

A Universal Equipment for Biaxial Stretching of Polymer Films^{*}

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Abstract A biaxial stretching equipment was designed and constructed to enable fundamental studies of the relationship between film processing conditions and structures of oriented film products. With programmable drive motors and scissor-like mechanism, all stretching modes, including uniaxial stretching with constant and free width, simultaneous and sequential biaxial stretching, can be applied to a square-shaped sheet. Parameters related to film stretching manufacturing, such as temperature, draw ratio and stretching speed can be set independently to meet the requirement of different polymers. The force information during stretching is recorded by two miniature tension sensors in two directions independently, which can monitor the mechanical stimulus and stress response. Using this equipment, experiments are conducted to investigate the influence of stretching parameters on the structure of polypropylene films, which provides an effective method to tailor the processing conditions to obtain the films with desired properties.

Keywords: Biaxial stretching; Processing/structure relationship; Dynamics of stretching.

INTRODUCTION

Biaxial stretching is the process of stretching polymers in both machine direction (MD) and transverse direction (TD). During stretching, polymer chains orient towards two perpendicular directions, producing polymer films with balanced properties in MD and TD^[1]. Also, due to the long-chain nature of semicrystalline polymers, stretching processing is an effective method to modify the crystallization features of the polymers and thus obtain films with excellent mechanical, optical and thermal properties^[2]. Another advantage of biaxial stretching is very high production rates. To date, the speed of the largest commercial tenter BOPP line of Brückner can be 500 m/min with film widths up to 10 meters^[3].

Though biaxial stretching process enjoys a great success throughout the polymer industry since early 1950's^[4], studies to establish the processing-structure-property relationships are still lacking. Much effort has relied on trial and error rather than an understanding of the basic principles behind the film stretching process, which is disadvantageous to developing new and unique film products. Laboratory evaluations are able to provide quantitative information such as mechanical stimulus (strain) and response (stress)^[5–7], providing an effective way to study the influence of processing conditions on film structure and properties. Such studies are useful to predict how new resins and formulations will behave on commercial lines, saving both time and effort^[1].

Typical laboratory stretchers are T.M. Long stretcher^[8] and Karo IV unit from Brückner^[3], which have been widely used in the community. However, they are costly and most research institutes cannot get access to the

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machines^[9]. Much effort has been tried to break the ice. On the basis of the Instron testing machine, Gilbert built a biaxial stretcher which can impose different modes of stretching to different polymers. Nevertheless, the maximum draw rate and draw ratio (both axes) of the equipment are restricted to 500 mm/min and 3.8, respectively^[9, 10]. Recently, Cakmak and co-workers designed and constructed a highly integrated biaxial stretching system, allowing simultaneous monitoring of stress-strain information and optical behavior of polymer films during biaxial stretching. However, stretching speed of the system is limited to 15 mm/s, which is much lower than that of commercial stretchers with 500 mm/s^[4]. In our previous work, a constrained uniaxial tensile apparatus was constructed to obtain stress-strain curve during stretching as well as subsequent structure evolution by *in situ* X-ray scattering technique. Nevertheless, simultaneous biaxial stretching cannot be achieved through the apparatus^[11]. It is recommended that a biaxial stretching equipment comparable to commercial stretcher should be constructed in a lab, making it more convenient for researchers to figure out the basic principles behind film stretching processing process.

In this work, a biaxial stretching equipment was designed and constructed. All film processing parameters can be comparable to commercial stretchers with maximum draw ratio (λ) of 10 (both axes), maximum stretching speed (v) of 500 mm/s and operating temperature up to 250 °C, respectively. Also, all stretching modes, including uniaxial stretching with constant (UCW) and free (UFW) width, simultaneous (SB) and sequential (SEQ) biaxial stretching^[12] related to polymer film stretching manufacturing can be achieved by the equipment. Stress-strain curves during the stretching process can be obtained, which offer the possibility to examine the dynamics of the stretching process^[1]. Combining with the structural information about final stretched films by off-line test, such as crystallinity and degree of orientation, the equipment helps to establish the relationship between processing conditions and structures. Such laboratory investigations based on the equipment provide extremely useful insight into how new polymers and processing technique will behave on commercial lines.

THE BIAxIAL STRETCHING EQUIPMENT

Integrated Layout of the Equipment and Parameter Selection

Figure 1 gives the mechanical assembly drawing of the biaxial stretching equipment (a, front view; b, top view), which contains three main subassemblies. The stretching took place discontinuously as the stretching frame was moved horizontally among different zones. After the specimen had been inserted on the stretching frame, the latter was moved from the loading zone to the preheating zone. The moment the temperatures of both preheating oven and stretching oven reach the set value, the stretching frame is shifted from the preheating oven to the stretching oven. After stretching, the frame is returned to the loading zone and the film is taken out for other

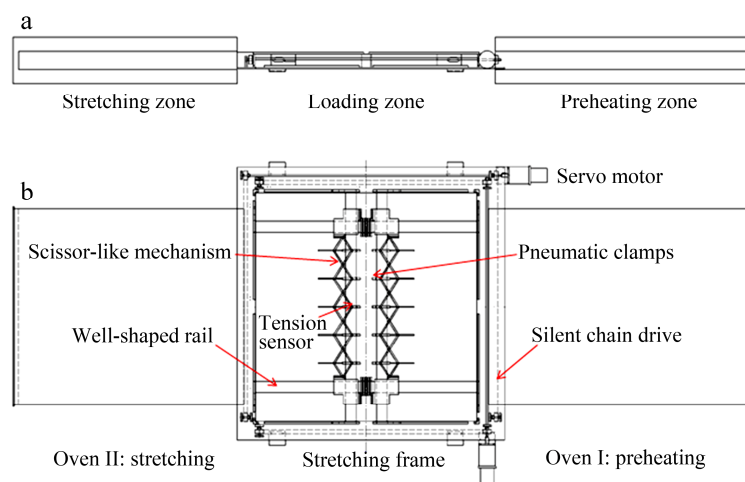


Fig. 1 Mechanical assembly drawing of the biaxial stretching equipment (a, front view; b, top view)

characterization. The preheating oven can also be used for heat treatment and relaxation of films to control dimensional properties of films^[13]. During such a stretching experiment the polymer film experienced a similar stretching processing as encountered under commercial processing conditions.

Parameter selection of the equipment is on the basis of Karo IV unit from Brückner, with which practically any polymers can be evaluated. The unit has stretching speed (v) of 500 mm/s; initial sample dimensions ($L_0 \times L_0 \times H$) of 90 mm $\times$ 90 mm $\times$ 4 mm; stretching ratios of 10.4 on both axes and operating temperature of 300 °C (400 °C). The technical data of our equipment are listed in Table 1, which are comparable with those of the Karo IV unit. The equipment is distinguished by the small size in height direction, which can be combined with on-line measurement such as X-ray scattering system with a vertical layout we have constructed. Each subassembly of the equipment will be described further in detail below.

Table 1. Technical data of the biaxial stretching equipment

Technical data	
Stretching ratios on both axes	10:1
Stretching speed	500 mm/s
Operating temperature	250 °C
Max. sample thickness	4 mm
Min. sample dimensions	104 mm $\times$ 104 mm
Stretching force per axis	1700 N
Machine dimensions	L $\times$ W $\times$ H (m) 6 $\times$ 2.2 $\times$ 0.32

Stretching Frame and Power System

Two servo motors drive the chain sprocket system, making the well-shaped rails in perpendicular directions move independently. Therefore, typical deformation modes encountered in the industrial processes, including UCW, UFW, SB and SEQ stretching can be imposed on polymer samples. Linked by scissor-like mechanism, pneumatic clamps can move apart in exact proportion along their respective rails. This produces an even distribution of the stretching force along four edges of the sample, ensuring that the polymer sheets are deformed homogeneously. Programmable servo motors are selected as power system as they possess varieties of advantages such as quick response and accurate positioning. Besides, they can operate steadily and the control system is simple. To determine the specification of servo motor, calculations have been carried out and all formulas are listed in Table 2. According to Formula (1), the maximum strain rate of the equipment can be 4.8 s^{-1} at the beginning of stretching with maximum v and initial sample length L_0 of 500 mm/s and 104 mm, respectively. In our previous work, the viscosity of $\eta = 1.78 \times 10^5 \text{ Pa}\cdot\text{s}$ is taken as reference to calculate the stretching force^[11]. Considering that the viscosity of sample during biaxial stretching is twice as much as that during uniaxial stretching, a stress of $1.7 \times 10^6 \text{ Pa}$ is generated (Formula (2)). The maximum stretching force of each well-shaped rail is 707 N (Formula (3)).

Table 2. Formulas used in this work

Parameter	Formulas	
Strain rate	$\dot{\epsilon} = 2v/L$	(1)
Stress	$\sigma = \dot{\epsilon} \times \eta$	(2)
Stretching force	$F = \sigma \times A$	(3)
Rotate speed	$n = 60v/\pi D$	(4)
Torque	$T_a = FD/2$	(5)
Friction	$F_0 = fp\pi d^2/4$	(6)

$\dot{\epsilon}$ is the strain rate, v the speed of each well-shaped rail, L the sample length, σ stress, η sample viscosity, F stretching force, A cross-section area of sample, n rotate speed of each servo motor, D diameter of the silent chain wheel, T_a torque of servo motor, F_0 friction between sample and each clamp, f friction coefficient, p nitrogen pressure, d diameter of cylinder block.

Chain sprocket system is employed to transfer the rotation of motor to the rectilinear motion of well-shaped rails, which has the advantage of operating quietly and smoothly. Diameter of chain sprocket and chain width are chosen to be 62.8 mm and 28.5 mm, respectively. The system can bear the load of about 20 kN, certainly meeting our needs. According to formulas (4) and (5), the servo system generating torque of 44 Nm and rotation speed of 152 r/min is needed. Taking into account the efficiency of work transformation, a combination of servo motor with power of 850 W and a 1/10 reducer is employed, which produces a rated torque of 53.9 Nm and a rated rotation speed of 150 r/min, respectively.

The clamps of the equipment are based on tailor-made cylinders, which are mounted on grip bases and moved with sliding block along well-shaped rail. The schematic of clamping system is shown in Fig. 2. Each cylinder contains two stainless steel tubes, seal cover, cylinder block, piston and piston return spring. When clamping is needed, nitrogen gas (N_2) pressure is applied to the piston to compress the sample. On the other hand, N_2 out of the cylinder is conveyed to another cylinder to work as N_2 in, forming a series-wound clamping system. On release of the gas pressure, the piston return spring pushes the piston back and release of sample is finished. Magnetic valves are installed at the N_2 inlet of the first cylinder and N_2 outlet of the last cylinder to control the on-off of the gas. Due to the compensation of N_2 with constant pressure, the grips close progressively as the sample thins during stretching, which can prevent the sample from creeping down^[9]. During stretching, it is the friction between clamps and sample that pull the sample open. The value is 142 N for each clamp as each well-shaped rail supports five pieces of clamps. Taking friction coefficient between samples and clamps of 0.65, the pressure of N_2 should be 4.3 MPa to produce friction of 142 N (Formula (6) in Table 2) as the diameter of the cylinder block is 8 mm. The N_2 pressure of homemade cylinder is set to be 5 MPa.

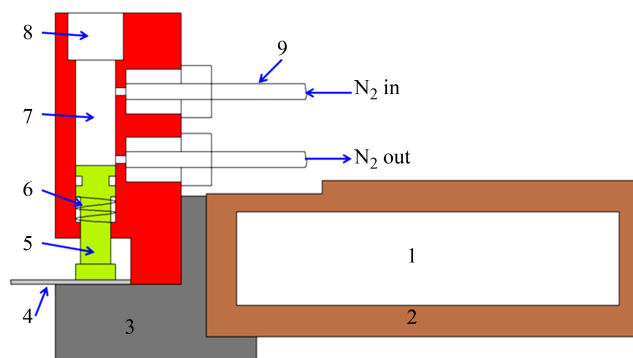


Fig. 2 Schematic of the pneumatic clamps (1, Well-shaped rail; 2, Sliding block; 3, Grip base; 4, Sample; 5, Piston; 6, Piston return spring; 7, Cylinder block; 8, Seal cover; 9, Stainless steel tubes)

Air Heating Circulation System

Air heating circuit system is used to provide the desired temperature condition for film stretching. Two industrial air heaters with the power of 15 kW are employed to heat the preheating oven and stretching oven, respectively. As the circuit systems are the same for both ovens, only the system of stretching oven is described for brevity. Hot air from the heater is divided into two branches through a T-shaped pipe. Then the two branches are driven into the air inlet manifold of the upper and down parts of oven through a short section of insulated ducting. A special pipe network with evenly distributed vents is connected with the air inlet manifold and air flows outward from these vents. Since the upper and lower parts of the oven have the symmetric pipe system, both faces of the sample can be heated, which not only increases heating efficiency, but also makes the sample heated homogeneously. Also, an insulated flexible ducting connected with the air outlet manifold of the oven completes the circuit by returning the air to the industrial air heater. Temperature control of the oven is achieved by adjusting the power supply to the heaters in response to temperature measurements made inside the oven using a thermocouple. Two measurements have been taken to make the temperature in the oven homogeneous. First, as mentioned above, two symmetric pipe network with evenly distributed vents can heat both faces of the sample.

Second, two air fans with diameter of 150 mm are installed directly in front of two faces of the sample to mix the air around the sample homogeneously. They also speed up the air convection heat transfer and heat the sample to the temperature of the oven quickly. Two thermocouples positioned on the middle clamps of the perpendicular rails are applied to monitor the temperature at different locations of the oven by stretching to different draw ratios^[13]. The oven has an operating range of ambient to 250 °C.

Control System

An integrated Labview program is compiled to control the operation of the equipment, including stretching of samples, tension data acquisition as well as the movement of stretching frame among different zones. During stretching process, two servo motors are driven by the Labview program through which various deformation modes, draw ratios and stretching speed for each axis and the strain rate profile (Cauchy or Hencky) can be presetted as needed. With regard to the tension data acquisition, two miniature tension sensors are mounted behind the other two middle clamps of the perpendicular rails. Tension evolution of both directions during stretching can be tracked by the two sensors, which can work normally below 300 °C with 0.05% accuracy. The program is used to collect the tension information through a National Instrument NI 6008 data acquisition card^[14]. Also, when shift of stretching frame among different zones is needed, the program drives the servo motor, the rotation of which can be transferred to rectilinear motion of stretching frame through ball screws.

PP FILMS STRETCHING MEASUREMENTS

Influence of stretching modes on structure of polypropylene (PP) films is studied using this equipment. The PP casting sheets used in this study were kindly supplied by a film company in Hefei, China, resulting from the extrusion of the molten polymer onto the water-cooled cast roll. A stretching experiment was started by loading the specimen in the frame. Each sheet with thickness of 0.7 mm was cut into a square and gripped by clamps in both MD and TD while the film width in TD is free of constraint under UFW stretching. The actual specimen size was 104 mm × 104 mm. For each experiment, the preheating and stretching temperature were chosen as 110 °C and 152 °C, respectively. Immediately after stretching, the stretching frame was moved to the preheating zone and the film was heat fixed under 125 °C for 10 min. The casting sheets and films obtained under different stretching modes were analyzed for morphology, orientation, melting behavior and crystallinity. Small-angle X-ray scattering (SAXS) 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 a multi-wire proportional chamber detector (Bruker, Hi-star) with a resolution of 1024 × 1024 pixels (pixel size of 105 μm). In order to obtain X-ray scattering through the thickness of stretched films, several layers of thin films were stacked together. The melting and crystallization behavior of the casting sheet and films were 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 fully crystalline PP has a heat fusion of 209 J/g^[15]. During each experiment, tension sensors in both directions are used to track the stress evolution.

Figure 3 presents the pictures of PP films obtained during different stretching modes at 152 °C with v of 20 mm/s. As can be seen from the pictures, homogeneous films were obtained after all stretching modes and the influence of clamps on the homogeneity of films can be neglected. The casting sheet gripped by pneumatic clamps before stretching is shown in Fig. 6(e).

Figure 4 shows 2D SAXS images of PP casting sheet and PP films under different stretching modes. Before stretching, 2D SAXS pattern shows a scattering ring with a little higher intensity in the vertical direction, indicating the formation of crystals with weak orientation in casting sheets. As for films obtained by UCW and UFW stretching, a scattering maximum concentrates in the meridional direction. It indicates the formation of highly oriented lamellar stacks with a layer normal to the stretching direction. Interestingly, the scattering maximum under UCW mode has a narrower distribution along the azimuthal angle than that under UFW mode. The phenomenon manifests that the lamellae formed after UCW stretching have larger lateral size than those

after UFW stretching^[2, 16]. Under UCW stretching, constraint in TD can limit the orientation of molecular chains towards MD to some degree^[17, 18], leading to smaller nucleation density comparing to UFW mode. Thus lamellae nucleated from those nuclei have more space to grow to large lateral size before lamellae impingement occurs^[19, 20]. While for films stretched by SB and SEQ modes, weak scattering signal disperses along the azimuthal angle, indicating that there is no preferred orientation for films after biaxial stretching. The reason is that biaxial stretching tends to orient the polymer chains in the plane of the film while maintaining in-plane isotropy.

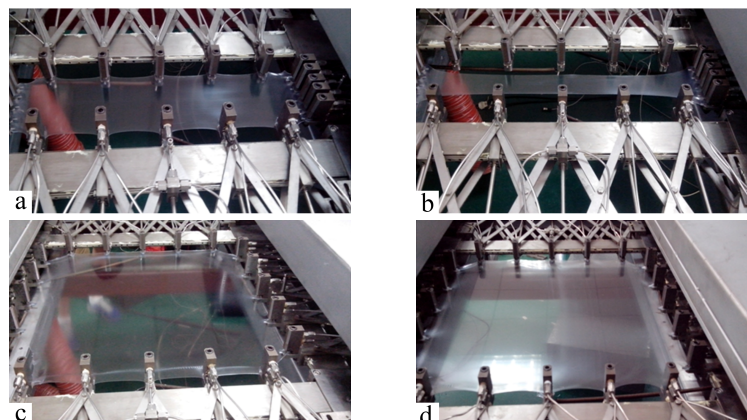


Fig. 3 Pictures of PP films obtained during different stretching modes at 152 °C with v of 20 mm/s: (a) UCW stretching, $\lambda = 5 \times 1$; (b) UFW stretching, $\lambda = 5$; (c) SB stretching, $\lambda = 5 \times 5$; (d) SEQ stretching, $\lambda = 5 \times 5$

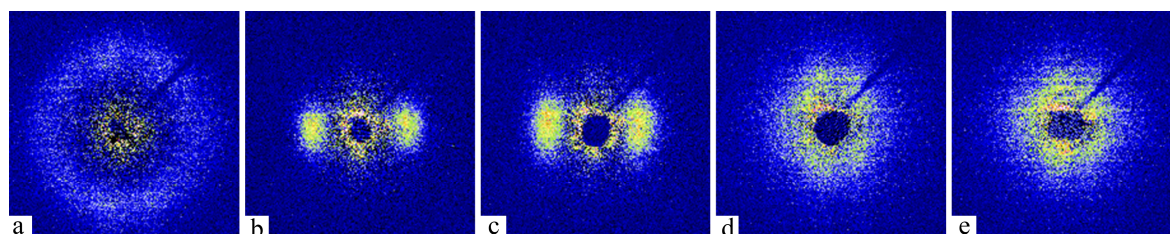


Fig. 4 2D SAXS images of the PP casting sheet (a), PP films under UCW (b), UFW (c), SB (d) and SEQ (e) stretching, respectively

Figure 5 shows the thermograms of casting sheet before stretching and stretched films under UCW, UFW, SB and SEQ modes, respectively. The quantitative results are summarized in Table 3. Generally, the melting point of the films after stretching is higher than that of casting sheets. Since during stretching, spherulitic crystals are partially transformed into high-melting point species as fibrous crystals. Corresponding to the higher melting temperature, films under all stretching modes have a higher degree of crystallinity than that of casting sheets. Because of rapid cooling from the melt, the casting sheet has a relatively low crystallinity. During stretching at temperatures near the melting point, the initial crystalline morphology is partially destroyed and a new, highly ordered one is generated by strain induced crystallization^[21].

Table 3. Melting point and crystallinity of the PP casting sheet (CS); PP films under UCW, UFW, SB and SEQ stretching, respectively

	Melting point (°C)	Crystallinity (%)
CS	161.2	42.5
UCW	167.5	46.4
UFW	167.9	48.4
SB	166.0	44.3
SEQ	170.0	44.1

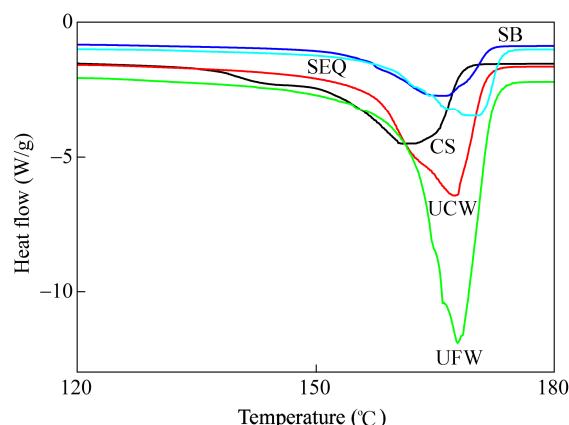


Fig. 5 Differential scanning calorimetry thermograms of the PP casting sheet (CS); PP films under UCW, UFW, SB and SEQ stretching, respectively

During stretching, stress-strain curves are recorded, which are presented in Fig. 6. As the film was stretched sequentially in MD and TD, two groups of curves (Fig. 6d₁ and Fig. 6d₂) were obtained for SEQ stretching. Under UCW stretching (Fig. 6a), the stress in MD increases almost linearly with a draw in the first stage and then reaches the yield point. After exceeding the yield point, the evolution of stress undergoes at first declination and then a plateau-like region. Interestingly, stress in TD increases first, and then reaches a plateau, which originates from the film shrinkage. While for UFW stretching (Fig. 6b), the stress in MD experiences a yield point followed by weak strain hardening-typical phenomenon of semi-crystalline polymers in tensile experiments. The differences between the two uniaxial stretching modes are: after the yield point, the stress in MD experiences a plateau-like evolution for UCW stretching while the stress shows weak strain hardening behavior for UFW stretching. Due to constraint in TD, molecular chains are no longer primarily oriented in the MD but tend towards TD, similar to biaxial orientation. While for UFW stretching, molecular chains are mainly oriented

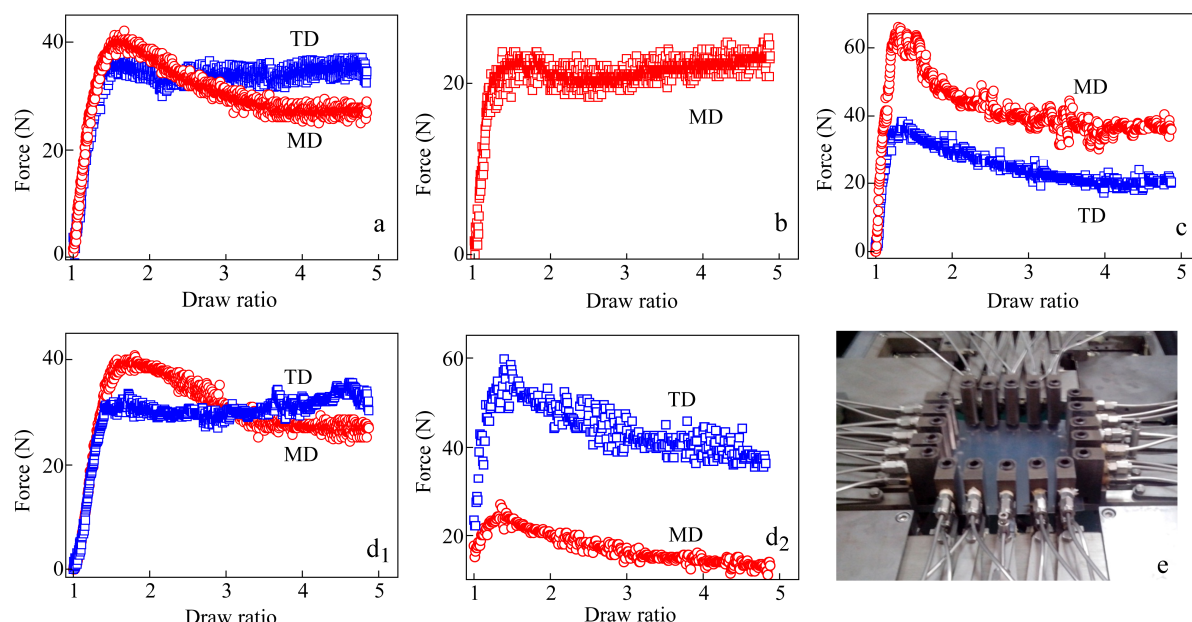


Fig. 6 Stress-strain curves recorded during UCW stretching (a), UFW stretching (b), SB stretching (c), first step (d₁) and second step (d₂) of SEQ stretching, respectively; (e) Picture of PP casting sheet gripped by pneumatic clamps before stretching

towards MD, resulting in higher degrees of orientation^[18]. Figure 6(c) gives the stress evolution of both directions under simultaneous biaxial mode. They both show the linear increase-yield point-slight decrease-plateau behavior similar to the stress in MD under UCW mode. No strain hardening phenomenon is observed as the rate of orientation is at a competitive disadvantage with rate of chain relaxation under low stretching speed. Also, during SB stretching process, molecular chains randomly orient in film plane and efficiency of orientation towards one direction is not so high as uniaxial stretching. The higher stress level in MD than in TD is attributed to the pre-orientation of molecular chains of casting sheets towards MD. During SEQ stretching, the films are first stretched in MD in UCW mode, then it is stretched in the perpendicular TD also in UCW mode. As a consequence, the stress-strain curves obtained in the first step are the same with UCW stretching. During transverse stretching, evolution of stress starts from a certain level and achieves higher value. During first-step, a well defined crystalline phase oriented in MD, similar to UCW stretching. These crystallites serve as physical cross-linking points, the transverse stretching becomes very efficient in transmitting lateral forces deep into the structure including all crystallites, resulting in higher stress level^[12].

More experiments with different draw ratios and stretching speeds were also conducted to study the influence of processing conditions on structure of PP films (data are not shown here for brevity). Optical properties (haze, gloss) and mechanical properties (Young's modulus, tensile strength, elongation at break) of the films will be further explored, providing an approach to establish the relationship of processing/structure/property. The testing experiments demonstrate that the biaxial stretching equipment is capable to study the influence of processing conditions on film structure, exploring the film stretching process in industry effectively.

CONCLUSIONS

A universal biaxial stretching equipment is designed and constructed, which allows the measurement of strain-stress in both directions during stretching. All parameters related to film manufacturing, such as deformation modes, stretching temperature, drawing speed and strain rate profile can be set accurately and independently. In addition, the technical indexes that the equipment can achieve are comparable to those of the commercial stretching unit. Using the equipment, stretching measurements of PP casting sheets were conducted to investigate the influence of stretching conditions on structure of films. SAXS results show that oriented lamellae are formed under UCW and UFW stretching, while no preferred oriented structures are formed after SB and SEQ stretching. DSC tests indicate that stretching can increase crystallinity and melting point of films comparing to casting sheets. The equipment provides an effective method to get a deeper understanding of film stretching in industry.

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