Bowing Phenomenon in Double Bubble Tubular Film Processing for PA 6

This report is to discuss the bowing phenomenon and the sag elimination tension for biaxial oriented PA 6 film process, which consists of double bubble tubular film process and the thermosetting tentering.

The bowing phenomenon does not occur in the heat set of the triple bubble tubular process. However, there is a serious problem of the bubble stability when the sufficient thermosetting by the tubular process is carried out, so the heat set by the tenter process is required. The bowing phenomenon arises when the annealing is carried out by the tenter method. The bowing distortion becomes large when the annealing temperature is high and film relaxation ratio is large.

It is possible to reduce the bowing distortion when thermosetting is carried out in a two-step tenter process. The shrinkage stress of the stretched film was relaxed in the heat treatment during the second step whose temperature is higher than the first one. A two-step heat treatment was able to decrease the bowing distortion compared with the one-step heat treatment.

When the bowing phenomenon occurred, shrinkage percentage in hot water changes along the cross-direction of a film. The humidity expansion coefficient in the film which increases as the increasing shrinkage percentage in hot water is related to the molecular orientation. The sag also increased with increasing the bowing distortion. The bowing distortion and the sag elimination of tension have proportional connection.

1 Introduction

The plastic film is used in the various applications and the requirement for the film characteristics becomes severe year by year in proportion to the diversification of applications. Therefore, the advanced production technology of plastic films is needed. Especially, further upgrading of the technology is required for the production of PA 6 film in order to demonstrate the features of PA 6. The research on biaxially oriented films remains fundamental and basic, which is not applicable to the commercial production.

Serious technical problems to be solved in the stretching of film are the bowing phenomenon, which causes the non-uniformity of film characteristics in the transverse direction, and the thickness unevenness of film, which causes the planar waviness of film and the wrinkle in the winding of film.

The bowing phenomenon relates to the discrepancy between the direction of the molecular orientation axis at the center and the direction at the edge of film width, which causes the anisotropy of film characteristics such as the heat shrinkage, the refractive index and mechanical properties. The difference between the direction of the molecular orientation axis and the line direction becomes large towards the edge of film in the film width. So, when customers require a film without the bowing in the property, film manufacturers usually have to supply only the mid-section of the film width. Reducing the bowing is one of the most important technical issues in the manufacturing of biaxially oriented film.

Papers on the bowing phenomenon have been published by Kase et al. [1 to 4] and Sakamoto [5 to 7]. Kase et al. theoretically analyzed the deformation behavior of thin uneven elastic film in the tenter process by using the finite difference method. Sakamoto investigated the changes of the refractive index ellipsoid of the biaxially oriented PET film, which was conducted by a laboratory batch drawing. But as far as the bowing phenomena are concerned, there is no proof that such a batch drawing is equivalent to industrial tenter stretching. He also reported the theoretical analysis of the refractive index ellipsoid due to bowing which occurs in the industrial tenter process. However, these theoretical predictions have not been confirmed experimentally. Previous studies done by the authors were concerned about observations of bowing phenomenon throughout the tenter process. Their analyses were used finite element method calculation discussing the countermeasure for reducing bowing distortion. But these papers are not systematically described.

Afterwards, the research using a biaxial tenter stretching production machine on polyester film was carried out by Yamada et al. [8 to 13]. The bowing behavior in the equipment was quantitatively analyzed. Reduction the bowing distortion has been proposed on the basis of these results.

However, research of heat set technology on the PA 6 film was not carried out in detail. Research of the tenter method biaxial stretching technology has been reported, and research on the double bubble tubular biaxial technology has not been
made at this time. In addition, annealing heat setting technology in double bubble tubular process will be studied this time.

In order to commercialize biaxial stretching PA 6 film for application such as the retorting reliable thermosetting is necessary. Because of the above reasons, it is necessary to carry out the thermosetting using the tenter equipment. The bowing phenomenon arises as a destiny, when the thermosetting is carried out using the tenter equipment.

After the tubular biaxial stretching, the reduction of the bowing phenomenon in the tenter thermosetting is required. This is the main purpose in this study.

The effect of the PA 6 resin, peculiar moisture absorption characteristics, was examined. It is a problem that biaxial stretched PA 6 film absorbs moisture. When moisture is absorbed, sag is caused. Sag causes the trouble in printing and in lamination. The relation with sag, this became the disturbance in the secondary working, was also evaluated. Therefore, it is important to reduce the bowing phenomenon, to minimize sag, and to control these factors, which should be examined in detail.

2 Experimental

2.1 Equipments

Apparatus used for the double bubble tubular film process is shown in Fig. 1. Using an extruder (L/D = 24) with the diameter of 40 mm equipped with a circular die with the diameter of 75 mm and the lip clearance of 1 mm and with a water-cooling ring having the diameter of 90 mm non-stretched film was produced at a resin temperature 265 °C and blow-up ratio of 1.2.

This raw film was stretched simultaneously in the machine and transverse directions by using inside bubble air, a drawing machine composed of two pairs of pinch rolls and a heating furnace (a far infrared radiation heater is self-contained).

The stretched film was heat-set using a heat treatment device. The evaluation was carried out using two kinds of thermosetting equipments this time. They are triple bubble thermosetting method and tenter thermosetting method. Apparatus used for the triple bubble tubular thermosetting process is shown in Fig. 2. The comparative evaluation of one-step thermosetting and two-step thermosetting was carried out on the tenter thermosetting method.

2.2 Material

The material used was Ube 1024FD15 (PA 6) with mean molecular weight of 24000 and the relative viscosity of \( \eta_r = 3.75 \) in 98 % sulfuric acid as a solvent.

2.3 Definition of Bowing Distortion

Bowing phenomena has two meanings. One is geometrical bowing, in which a straight line drawn across the film width at

![Fig. 1. Schematic of double bubble tubular film process, top: tenter one-step annealing process, bottom: tenter two-step annealing process](https://example.com/fig1.png)
the entrance of a tenter changes into bow shape at the exit of the tenter. The second is the characteristic bowing which means the anisotropic main axis of film characteristics such as heat shrinkage, optical refraction index, and mechanical properties is varied along in the film width. In this paper, the geometrical bowing is used under the assumption that it corresponds to the characteristic bowing.

As shown in Fig. 3, bowing distortion (B) is expressed as the percentage of bowing distance \( \Delta B \) divided by the film width \( L \). When the film center lags behind the film edge, the bowing distance \( \Delta B \) is considered to be positive.

\[
B = \frac{\Delta B}{L} \times 100 \%.
\] (1)

### 2.4 Experimental Method

The process conditions of non-stretched film is 265 °C for resin temperature at the die exit, 1.2 for blow up ratio, and 6.0 for draw down ratio respectively. Film was quenched in water at 18 °C to lower crystallinity. The stretching device consists of a heating/stretching furnace and an air ring. The air ring, which injects air downward at an angle of 45°, was installed at the upper part of the heating furnace. The tenter annealing process in the one-step is used.

The standard condition for the tenter process was set at 200 °C for thermosetting temperature (hot air temperature) and 10 % for relaxation ratio respectively. It is the mechanism, which perpendicularly blows hot air from top and bottom holding the double bubble end division in the clip. To hold the double end division in the clip, for holding the contraction, the bowing distortion were evaluated by the change of thermosetting temperatures and film relaxation ratio:

- annealing temperature (180, 190, 200, 210, 220 °C),
- relaxation ratio (0, 10, 20, 30 %).

The tenter thermosetting process in two-step, the standard condition for tenter process in the first stage was set at 170 °C for thermosetting temperature (hot air temperature) and 0 % for relaxation ratio respectively. The standard condition for tenter process in second stage was set at 200 °C for thermosetting temperature (hot air temperature) and 10 % for relaxation ratio respectively. Many straight lines across the non-stretched film width were drawn on the surface of the film at the entrance of the stretching furnace. After film winding, a line sample was collected. The evaluation procedure for bowing phenomenon was followed by Fig. 3. The bowing distortion was calculated by Eq. 1.

### 2.5 Shrinkage Percentage and Density

In PA 6 film for retort food, it is soaked in hot water of high temperature in order to conduct the sterilization and processing. It is necessary to hold the shrinkage percentage of a film small by thermosetting. The relationship among thermosetting temperature, film shrinkage percentage and film density was examined.
2.6 Shrinkage Percentage and Orientation In-plane

By changing heat set and cross-direction position, the value of the shrinkage percentage pattern in plane was measured. By measuring the length of the marked line under the 23°C, 50% relative humidity, after a day of drying, as a conditioning after the shrinkage percentage pulled the table line in each direction in the film; in plane every 100 mm and after it spread in the hot water of 120°C condition for 30 minutes, the shrinkage percentage was calculated.

Molecular orientation condition of the film in plane was evaluated using the ultrasonic wave orientation measurement equipment. This equipment can evaluate the in plane orientation using the elastic modulus proportional for the square of conduction velocity of ultrasonic wave.

2.7 Sag Elimination Tension

Using the slitter machine, sag elimination tension was measured. Slit samples of 800 mm width was collected by the change of the thermosetting temperature, and each sag elimination tension was evaluated using the slitter machine. The relationship between bowing distortion and sag elimination tension was evaluated.

2.8 Film Humidity Expansion

Using the humidity expansion measuring instrument (Nihon sinkuu), the relationship between film position and humidity expansion coefficient of a film was quantitatively evaluated. The humidity expansion coefficient should be obtained by the sampling of the film position of diagonal 45°. The relative humidity was made to rise from 20 % RH to 80 % RH, and the humidity expansion coefficient was calculated by the measurement of the elongation percentage of a film.

3 Results and Discussion

3.1 Heat Set Temperature and Relaxation Ratio

The relationship between shrinkage percentage and thermosetting temperature is plotted in Fig. 4. The shrinkage percentage of a film is reduced with increasing thermosetting temperature. The shrinkage percentage shows the high value, as the temperature of hot water is higher. The relationship between film density and thermosetting temperature is plotted in Fig. 5.

The density of a film increases with a rise in the annealing temperature. The crystallinity increased with increasing temperature. It is necessary to reduce to less than 5% the shrinkage percentage of a film for the application of the retorting. It is necessary to set the thermosetting temperature near 210°C for this reason.

The relationship between bowing distortion and annealing temperature is plotted in Fig. 6. The bowing distortion tend to increase rapidly over the thermosetting temperature over 200°C. It is estimated that the vertical deformation increases, as the vertical shrinkage force, which arises in thermal treat-
ment equipment, approaches the central of a film. Thermal contraction is generated in the high temperature area, because of that; the bowing phenomenon expands by the heat treatment process, which is oriented biaxial. It is estimated with that the thermal contraction becomes a maximum in the region, which reached the maximum temperature and that bowing distortion takes the highest value. The bowing line showed the bilaterally symmetric shape.

The relationship between bowing distortion and relaxation ratio is plotted in Fig. 7. When the film relaxation ratio is increased, the bowing distortion proportionally increases. Lateral restraining force of a film decreases, when the width relaxation ratio increases. Therefore, it is estimated that the deformation of a film increases and that the bowing distortion increases as a result. It is also based on influence in the wait of the contraction stress of the MD direction, since film width orientation tensile force lowers. The effective product width decreases too.

3.2 Double Bubble Stretching and Triple Bubble Thermosetting Process

Table 1 shows the relationship between thermosetting process and bowing distortion. As a result of evaluating bowing distortion of a film after the double bubble biaxial stretching, the bowing distortion was 0. The line pulled in non-stretched film before the stretching straight completely did not change in stretching, after the strain was not generated. The bowing phenomenon completely did not arise from this in the double bubble stretching process from the experiment.

In the tubular process, the bubble internal pressure controls the lateral stretching force. Therefore restraining force in the width orientation becomes uniform in all position. Where the cross-direction force of a film also extends in the uniform condition, because the clip is not used for the stretching in case of the tubular stretching process. It is considered that in the tubular process, the bowing phenomenon is not produced as a result.

As a result of evaluating bowing distortion of a film after a double bubble biaxial stretching, the bowing distortion was 0. The line pulled in non-stretched film before the stretching straight is complete did not change in the thermosetting after the strain was not generated. The bowing phenomenon completed did not arise from this in the thermosetting of the triple bubble tubular thermosetting process from this experiment.

In the tubular thermosetting process, the bubble internal pressure controls the lateral shrinkage force. Therefore restraining force in the width orientation becomes uniform in all position. Where the cross-direction force of a film also extends in the uniform condition, because the clip is not used for the thermosetting in case of the triple bubble thermosetting process. It is considered that in the tubular thermosetting process, the bowing phenomenon is not produced as this result too.

However, the bubble showed the gourd phenomenon with heightening the annealing/heat setting temperature and some unstable tendencies were shown, and the good sample became difficult to be obtained. Mechanical heat setting using the tenter equipment is necessary in order to carry out sufficient heat setting. The stretching was carried out in the double bubble, and the thermosetting should use the tenter process.

3.3 Annealing in the Two-step Tenter Process

The bowing distortion increased in the tenter method heat set. Then tenter heat set was conducted in the two-step process and the influence on the bowing distortion was examined. The relationship between bowing distortion and annealing temperature in two-step is plotted in Fig. 8. It was possible to hold the bowing distortion smaller than the one-step annealing system in a two-step annealing system (Table 1).

<table>
<thead>
<tr>
<th>Thermosetting system</th>
<th>Bowing distortion (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>① Tubular stretching system</td>
<td>0</td>
</tr>
<tr>
<td>② Tubular thermosetting system</td>
<td>0</td>
</tr>
<tr>
<td>③ One-step tenter process</td>
<td>7.5</td>
</tr>
<tr>
<td>④ Two-step tenter process</td>
<td>5.2</td>
</tr>
</tbody>
</table>

Table 1. Relationship between one-step annealing and two-step annealing system
The effect of two-step thermosetting will be estimated as following. It was possible that the contraction stress of a film was reduced in the processing of one-step. It became possible the fact of two-step in which initial contraction stress in the high temperature processing was kept small, and the bowing distortion was reduced as a result.

The bowing effect of decreasing is confirmed, when the quenching temperature decreases further than glass transition temperature.

### 3.4 Film Shrinkage Percentage Pattern In-plane and Film Orientation

The relationship between orientation pattern and shrinkage pattern is shown in Fig. 9. The estimated result at three positions is shown. (central position and both edge positions). In the edge, the tendency in which the shrinkage percentage of diagonal 45° differed was shown, though the central had as a result of evaluating the film shrinkage percentage pattern in the each direction. Both edges showed the symmetrical shrinkage percentage pattern for the central axis.

In the edge, the main shaft had turned to the MD direction on the central as a result of evaluating the film orientation pattern, and the tendency in which the main shaft tilted a little was shown. Both edges showed the symmetrical orientation pattern for the central axis. The effect of the bowing phenomenon on the film physical property was confirmed.

### 3.5 Sag Elimination Tension

Fig. 10 shows the slitter machine. The roll film of the fixed width was collected from the film in which the thermosetting temperature changed, and the sag elimination tension was calculated using the slitter machine.

The relationship between thermosetting temperature in two-step and sag elimination tension is plotted in Fig. 11. The tendency in which the sag elimination tension as a film in which the big thermosetting temperature of the bowing distortion as a result is high increased was shown.

The relationship between bowing distortion and sag elimination tension is plotted in Fig. 12. There was in the proportional connection shown in the Fig. 12. It is important to reduce the bowing distortion in order to decrease the film sag.

### 3.6 Shrinkage Percentage and Humidity Expansion

It is considered that the phenomenon of sag originates from the elongation of the minute quantity of a film in the film width orientation. Fig. 13 shows the humidity expansion. It is considered that the elongation of film is influenced by humidity expansion in the moisture absorption (Fig. 13).

Table 2 shows the relationship between shrinkage percentage and humidity expansion coefficient. It tried to compare the humidity expansion coefficient in the direction in which
the shrinkage percentage in the place where the anisotropy is big in the product edge differed; it is greatly different in film and edge in the central, when humidity expansion coefficient at 45° and 135° was measured.

The humidity expansion coefficient of the direction of the big hot water shrinkage percentage showed the high value. Likewise, the humidity expansion coefficient of the smallish direction of hot water shrinkage percentage showed the small value. It was confirmed that there were shrinkage percentage and humidity expansion coefficient as a result in the proportional connection. In the position of which the anisotropy of a film is big, this fact means the film in which elongation quantity is in the moisture absorption is big. It is regarded as sag arising, since the moisture extends the strain of the edge by bowing absorption excessively. It was proven that sag increased, as the bowing distortion increases.

The tension by the slitter machine had to be increased in order to remove sag that occurs in extending the film edge. It is understood that there is the correlation as a result between bowing distortion and sag elimination tension (Fig. 14). The printing can smoothly carry it out, if there is no sag in a film.

### Table 2. Film shrinkage percentage pattern and humidity expansion coefficient

<table>
<thead>
<tr>
<th>Position</th>
<th>Film shrinkage percentage %</th>
<th>Humidity expansion coefficient 1/ %</th>
</tr>
</thead>
<tbody>
<tr>
<td>➀ Center</td>
<td>45°</td>
<td>3.7</td>
</tr>
<tr>
<td>➁ Center</td>
<td>135°</td>
<td>3.8</td>
</tr>
<tr>
<td>➂ Edge</td>
<td>45°</td>
<td>5.7</td>
</tr>
<tr>
<td>➃ Edge</td>
<td>135°</td>
<td>2.2</td>
</tr>
</tbody>
</table>

### 3.7 Shrinkage Stress

The triple bubble tubular annealing/heat setting process is a method for thermosetting by the inside bubble pressure, using the device shown in Fig. 15. In principle, shrinkage stress may be calculated in accordance with the tubular theoretical equation analyzed by the authors et al. [14]. The stresses at the end point of heat set may be obtained by the following equation:

$$\sigma = \Delta P \cdot D/2t,$$

(2)

![Fig. 15. Measurement method of shrinkage stress in a triple bubble thermosetting system](image-url)
where $\sigma$: shrinkage stress in the TD direction, $\Delta P$: internal bubble pressure, $D$: bubble diameter at end point of heat set, $t$: film thickness at end point of heat set.

In the triple bubble tubular film process, the shrinkage stress in the width direction may be calculated by measuring the inside bubble pressure. Fig. 16 shows the relationship between shrinkage stress and annealing temperature which illustrates that the larger thermosetting temperature is the lower shrinkage stress. The actual film temperature is about 26% lower (in °C) than that shown in Fig. 16. Fig. 17 shows the relationship between shrinkage stress and relaxation ratio illustrating the larger relaxation ratio and the lower shrinkage stress. It was verified that the tensile force which work in back direction in heat set increased and that the bowing distortion increase as a result, as the shrinkage stress was bigger. It is regarded as reducing the shrinkage stress in the one-step heat set, when heat set was carried out from this fact in the two-step process. Reducing bowing distortion in heat set by the tubular process is difficult, to basically avoid the bowing phenomenon.

**4 Conclusion**

This report is discussed on the bowing phenomenon and sag elimination tension for biaxial oriented PA 6 film process, which consists of double bubble tubular film process and thermosetting tentering one.

The bowing phenomenon does not occur in the heat setting of the triple bubble tubular process. However, there is a problem of bubble stability when sufficient annealing by the tubular process is carried out.

The bowing phenomenon arises when the annealing is carried out by the tenter method. Bowing distortion becomes large when the annealing temperature is high and film relaxation ratio is large.

It is possible to reduce the bowing distortion when heat setting is carried out in the two-step of tenter process. Shrinkage stress of the stretched film was relaxed in the thermosetting of the second step whose temperature is higher than the first one. The two-step thermosetting was able to decrease the bowing distortion compared with the one-step of tenter process.

When the bowing phenomenon occurred, shrinkage percentage in hot water changed along the cross-direction of a film. The humidity expansion coefficient in the film increases with increasing shrinkage percentage in hot water is related to the orientation of the molecule. The sag also increased with increasing the bowing distortion. The bowing distortion and the sag elimination tension have the proportional relation.

It is necessary to decrease the sag of a film in order to carry out smooth and clear printing.

**References**


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