesting findings, which allow us to establish the relationship between processing-structure-mechanical property of BOPP films. For laboratory investigation, we derive two observations. (i) Contents of PE show negative correlations with the modulus of BOPP films. (ii) Stretching at higher temperature results in lower stress level and lower modulus of stretched films. The modulus of PE is lower than that of PP, thus the stretched film containing PE has lower modulus than film composed of pure PP^[24]. Also, PE is melted under draw temperature, acting as flow media. After MD stretching is stopped, molecular chains flow more easily towards TD as PE content increases^[25], resulting in higher orientation level of molecular chains along TD. Thus, the modulus of films in MD decreases with increasing PE content. As for the effect of draw temperature, stretching at lower temperature results in higher stress level, which induced more oriented structure in both the directions^[26]. Therefore, the modulus of stretched films in MD declines with draw temperature^[27]. As mentioned above, the modulus can be used to represent the softness of polymer films. As a consequence, adding PE into PP matrix and increasing draw temperature can both improve the softness of BOPP films.

Based on the results from laboratory investigations, BOPP films containing different contents of PE are produced on commercial line. From the results, two observations can be extracted. (i) The molecular chains and crystals of stretched films are more oriented towards TD. Increasing content of PE can increase the orientation level along TD. (ii) The height of closed circle and Young's modulus in MD decrease with the increment of orientation level caused by the increasing of PE contents. Firstly, the draw ratio in TD is larger than that in MD. In addition, after initial stretching in MD, subsequent TD stretching is more efficient as stretching has already induced orientation and crystallization of molecular chains during MD stretching^[28]. Therefore, the crystals are more oriented in TD than in MD^[29]. On the other hand, PE is in the molten state during stretching in TD at 150 °C, which facilitates the motion and orientation of PP chains, resulting in higher orientation level along TD with PE content increasing. With regard to the mechanical properties of BOPP films, the relative orientation level along MD declines with increasing PE contents, resulting in the lowering of Young's modulus and flexural modulus in MD as well as the height of closed circle. As a consequence, the softness of BOPP films is improved. Besides, the relationship of processing (PE contents), structure (orientation level of crystals) and properties (softness) is established. The effect of stretching temperature and draw ratio on structure and property of BOPP films produced on commercial line will be further explored.

CONCLUSIONS

Two parameters can be employed to represent the softness of BOPP films: one is the height of a closed circle formed by folding a piece of film with specific dimension; the other is the Young's modulus. Laboratory investigations are carried out to figure out the effect of formula and processing conditions on film softness. Increasing PE content from 0% to 30% can decrease the modulus of BOPP films from 1788 MPa to 560 MPa. Higher stress level under lower draw temperature can induce formation of crystals with higher orientation level and thus higher modulus of BOPP films. As a consequence, adding PE into PP matrix and increasing draw temperature are effective methods to improve softness of films. On the basis of laboratory studies, PE are added into PP to improve softness of BOPP films produced on commercial line. Increasing the PE content from 0% to 25% can improve the orientation parameter along TD from 0.68 to 0.83. The height of the closed circle and modulus along MD are thus declined, improving the film softness. Also, the relationship of processing, structure and properties is well established.

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