THE EVALUATION OF STRETCHABILITY OF POLYPROPYLENE AND POLYSTYRENE FILMS USING A NEWLY DEVELOPED TESTING MACHINE

EGOSHI KENTARO[*1,3], KANAI TOSHITAKA[2] AND TAMURA KAZUHIRO[3]

[*1] *ETO Co., Ltd. 3403-1 Shake, Ebina city, Kanagawa, 243-0424 Japan*
[*2] *KT Poymer, 2411-3 Kuranamidai, Sodegaura, Chiba, 299-0243, Japan*
[*3] *Kanazawa Univerity,Kakuma-machi, Kanazawa, Ishikawa, 920-1192 Japan*

ABSTRACT

The evaluation method of the stretchability for a biaxially oriented film during the stretching process was newly developed using in-situ measurement test machine. Stress, retardation, three dimensional refractive indexes, light scattering image and birefringence distribution of films can be obtained in a short time. This stretching test machine was applied to both crystalline polymer such as polypropylene and non-crystalline polymer such as polystyrene.

As results of the measurements, the stress of crystalline polymer increased with increasing the preheating time. One the other hand, the stress of non-crystalline polymer did not increase with increasing the preheating time. It means that crystalline polymer is required to set up the optimum stretching condition such as preheating time for good biaxial oriented film. Furthermore, the birefringence distribution and the thickness uniformity of stretched film were measured. It was found that the stretchability could be evaluated with a small amount of sample using the biaxial stretching test machine.

Key words: biaxial stretching process, preheating time, stretchability, birefringence distribution, thickness uniformity

1. Introduction

The biaxial stretching films are widely used for both industrial products and our daily necessaries package. Recently, the films with thin and uniform thickness in the production of stretching films are required from the reduction of raw materials. In the production lines of 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, retardation-strain curve, three dimensional molecular orientation and crystallization of crystalline polymer are keypoints for evaluation of stretchability during the biaxial stretching[1]. Previous researches studied the relationship between the stretching force and stretching ratio for various samples[2-4]. However, the relationship among the stresses, the deformation of spherulite and retardation behavior as a function of the stretching ratio during the biaxial stretching process were not reported.

Moreover, the simultaneous measurement of stresses, birefringence and three dimensional molecular orientations are also essential for the evaluation of stretchability. Several kinds of retardation measurement methods have been developed[5,6]. However, the simultaneous measurement data for the stresses, birefringence and three dimensional molecular orientation during the biaxial stretching process were not reported.

On the other hand, the in-situ measurement of the stress and retardation during the uniaxial stretching using Photo Elastic Modulator (PEM) was previously proposed[7]. This method is superior in terms of high sensitivity and short measurement time. With the advantages of PEM, the stress and retardation in the 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. Moreover, there is also no investigation of the relationship between the stress and retardation behavior during the biaxial stretching process. As well as, there are no reports of the stretchability of crystalline polymer and non-crystalline polymer under the stretching condition such as the preheating time.

In this paper, we describe results of crystalline polypropylene sample and non-crystalline polystyrene sample during the biaxial stretching process obtained by a newly developed test machine.

2. System construction

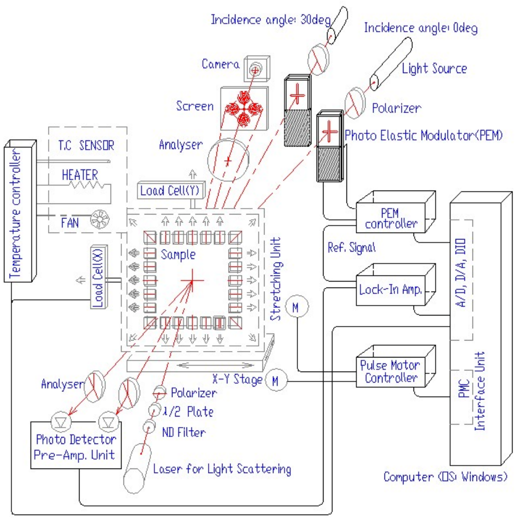

Fig.1: System construction

Fig. 1 illustrates the schematic diagram for the biaxial stretching test machine. The apparatus was designed and 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, 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, and three dimensional molecular orientation of biaxial stretching film in vertical and inclined incidence of laser beam, and the light scattering system for monitoring the spherulite conformation and stero-crystal size. The light source axis on the double birefringence and light scattering measurement system was adjusted to overlap in a center of the biaxial stretching film on the stretching unit.

3. Sample

3.1.1 Crystalline Sample

Polypropylene is one of crystalline polymers which has high performance such as high formability, heat-resisting property, high transparency and cost performance, so polypropylene film is widely used for food packaging and industrial films.

3.1.2 Non-Crystalline Sample

Polystyrene is one of non-crystalline polymers which has high performance such as high transparency, high hardness, high formability and cost performance, so polystyrene is widely used for electrolic products, food container and display case.

3.1.3 Characterictics of resin

The characteristics of polypropylene(PP) and polystyrene(PS) are shown in Table1

Table 1: Characteristics of resins

Sample	MFR g/10min	Mw×10^5 g/mol	Mw/Mn [-]	Tm ℃
PP	3	3.6	5.0	160
PS	4	-	-	-

3.2 Experimental conditions

Polypropylene sample: The non-stretching sheet with 500 micrometers as a film thickness was cut to size at 85 square milimeter 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 90, 120, 150, 180 sec before stretching.

Polystyrene sample: The non-stretching sheet with 1200 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 124℃, stretching speed at 25 mm/sec, and preheating time at 90, 120, 180, 240 sec before stretching

4. Results and discussion

4.1 Polypropylene sample

4.1.1 Simultaneous measurement of stresses and retardations during sequential biaxial stretching

Fig.2A showed the results of the MD stress behavior and Fig.2B showed the TD stress behavior during the sequential biaxial stretching. It was found that the MD and TD stresses differently increased with increasing the preheating time.

The spherulite of crystalline polymer was observed as four leaf clover on the screen of light scattering system. As the results of light scattering, the spherulite was deformed by increasing stresses and then it disappeared at MD5×TD3.

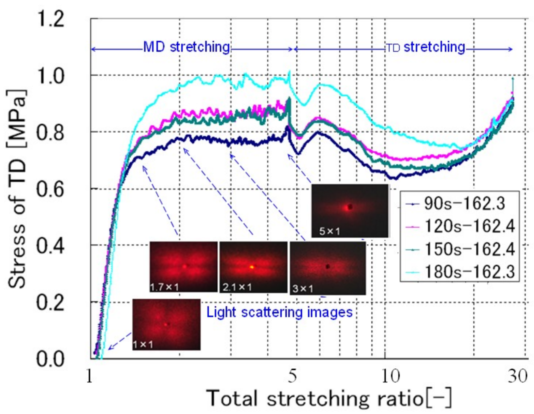

Fig.2A: MD stress at several preheating times

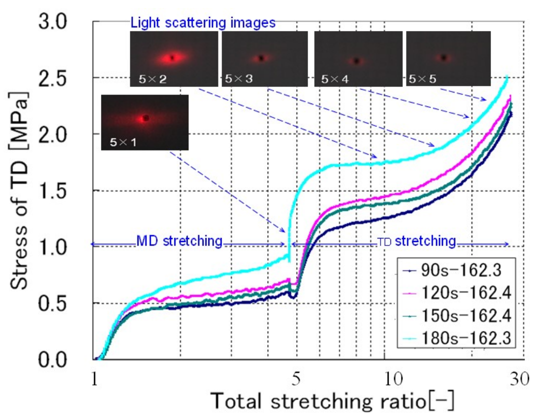

Fig.2B: TD stress at several preheating times

Fig.2C showed the results of the vertical incidence angle(0°) of retardation, and Fig.2D showed the results of the inclined incidence angle(30°) of retardation during the sequential stretching process. The retardation showed the increase in MD and the decrease in TD during the biaxial stretching process. It was found that the vertical and the inclind incidence of retardations also increased with increasing the preheating time.

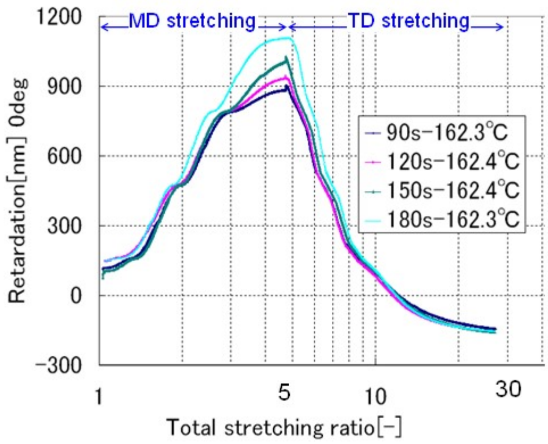

Fig.2C: Vertical incidence angle of retardation at several preheating times

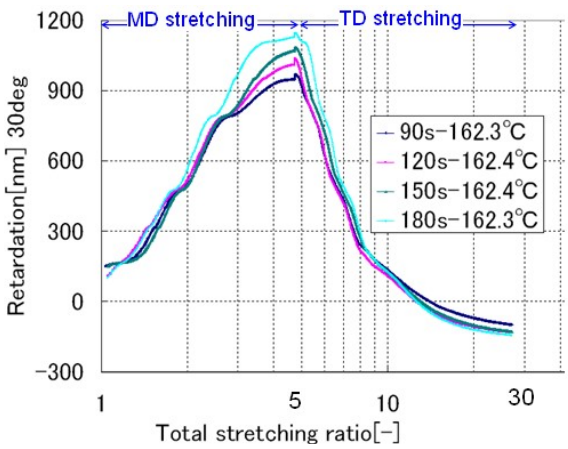

Fig.2D: Inclined incidence angle of retardation at several preheating times

Futhermore, the refractive indexes can be calculated from the results of retardations which were measured by the double birefringence measurement sytem as shown in Fig.3A(120sec) and Fig.3B(180sec). It was found that the degree of plane orientation at 120sec of preheating time is smaller than at 180sec. It means that the preheating time at 120sec gives easier stretching than at 180sec.

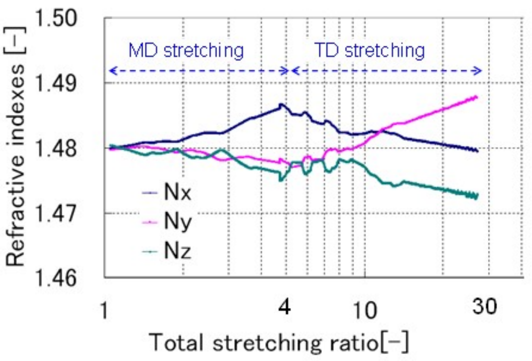

Fig.3A: Refractive indexes at 120sec of preheating time

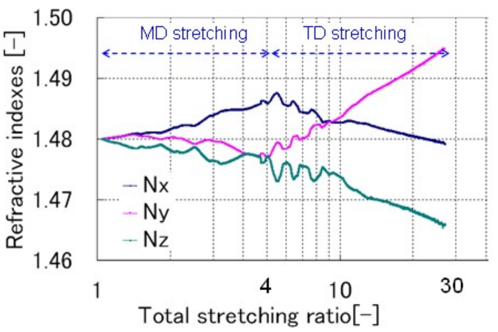

Fig.3B: Refractive indexes at 180sec of preheating time

4.1.2 Retardation distribution of stretched film

The retardation distribution at the center area of stretched film were easily measured by XY mapping controlled system as shown in Fig.4A, and the standard deviation (SD) of retardation distribution was calculated as shown in Fig.4B. It was found that the retardation distribution shows the difference in color all over the image. Nonuniform retardation distributions in color all over the image were shown at a short preheating time and long preheating time. On the contrary, more uniform distribution was shown at 120sec of preheating time which means more uniform retardation distribution. It means that it has the optimum preating time condition for the better uniformity of biaxial stretching film.

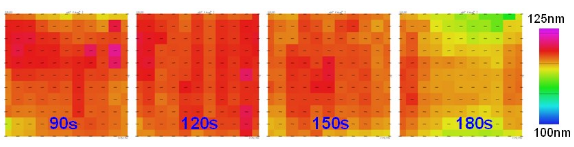

Fig.4A: Retardation distribution of stretched film at several preheating times

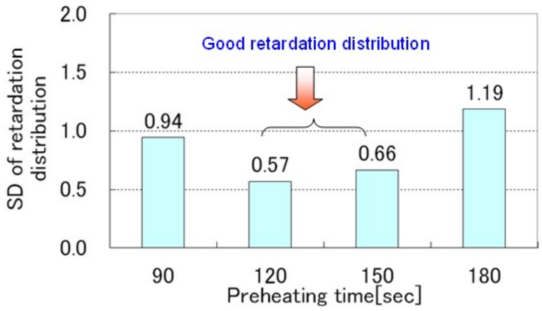

Fig.4B: SD of retardation distribution of stretched film at several preheating times

4.1.3 The thickness uniformity of stretched film

The film thickness at the center area of stretched film was measured by dial gauge, Mitsutoyo Teclock PG-01J. The thickness uniformity calculated by equation 1 was shown in Fig.5. It was found that the thickness uniformities showed bad values in the short and in the long preheating time such as 90sec and 180sec, and then they showed good values at the middle of preheating time such as 120sec to 150sec. It means that the stretching under the optimum preheating time condition is better for thickness uniformity of stretched film.

$$\text{Thickness uniformity [\%]} = (\text{max-min})/(2\times\text{ave})\times 100 \qquad (1)$$

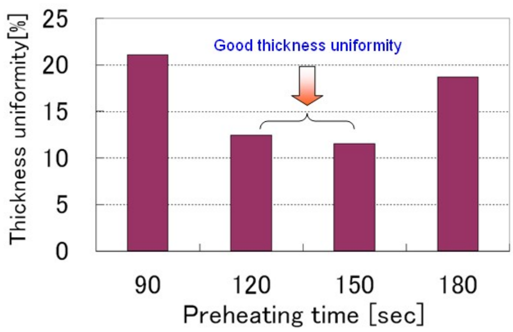

Fig.5: Thickness uniformity of stretched film at several preheating times

The results of the standard deviation of retardation distribution are shown in Fig.4B and the results of thickness uniformity are shown in Fig.5. It was found that the small standard deviation of retardation distribution is good thickness uniformity of biaxial stretched film. In other word, they are closely related to each other, and then the thickness uniformity can be easily evaluated by the measurement of retardation distribution.

4.1.4 DSC measurement of non-stretch film

The preheated non-stretch sample was heated from -20℃ to 220℃ at 20℃ per minute for DSC measurement of several preheating time conditions as shown in Fig.6. It was found that the low melting temperature component decreased, and the endothermic peak moved to high temperature and the latent heat of fusion increased with increasing the preheating time. It means that the increasing of preheating time caused the crystallinity and the melting temperature become higher.

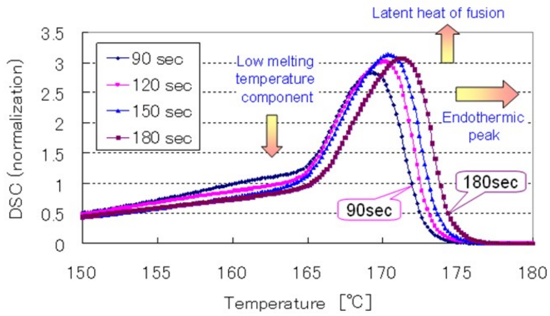

Fig.6: DSC measurement of non-stretch film at several preheating times.

4.2 Polystyrene sample

The stresses and retardation of polystyrene sample can simultaneously measure during the sequential biaxial stretching process under several preheating time conditions.

Fig.7A showed the results of the MD stress and Fig.7B showed the TD stress behavior. Fig.7C showed the vertical incidence(0°) of retardation behavior and Fig.7D showed the inclined incidence(30°) of retardation behavior during the sequential biaxial stretching process. It was found that all of preheated samples showed the same stresses behaviors, and the same retardation behaviors at the several different preheating time conditions. It means that the stresses and retardations of polystyrene are not much influenced with increasing preheating time condition during the sequential biaxial stretching process.

As the results shown in Fig.7A, the spherulite can not be observed as four leaf clover on the screen of light scattering system. It means that non-crystalline polymer does not have spherulites like as crystalline polymer which has spherulites as shown in Fig.2A.

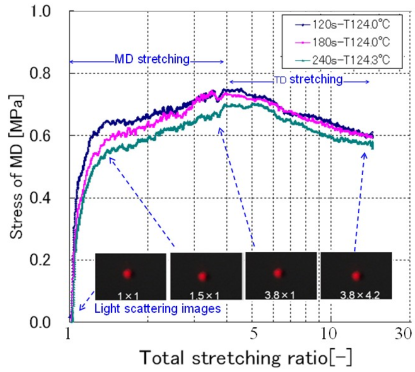

Fig.7A: MD stress of polystyrene at several preheating times

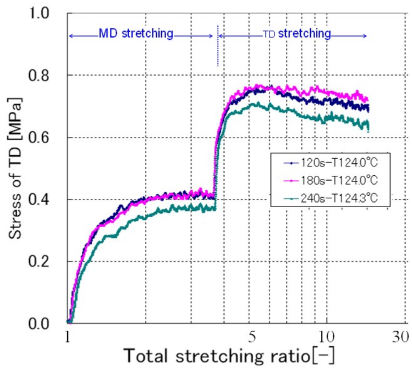

Fig.7B: TD stress of polystyrene at several preheating times

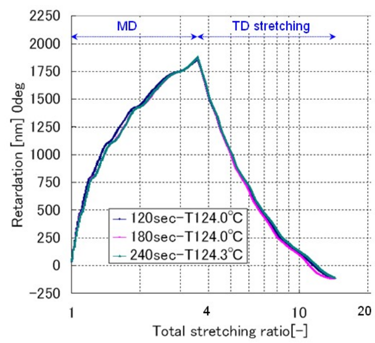

Fig.7C: Vertical incidence of retardation at several preheating times

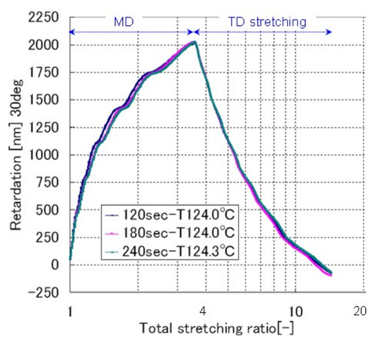

Fig.7D: Inclined incidence of retardation at several preheating times

4.3 Schematic crystalline structure

The Fig.8A explained the changing of polypropylene crystalline lamella under the increasing of preheating time condition. The crystalline lamella became thick and amorphous region get thinner due to the increasing of preheating time, that cause the endothermic peak moved to high temperature as shown in Fig.6, and the increasing of stresses as shown in Fig.2A.

On the other hand, as expressed in Fig.8B non-crystalline sample does not have an ordered structure like as crystalline sample which has an ordered structure, so that the stresses and retardations do not increase with increasing the preheating time.

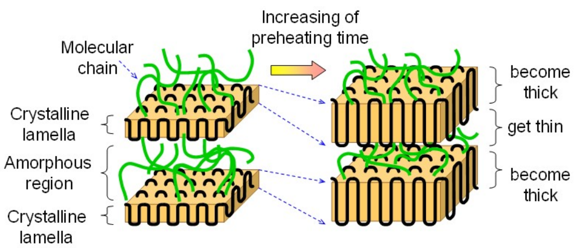

Fig.8A Crystalline structure of crystalline polymer under the increasing of preheating time

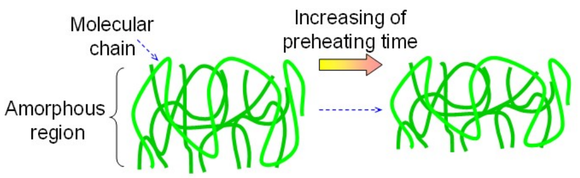

Fig.8B Amorphous region of non-crystalline polymer under the increasing of preheating time

5. Conclusion

1) A newly developed test machine can simultaneously measure the data for evaluation of stretchability.

2) Crystalline sample

The stretchability of crystalline polypropylene sample was influenced by the increasing of preheating time. In order to obtain a good stretchability of crystalline film, the production under the optimum preheating time condition is very important.

3) Non-crystalline sample

The stretchability of polystyrene is not much influenced by the increasing of preheating time.

4) As the thickness uniformity is closely related to the standard deviation of retardation distribution, the thickness uniformity can be easily evaluated by the measurement of retardation distribution.

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