The Evaluation of Biaxial Stretchability of Polypropylene Films Using a Newly Developed Test Machine

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Abstract. The evaluation method for a biaxially oriented film was developed using in-situ measurement during the stretching process. It could obtain basic data such as stress-strain curve, birefringence, light scattering, three-dimensional refractive indexes and birefringence distribution as functions of time, temperature and percentage in elongation, the information of superstructure could be obtained by measuring the birefringence and light scattering during the biaxial stretching process with a small piece of polymer sample. From the experimental results, stretchability, thickness uniformity and spherulite size could be evaluated simultaneously. Furthermore, as the thickness uniformity is closely related to the standard deviation of birefringence distribution, the thickness uniformity can be also evaluated by the retardation distribution

Keywords: biaxial stretching; in-situ measuring; birefringence distribution; light scattering; three-dimensional refractive indexes.

INTRODUCTION

The biaxial stretching films are widely used for both industrial products and our daily necessaries. Recently, the films with thin and uniform thickness in the production of stretching films are required from the reduction of raw materials and high quality films. In the production lines of the stretching films, one needs to understand not only the stretchable resin property, but also the optimum stretching conditions for the film resin. Besides, the evaluation of stretchability of the films becomes important in the development of the stretching film. The stress-strain curve and crystallization of crystalline polymer are keypoints for evaluation of stretchability during the biaxial stretching. The stress-strain behavior of the biaxial stretched film has been examined[1]. The relationship between the stretching force and stretching ratio of various samples was studied[2]. However, the relationship of both the stresses and retardation behavior as a function of the stretching ratio during the stretching process and the deformation of spherulite and stretching ratio have not been shown. The crystallization behavior and the higher order structure formation of crystalline polymer have been measured by the light scattering, but the deformation of spherulite by the stresses during the stretching process has not yet been done.

Moreover, the simultaneous measurement of stresses, birefringence and three dimensional molecular orientations are also essential for evaluation of stretchability. However, the simultaneous measurement data for the stresses, birefringence, spherulite and three dimensional molecular orientations during the biaxial stretching process have been not shown.

On the other hand, the in-situ measurement of the stress and retardation during the uniaxial stretching by using Photo Elastic Modulator(PEM) was previously proposed[3]. This method is superior in terms of high sensitivity and short measurement time. With the advantages of PEM, the stress and retardation in simultaneous measurement during the uniaxial stretching process can be measured even if the stretching speed is fast[1].

Above all, there are no reports of the simultaneous measurement of stress, birefringence, three dimensional refractive indexes and light scattering during the biaxial stretching process. In this paper, we describe on the results of crystalline polypropylene sample during the sequential biaxial stretching process obtained by a newly developed test machine.

SYSTEM CONSTRUCTION

FIGURE 1 illustrates the schematic diagram for the biaxial stretching test machine. The apparatus was constructed to carry out the simultaneous and the in-situ measurement of stress, birefringence, three dimensional refractive indexes and light scattering during the biaxial stretching process. The test machine was equipped with stretching unit and XY mapping controlled and driven by a computer, two load cells for the measurement of stresses of the biaxial stretching film, double birefringence measurement system using Photo-Elastic Modulator (PEM) for the measurement of retardations: the vertical and inclined incidence angle of laser beams respectively, and three dimensional molecular orientations of the biaxial stretching film, and the light scattering system for monitoring the spherulite conformation and spherulite size. The light source axis on the double birefringence and light scattering measurement system was adjusted to overlap in the center of the biaxial stretching film on the stretching unit.

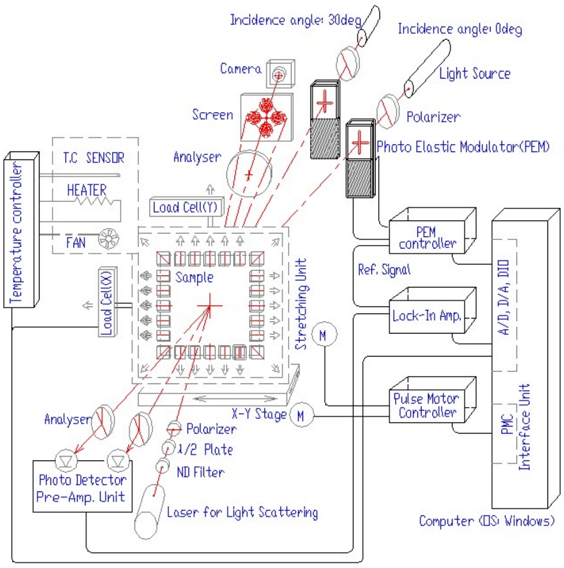

FIGURE 1 A diagram of a newly developed test machine

SAMPLE

Polypropylene is one of crystalline polymers which have high performance such as high formability, heat-resistance, high transparency and cost performance, so polypropylene film is widely used for food packaging and industrial films. Polypropylene is also the crystalline polymer which has highly ordered arrangement of molecules structure.

The resin of iPP sample, having MFR=3g/10min, Mw=3.6×105g/mol, Mw/Mn=5.0 and Tm=160℃, was produced by Prime Polymer(Japan). The non-stretching sheet with 500 micrometers as a film thickness was cut to size at 85 square millimeter of piece in use. The stretching conditions were fixed with the chamber temperature at 162℃, stretching speed at 25 mm/sec and preheating time at 120 sec before the biaxial stretching.

EXPERIMENTAL RESULTS AND DISCUSSION

Measurement of stresses and retardations

The stresses and retardations were simultaneously measured during the sequential biaxial stretching with stretching ratio at MD5×TD6.7 times as shown in FIGURE 2(a). As the results, the MD stress(blue line) and TD stress(pink line) are highly increasing in the beginning of MD stretching, and then they changed the increasing behavior where the MD yield point appears due to the neck-like deformation occurs during the MD stretching process. In the beginning of TD stretching, TD stress is dramatically increasing due to the neck-like occurs during the TD stretching process. Finally, MD and TD stresses are increasing again for the even-stretching in the last of TD stretching. The retardations of vertical incidence and inclined incidence angle of laser beams show the increasing in MD stretching and the decreasing in TD stretching process due to the sequential biaxial stretching. The variation of retardation in TD stretching is greater than the variation of retardation in MD stretching process at the middle stage of TD stretching. It means that the polypropylene has a high molecular orientation at the end of the sequential biaxial stretching.

Refractive indexes behavior during sequential biaxial stretching process

The refractive indexes which are information of molecular orientation were calculated from the results of retardations shown in FIGURE 2(b). Here, n_x and n_y are refractive indexes of MD and TD stretching direction, and n_z is refractive index of film thickness direction. The relationship between refractive indexes and stretching ratio shown: In the MD stretching, n_x is increasing due to the increasing of MD stress, and n_z is decreasing due to the decreasing molecular orientation in the film thickness direction. Next, in the TD stretching, n_y is increasing due to the increasing of MD stress, and n_z is still continuously decreasing due to the decreasing of film thickness during TD stretching process.

From the results, it found that the degree of n_y became higher than n_x where the variation of retardation in TD stretching is larger than in MD stretching. As well as from the increasing behavior in difference value of n_x and n_y, it was found that the stretched film is in a plane orientation according to the stresses and the sequential biaxial stretching ratio.

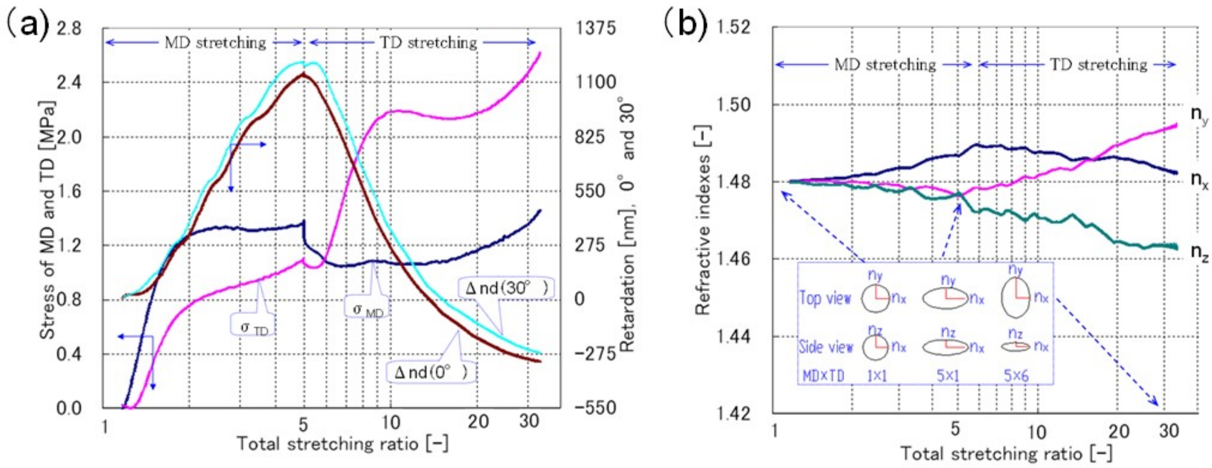

FIGURE 2

(a) Simultaneous measurements of stresses and retardations during the sequential biaxial stretching process

(b) Three dimensional refractive indexes behavior during the sequential biaxial stretching process

Spherulite deformation of stretched film at each sequential biaxial stretching ratio

The light scattering image of the stretched film was observed at each sequential biaxial stretching ratio shown in FIGURE 3(a). As the results of light scattering images as four-leaf clover pattern, it was found that the spherulite was deformed by the increasing of the sequential biaxial stretching ratio. And it was also obtained that the spherulite was dramatically deformed in the beginning of MD stretching region, and then it was starting to break up at MD2×TD1 stretched ratio where the MD yield point appears. Lastly, it was already disappeared at MD5×TD3 stretched ratio where the TD neck-like deformation finished before the TD stress increases again in the last of TD stretching.

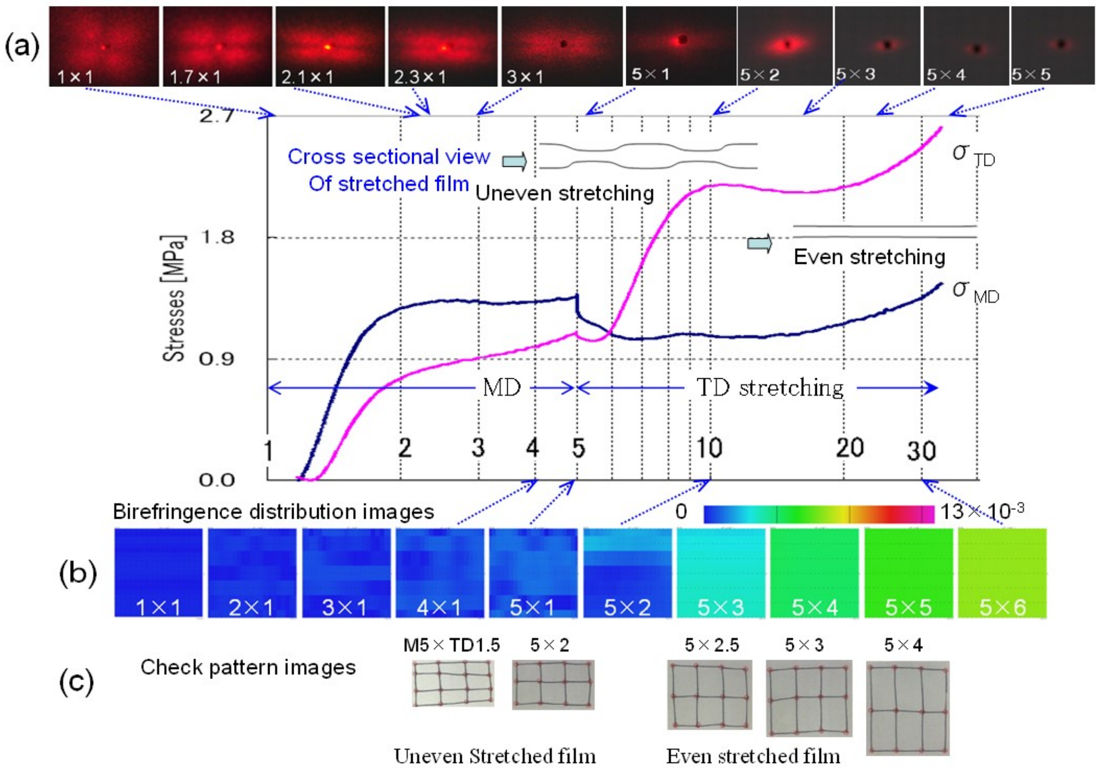

FIGURE 3

(a) Light scattering images at several stretching ratio of sequential biaxial stretching process

(b) Birefringence distribution images at several stretching ratio of sequential biaxial stretching process

(c) Stretched film pattern images at several TD stretching ratio

Birefringence distribution of stretched film at each sequential biaxial stretching ratio

The birefringence distributions at the center area of the stretched film were measured by XY mapping controlled system, and the standard deviation (SD) of birefringence distribution was calculated as shown in FIGURE 3(b).

As the results, it was found that the birefringence distribution images showed unevenly distributed results due to the neck-like until the spherulite had already disappeared at the stretched ratio MD5×TD3, and they showed good distribution from the stretching ratio where the MD and TD stresses were increasing behavior again in the last stage of TD stretching.

Neck-like deformation during sequential biaxial stretching process

Furthermore, the 5 mm^2 check pattern was drawn visually to check the stretched film pattern at several stretching ratios as shown in FIGURE 3(c). From the results of check pattern images, the uneven stretching occurred in the beginning of TD stretching (5×1.5 times) and then finally the stretching pattern became evenly after the TD yield point appeared at the stretching ratio where MD and TD stresses increased again in the last of TD stretching. It means that the stretched film had the neck-like stretching during the sequential biaxial stretching before the even stretching in the last stage of TD stretching.

Relationship between SD of birefringence distribution and thickness uniformity

Furthermore, in order to investigate the thickness variation at each sequential stretching ratio, the thickness average at 150 mm^2 of the stretched films was measured by dial gauge (Mitsutoyo Teclock PG-01J), and then the thickness uniformity could be calculated from equation (1) as follows.

$$\text{Thickness uniformity}[\%] = (\text{maximum thickness} - \text{minimum thickness}) / (2\times\text{average thickness})\times 100 \qquad (1)$$

The results of thickness uniformity and standard deviation of birefringence distribution of the stretched film at several TD stretching ratios are shown in TABLE 1 and FIGURE 4. As the results, it was found that the standard deviation of birefringence distribution and the thickness uniformity are improved with the increasing of sequential biaxial stretching ratio. It means that they are closely correlated in the TD stretching process.

TABLE 1 Results of standard deviation of birefringence distribution and thickness uniformity at several TD stretching ratio

Sequential stretching ratio	5×2	5×2.5	5×3	5×4	5×5	5×6
SD of birefringence distribution($\times10^{-3}$)	0.544	0.426	0.195	0.042	0.021	0.018
Thickness uniformity [%]	60.7	48.1	29.7	18.4	12.7	11.9

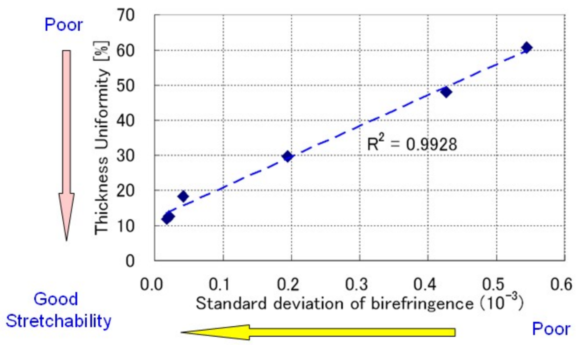

FIGURE 4 Relationship between standard deviation of birefringence distribution and thickness uniformity

CONCLUSIONS

We can summarize the stretchability of crystalline polypropylene film during the sequential stretching process obtained by a newly developed test machine and make conclusions as follows:

1. A newly developed test machine can simultaneously measure the data for evaluation of stretchability.
2. The even stretching was begun at MD5×TD3 where the spherulite was broken up and the neck-like stretching finished, so the thickness uniformity and the SD of birefringence became good.
3. In order to obtain a good stretchability of the stretched film, the proper stretching ratio should be above MD5×TD5.
4. As the thickness uniformity is closely related to the SD of birefringence distribution, the thickness uniformity can be easily evaluated by the retardation distribution.

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