The Effect of Molecular Structure of Polypropylene on Stretchability for Biaxially Oriented Film

Satoshi Tamura,1,2 Itaru Kuramoto,1 Toshitaka Kanai2,3
1 Research and Development Division, Prime Polymer Co. Ltd., Sodegaura-city, Chiba 299-0265, Japan
2 Division of Material Sciences, Graduate School of Natural Science and Technology, Kanazawa University, kakuma-Machi, kanazawa-City, Ishikawa 920-1192, Japan
3 Research and Development Laboratory, Idemitsu Kosan Co. Ltd. Ichihara-city, Chiba 299-0193, Japan

Biaxially oriented polypropylene films are widely used for food packaging and industrial films. Recently, machine speed has been increasing in order to obtain higher production rate, and film thickness has become thinner to reduce the environmental load. The customers' requirements for better production ability and thinner films have been becoming more demanding, but their demands are not always met because of lack of film stretchability in the final product. To meet the demands, research on stretchability has been conducted with the goal of finding the optimum polypropylene molecular structure for developing a new product by analyzing stretching force–strain data using a table tenter, which was thought to be the parameter of stretchability. It was found that low crystallinity and wide molecular weight distribution were effective in improving the stretchability from the table tenter test. By running the test with a sequential and biaxially oriented stretching machine, it was verified that samples produced by the above designed polymer indicated good thickness uniformity, which was considered to be the barometer of stretchability. Furthermore, it was concluded that analyzing the stretching force–strain data obtained from a table tenter is a good method to predict machine speed and film thickness. POLYM. ENG. SCI., 52:1383–1393, 2012. © 2012 Society of Plastics Engineers

INTRODUCTION

About 40 million tons per year of polypropylene (PP) are currently produced in the world [1]. Biaxially oriented PP (BOPP) film accounts for a large amount of PP, since it is suited for food packaging films or industrial films and because it has high performance in terms of mechanical and optical properties. Recently, higher production speeds are required to reduce the production costs, and thinner films are requested for the purpose of reducing the environmental load. Yet, there are some cases when PP products do not satisfy the demands due to a lack of stretchability. In those cases, film breaking occurs at transverse direction (TD) stretching process when it is produced by a sequential and biaxially oriented stretching machine (Fig. 1). Film quality also drops because of wrinkles created by low thickness uniformity.

To overcome the problem, various studies have been performed by many researchers. For example, the stretchability of various stretching process was studied [2–7], and the relationship between the stress–strain curve of uniaxial stretching and deforming of PP crystal was studied using a spectral birefringence technique by various researchers. However, the results did not show any relationship with stretchability [8]. Phillips and Nguyen reported the relationship between stress–strain curve and the crystal structure of biaxially oriented film, but there was no indication of the influence of the stress–strain curve on the stretchability [9].

Kanai et al. gave a report on the prediction of film thickness uniformity using the stress–strain curve in their reports [10, 11]. The relationship between strain (stretching ratio) and stretching force is indicated in Fig. 2. In this article, the stretching process can be divided into three regions in terms of thickness uniformity. Where there is lower stretching ratio at the beginning of stretching (region I), the stretching force increases in proportion to stretching ratio showing local maximum force. At the middle stage (region II), the stretching force indicates the flat line, and at the last stage (region III), the stretching force increases again with increasing stretching ratio [12–13]. Figure 2 also shows the thickness uniformity with the cross-sectional view of film. It is well known that PP is stretched unevenly, which is called neck-like deformation at a lower stretching ratio. From the begin-

Correspondence to: Satoshi Tamura; e-mail: Satoshi1.Tamura@primepolymer.co.jp
DOI 10.1002/pen.22180
Published online in Wiley Online Library (wileyonlinelibrary.com).
© 2012 Society of Plastics Engineers

POLYMER ENGINEERING AND SCIENCE—2012
ning to the middle stage of stretching, thickness uniformity gradually worsens. At the later stage (region III), thickness uniformity improves because the highly stretched part has a higher stretching force than the lower stretched part, and the lower stretched part becomes easier to stretch with a low stretching force. This has been observed in uniaxial, simultaneous biaxial and sequential biaxial stretching experiments in other resin such as polyethylene terephthalate (PET) [14] and polyetheretherketone (PEEK) [15].

There are some reports regarding the relationship between the stress–strain curve and molecular structure of PP. The stretching force at the yield point was related to the degree of crystallinity as reported by Butler et al. [16], deformation of crystal as reported by Phillips et al. [17], and the thickness of lamella as reported by Kanai et al. [11, 13]. Consequently, the crystallinity of PP is considered to be the influence factor to the stretching force at the yield point. However, Kanai reported that the stretching force at a later stage changes with the amount of the component with a long relaxation times measured by melt viscoelasticity. It is known that high molecular weight components cause long relaxation times, but the flow-ability falls only if the amount of high molecular weight components is increased. To maintain the flow-ability in the case of increasing of the high molecular weight components, widening the molecular weight distribution (MWD) is considered to be a good method of controlling the stretching force at a later stage.

However, there is no research showing the relationship between the stretching force obtained by table tenter and the actual thickness uniformity data obtained by a sequential and biaxially oriented stretching machine. It is suspected that uneven thickness of the film is the cause of film breaking during the stretching process, and this is also believed to be one of the reasons making thinner films difficult. Therefore, film thickness uniformity is considered to be an important barometer of stretchability. This report will show the prediction of stretchability using a table tenter, and the test results of film thickness uniformity using a sequential and biaxially oriented stretching machine.

EXPERIMENTS

Samples

All samples were polymerized by Ziegler-Natta catalyst with different donors to change the molecular structure of isotacticity and MWD. Sample A, which is generally used for BOPP film produced commercially in our company as a grade name of F-300SP, was used as a standard sample. Sample V with high isotacticity produced by our middle size of our pilot plant was prepared to investigate the relationship between stretchability and crystallinity. Then three samples with low crystallinity from B1 through B3 and two wide MWD samples C1 and C2 were prepared by our pilot plant to investigate the relationship between stretchability and resin properties (Table 1, Fig. 3). To decrease the crystallinity, B1 and B2 were copolymerized with small amount of ethylene, and the isotacticity parameter meso pentad values mmmm measured by carbon-13 nuclear magnetic resonance (13C-NMR) spectroscopy [18] of B3 was reduced. The MWD parameter $M_w/M_n$ measured by gel permeation chromatography (GPC) of B1 and B2 were narrower than that of the standard sample A. $M_w/M_n$ of C1 and C2 were larger than that of the standard sample A with the same isotacti-
TABLE 1. Resin properties of samples.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Unit</th>
<th>Standard</th>
<th>High crystallinity</th>
<th>Low crystallinity</th>
<th>Wide MWD</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>V</td>
<td>B1</td>
<td>B2</td>
</tr>
<tr>
<td>MFR</td>
<td>g/10 min</td>
<td>3.0</td>
<td>3.0</td>
<td>3.0</td>
<td>2.9</td>
</tr>
<tr>
<td>mm mol%</td>
<td></td>
<td>90</td>
<td>97</td>
<td>97</td>
<td>90</td>
</tr>
<tr>
<td>C2 = amount wt%</td>
<td></td>
<td>0.0</td>
<td>0.0</td>
<td>0.5</td>
<td>88</td>
</tr>
<tr>
<td>C7-II wt%</td>
<td></td>
<td>96.7</td>
<td>99.0</td>
<td>97.4</td>
<td>97.3</td>
</tr>
<tr>
<td>$T_m$ °C</td>
<td></td>
<td>161.4</td>
<td>165.7</td>
<td>159.7</td>
<td>158.4</td>
</tr>
<tr>
<td>$H_m$ J/g</td>
<td></td>
<td>99.6</td>
<td>111.0</td>
<td>99.4</td>
<td>100.2</td>
</tr>
<tr>
<td>$T_c$ °C</td>
<td></td>
<td>102.0</td>
<td>105.4</td>
<td>103.0</td>
<td>102.8</td>
</tr>
<tr>
<td>$M_w \left(10^{-5}\right)$</td>
<td>—</td>
<td>3.56</td>
<td>3.61</td>
<td>3.29</td>
<td>3.50</td>
</tr>
<tr>
<td>$M_n \left(10^{-4}\right)$</td>
<td>—</td>
<td>7.73</td>
<td>8.68</td>
<td>9.14</td>
<td>8.53</td>
</tr>
<tr>
<td>$M_w/M_n$</td>
<td></td>
<td>4.6</td>
<td>4.2</td>
<td>3.6</td>
<td>4.1</td>
</tr>
</tbody>
</table>

FIG. 3. DSC charts [(a), (c), and (e)] and GPC charts [(b), (d), and (f)] of each sample.
Heptanes insolubilities (C7-II) measured by soxhlet abstraction method were used to compare with the crystallinity of each sample. Thermal properties such as melting point (T_m), melting enthalpy (ΔH_m), and crystallizing temperature (T_c) were measured by differential scanning calorimetry (DSC). These seven samples with different properties in crystallinity and MWD were used to investigate the relationship with the stretchability.

**Stretching Test Using a Table Tenter**

A nonstretching sheet, with a thickness and width of 1 mm and 270 mm, respectively, was made by a sheet forming machine (Tanabe Plastics Co.) at a chilling roll temperature of 30°C. The sheet was stretched 4.6 times in the machine direction at 147°C by which all samples were stretched properly by a heated roll type stretching machine (Iwamoto Seisaku-sho Co.).

Uniaxially stretched sheet in the machine direction was cut to the size of 290 mm in length (machine direction) and 82 mm in width (TD) and was stretched in a TD by the table tenter (Iwamoto Seisaku-sho Co.). The stretching ratio was 9.2 at several temperatures after preheating for 1 min. At last, a BOPP film with a thickness of 24 μm was obtained. The stretching force was measured by a load cell that was equipped on a chuck of the table tenter to predict the stretchability of each sample.

The force data were determined as follows. A stress–strain curve of PP is shown in Fig. 4. It was defined that the local maximum force observed at the beginning of the strain as the stretching force at the yield point (F_y), the minimum drawing force observed in the middle of the strain as the minimum drawing stretching force (F_d), and the stretching force observed at the end of the strain as the stretching force at the maximum strain (F_m), respectively. The stretching ratio at F_d was defined as the R_d. These stretching force parameters were used to predict stretchability. The film thickness uniformity of PP-α was predicted to be good, because its F_m is bigger than F_y, i.e., the stretched part gets harder than the nonstretched part. The stretched part requires more effort to be stretched than the nonstretched part. The resin design that indicates the PP-α’s stress–strain curve, such as low F_y and high F_m, is believed to be necessary in order to improve the stretchability. Therefore, F_m/F_y is considered to be one of the parameters of stretchability. It is believed that PP resin with more than one F_m/F_y should have good stretchability, and PP resin with less than one F_m/F_y should have poor stretchability. The investigation on stretchability from the point of F_m/F_d is reported in this article.

**Stretching Test Using a Sequential and Biaxially Oriented Stretching Machine**

BOPP films were produced by a sequential and biaxially oriented stretching machine (Mitsubishi Heavy Industries Co.) using samples from B1 to C2 and the standard sample A. PP resins were extruded by an HM tandem-type extruder with a discharge amount of 390 kg/hr, and a nonstretched sheet with 270-mm width was made using a roll type casting machine at the roll temperature of 30°C and take-off speed of 55 m/min. After the sheet was stretched in a machine direction for 4.5 times at 138°C using heated rolls, BOPP film of 15 μm thickness and 1-m width was obtained through a tenter process with a stretching ratio to the TD 9.5 times. Final machine speed was 270 m/min, and the strain rate was 423%/s, which is close to that of a large size production line of BOPP film. The stretchability of each sample was verified by judging a thickness uniformity that is considered to be one of the important barometers of stretchability of BOPP film, which was measured by film thickness distribution.

**RESULTS AND DISCUSSION**

**Examination for Predicting a Stretchability Using a Table Tenter**

At first, the influence of crystallinity on stretchability was examined with samples A and V, which have quite different meso pentad values mmmm as parameters of crystallinity (Table 2, Fig. 5). F_y value of sample V with mmmm value 97 mol% at 164°C and 166°C indicated 2.49 kgf and 1.90 kgf, respectively. They were 2.5 times as high as those of the standard sample A with mmmm.
value 90 mol%. Stretching force at the yield point increased with increases of the sample’s crystallinity, but the estimation was made that the difference of crystallinity will be about $6^\circ C$, i.e., stretching force at the yield point of sample V will be equal to that of sample A if the stretching temperature of sample V was raised by $6^\circ C$.

However, $F_m$ values of sample V at $164^\circ C$ and $166^\circ C$ were 1.9 and 2.0 times as high as those of sample A at the same temperature. The ratios of $F_m/F_d$ values of sample V were smaller than those of sample A, because the MWD $M_w/M_n$ of sample V (4.2) was smaller than that of sample A (4.5).

The sheet made by low crystallinity PP sample A was stretched without breaking at the stretching temperature between $158^\circ C$ and $166^\circ C$, meaning sample A had a process window of $8^\circ C$. However, although the sheet made by high crystallinity sample V (97%) was successfully stretched from $166^\circ C$ to $172^\circ C$, it broke during stretching at $164^\circ C$. It was found that the stretchability of sample V was inferior to that of sample A, because the process window of sample V was $6^\circ C$, which means it is $2^\circ C$ narrower than that of sample A. A possible reason is the influence of the narrower composition distribution of sample V, compared with sample A (Fig. 6, Table 3).

It means that the difference in temperature between the molten part of 70 wt% and that of 40 wt% of sample V ($5.9^\circ C$), which is related to the process window of stretchability, is narrower than that of sample A ($7.7^\circ C$). Since a proper amount of molten part is necessary to stretch for BOPP film as was studied by Uehara et al. in double bubble tubular stretching process [19–22], widening the composition distribution is an effective method in improving its processability in tentering stretching process.

The stretching forces $F_y$, $F_d$, and $F_m$ obtained from these stretching curves were plotted in Fig. 7a. $F_y$ and $F_d$ of sample V indicated almost the same value as those of sample A at $164^\circ C$. $F_m$ values of sample V at $164^\circ C$ and $166^\circ C$ were 1.9 and 2.0 times as high as those of sample A at the same temperature. The ratios of $F_m/F_d$ values of sample V were smaller than those of sample A, because the MWD $M_w/M_n$ of sample V (4.2) was smaller than that of sample A (4.5).
sample A when its temperature was lowered by 6°C, but $F_m$ of sample V was lower than that of sample A as the stretching temperature was lowered. The stretching force ratios $F_m/F_y$ and $F_m/F_d$ calculated from each stretching force are plotted in Fig. 7b. Since both $F_m/F_y$ values of sample A and V increased with increasing of stretching temperature, stretchability was considered to be better at higher temperature in this range. However, since $F_m/F_y$ of sample V at the range of 164°C–170°C were <1.0, it would only enable us to produce BOPP film of sample V with poor film thickness uniformity even if the film breaking did not occur. In other words, sample V is required to be stretched at only from 170°C to 172°C to make BOPP film with a good thickness uniformity. Therefore, sample V has quite a narrow process window in terms of film quality although it has a process window of 6°C from 164°C to 170°C.

In the mean time, both $F_m/F_d$ values of sample A and V decreased gradually with increasing the temperature. As $F_d$ is a value that depends on average molecular weight and crystallinity, it is estimated that $F_m/F_d$ is related to the amount of high molecular weight component to the average of molecular weight for the same crystallinity samples. $F_m/F_d$ of sample V was lower than that of sample A, because the MWD of sample V was narrower than that of sample A. $F_m/F_d$ should decrease as the temperature rises, because the force of entanglement reduces as the temperature rises.

It was found that the stretching ratio $R_d$ at the lowest stretching force of sample V was larger than that of sample A (Table 2, Fig. 5). The stretching ratio of sample V must be higher than that of sample A to alter the crystallinity component. Therefore, stretchability of sample A is assumed to be better than that of sample V at the same temperature, even when the temperature was raised by 6°C to fit the stretching force at the yield point.

Further research for the purpose of grasping the relationship between the stretchability and resin properties was conducted using six samples from B1 to C2 and the standard sample A. The stretching force curve at 164°C obtained by table tenter is indicated in Fig. 8. The stretching forces $F_y$ at the yield point of samples from B1 to B3 with reduced crystallinity were lower than that of the standard sample A (Fig. 8a). The stretching forces $F_m$ observed at the maximum stretching ratio of samples C1 and C2 were larger than that of sample A. The effect of wide MWD was confirmed (Fig. 8b). The stretching force data at 164°C are indicated in Table 4.

The stretchability of each sample was predicted by connecting the stretching force parameter with MWD $M_w/M_n$ (Fig. 9). As a result, it was predicted that the stretchability of all samples except for B1 is assumed to be greater than that of the standard sample A, because $F_m/F_y$ of all the samples except for B1 was higher than that of A (Fig. 9a). Also, $F_m/F_y$ was determined as the parameter related to both crystallinity and MWD, because the data were able to fit into two lines with the same range of C7-II (heptanes’ insolubilities), which was one of the parameters of crystallinity. It was assumed that the stretchability improved with an increase of $M_w/M_n$ and a decrease of crystallinity from $F_m/F_y$ parameter. The stretchability of low crystallinity sample B1 did not improve compared with sample A, because the effect to worsen the stretchability by narrow $M_w/M_n$ was greater than the effect to improve the stretchability by low crystallinity in a more effective way.

However, it was recognized that the $F_m/F_d$ is a parameter in proportion to $M_w/M_n$ (Fig. 9b). The amount of the long-time relaxation component becomes larger when the MWD is widened, which causes strain hardening. It is important to select a suitable extruder to maintain its long-time relaxation component, because PP MWD is easily shortened by degradation under severe conditions in the extrusion process [23].

**Examination for Verifying Stretchability Using an Sequential and Biaxially Oriented Stretching Machine**

Next, an experiment was conducted to verify stretchability of each sample by measuring film thickness unique...
formity using a sequential and biaxially oriented stretching machine. BOPP film thickness distribution was controlled by checking its thickness with a β-ray thickness gauge during production, and reflected the data to die clearance distribution (Fig. 1) [24]. The film thickness uniformity data were measured after its distribution was roughly stabilized.

The film rolled on a winder was cut, and thickness \( t \) for 2000 points to the TD and 10 points to the machine direction was measured. The film thickness uniformity was evaluated to the TD using the standard deviation \( \sigma \) calculated by Eq. 1.

\[
\sigma = \frac{1}{2000} \sqrt{\sum_{n=1}^{2000} (t_n - \bar{t})^2}
\]

(1)

where \( t_n \) is the film thickness, and \( \bar{t} \) is the average of film thickness.

However, it was uncertain if \( \sigma \) was suitable for estimating the thickness uniformity, because we were unable to decide if \( \sigma \) was the best data for each sample due to the shortage of experiment time and sample amount. Furthermore, it was probable that the thickness distribution in the TD was different even if the lip clearance was the same, because each sample has its own flow feature. Therefore, film thickness variation coefficient in the machine direction \( \delta \) calculated by Eq. 2 was used, which enable to estimate the amount of difference between the film thickness at the same point to the TD. Since \( \delta \) is most unlikely to be influenced by experiment time or sample amount, even if film thickness distribution in the TD is not good, \( \delta \) can tell us the film thickness uniformity. Consequently, we came to the conclusion that the film thickness variation coefficient in the machine direction \( \delta \) is the most important barometer to evaluate the stretchability of each sample.

\[
\delta = \frac{1}{2000} \sqrt{\sum_{j=1}^{2000} \left( \frac{t_i - t_{i+1,j}}{C_0} \right)^2}
\]

(2)

where \( t \) is the film thickness, subscript \( i \) is the \( i \)th point to the machine direction, and subscript \( j \) is the \( j \)th point to the TD.

Before checking the film thickness, the stretching behavior on the machine direction stretching roll was observed. It could be seen from the result of all samples tests that vibrations of the stretching line on the stretching roll occurred for B1 and B2. The stickiness of B1 and B2

![Graphs showing stretching force and force ratio](image-url)

**FIG. 7.** (a) Stretching force and (b) stretching force ratio of sample A and V.

![Graphs showing strain-stress curve](image-url)

**FIG. 8.** Strain–stress curve of (a) low crystallinity samples and (b) wide molecular weight distribution samples.
to the roll is assumed to be much higher than that of other samples, because the accumulated weight ratio of melting parts of B1 and B2 at the roll temperature of 138°C is larger than that of other samples (Fig. 10, Table 5). Vibration occurred when the balance between the stickiness of resin to the roll and the stretching force was changed. Meanwhile, the narrower MWD of B1 and B2 is considered to be one of the causes of the vibrations (Fig. 11). Since it is a well-known fact that the phenomenon known as neck-in of the sheet easily occurs for resins with a narrow MWD due to the lack of melt tension, the thickness of the edge should be thicker than average (namely edge beads and the edge part easily detach from the stretching roll). Therefore, it is considered that the stretching line of B1 and B2 is not as stable as other samples.

Next, film thickness variation coefficients in the machine direction $d$ and the standard deviation to the TD $r$ were checked. Both $d$ and $r$ showed a good correlation with the stretching force ratio $F_m/F_y$ obtained by the table tenter (Table 6, Fig. 12). The film thickness uniformity of all samples except for B1 improved in comparison with the standard sample A as was predicted by the table tenter. However, the film thickness uniformity of lower crystallinity sample B3 obtained by a sequential and biaxially oriented stretching machine was not as good as was expected. It is supposed that $F_y$ of sample B3 obtained by the table tenter is assumed to be smaller than we estimated, because the stretching temperature of the sequen-

### TABLE 4. Stretching parameters of sample from A to C2 obtained by a table tenter.

<table>
<thead>
<tr>
<th>Samples</th>
<th>$F_y$ (kgf)</th>
<th>$F_d$ (kgf)</th>
<th>$R_d$</th>
<th>$F_m$ (kgf)</th>
<th>$F_m/F_d$</th>
<th>$F_m/F_y$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>1.02</td>
<td>0.79</td>
<td>5.33</td>
<td>1.06</td>
<td>1.29</td>
<td>1.34</td>
</tr>
<tr>
<td>B1</td>
<td>0.97</td>
<td>0.75</td>
<td>6.0</td>
<td>0.98</td>
<td>1.29</td>
<td>1.31</td>
</tr>
<tr>
<td>B2</td>
<td>0.93</td>
<td>0.74</td>
<td>5.60</td>
<td>0.99</td>
<td>1.25</td>
<td>1.33</td>
</tr>
<tr>
<td>B3</td>
<td>0.92</td>
<td>0.77</td>
<td>4.8</td>
<td>1.05</td>
<td>1.19</td>
<td>1.36</td>
</tr>
<tr>
<td>C1</td>
<td>0.98</td>
<td>0.82</td>
<td>5.3</td>
<td>1.11</td>
<td>1.20</td>
<td>1.35</td>
</tr>
<tr>
<td>C2</td>
<td>0.94</td>
<td>0.79</td>
<td>5.3</td>
<td>1.10</td>
<td>1.19</td>
<td>1.39</td>
</tr>
</tbody>
</table>

$F_y$: stretching force at yield point, $F_d$: minimum stretching force at drawing, $R_d$: stretching ratio at minimum stretching force, and $F_m$: stretching force at maximum ratio.

![FIG. 9. Relationship between $M_w/M_n$ and (a) $F_m/F_y$, (b) $F_m/F_d$.](image)

![FIG. 10. Accumulated weight ratio of the melting parts on DSC (wt%).](image)
tial and biaxially oriented stretching machine was 160°C, while the table tenter was at 164°C. On the other hand, \( F_m/F_d \) is also a good parameter to predict \( \delta \) and \( \sigma \) (Fig. 13). Low crystallinity sample B3 had a good correlation with other samples, because \( F_m/F_d \) is a parameter related to the MWD.

Finally, the relationship between the resin parameter and stretchability is indicated in Fig. 14. It is thought to be a good method to widen MWD to improve the film thickness uniformity parameters \( \delta \) and \( \sigma \). Although sample A has a wider MWD than sample B1, its \( \sigma \) and \( \delta \) were almost identical to that of sample B1. Sample A should have been stretched at a higher temperature than sample B1, because the melting point of sample A is higher than that of sample B1. However, two samples were stretched at the same temperature, and \( \delta \) and \( \sigma \) values of sample A did not meet the expected values. Therefore, it was concluded that reducing the crystallinity and widening the MWD is a good method to make a film with proper thickness uniformity.

**CONCLUSIONS**

Stretching force parameters obtained by a table tenter were investigated to predict the stretchability and to develop new materials to satisfy the demands of higher production speeds and thinner films. After the analysis on \( F_m/F_y \) and \( F_m/F_d \) was conducted, it was predicted that the stretchability of PP resin will improve by widening the MWD and reducing the crystallinity. It was also found that reducing the crystallinity is effective in widening the process window of the stretching temperature in the stretching process.

By means of the test using a sequential and biaxially oriented stretching machine, reducing the crystallinity of PP resin by copolymerization was found not to be a good method to improve the stretchability, because the vibrations of the stretching line on the MD rolls occurred and made the film thickness unstable. It was verified that the film thickness uniformity, which was the barometer of stretchability obtained from a sequential and biaxially ori-

<table>
<thead>
<tr>
<th>Vibration</th>
<th>A</th>
<th>B1</th>
<th>B2</th>
<th>B3</th>
<th>C1</th>
<th>C2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Accumulated weight ratio of melting parts at 138°C on DSC (wt%)</td>
<td>21.1</td>
<td>23.4</td>
<td>24.2</td>
<td>21.5</td>
<td>19.6</td>
<td>18.7</td>
</tr>
<tr>
<td>( M_w/M_n )</td>
<td>4.6</td>
<td>3.6</td>
<td>4.1</td>
<td>4.6</td>
<td>5.4</td>
<td>5.6</td>
</tr>
<tr>
<td>Sheet vibration</td>
<td>○</td>
<td>×</td>
<td>×</td>
<td>○</td>
<td>○</td>
<td>○</td>
</tr>
</tbody>
</table>

**TABLE 5. Sheet vibration on the MD stretching roll (○: not occurred ×: occurred).**

### Film thickness uniformity parameters \( \delta \) and \( \sigma \) obtained by a sequential and biaxially oriented stretching machine.

<table>
<thead>
<tr>
<th>Samples</th>
<th>A</th>
<th>B1</th>
<th>B2</th>
<th>B3</th>
<th>C1</th>
<th>C2</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \delta ) (( \mu m ))</td>
<td>0.62</td>
<td>0.63</td>
<td>0.50</td>
<td>0.48</td>
<td>0.39</td>
<td>0.35</td>
</tr>
<tr>
<td>( \sigma ) (( \mu m ))</td>
<td>3.2</td>
<td>3.8</td>
<td>3.3</td>
<td>3.1</td>
<td>2.7</td>
<td>2.1</td>
</tr>
</tbody>
</table>

**TABLE 6. Film thickness uniformity parameters \( \delta \) and \( \sigma \) obtained by a sequential and biaxially oriented stretching machine.**

**FIG. 11. Sheet vibration on the MD stretching roll.**

**FIG. 12. Relationship between \( F_m/F_y \) and film thickness uniformity parameters (a) \( \delta \), (b) \( \sigma \).**

DOI 10.1002/pen
oriented stretching machine, gave a good correlation with $F_{m}/F_{d}$ and especially $F_{m}/F_{a}$. That is, reducing the crystallinity of PP resin and widening the MWD were effective in improving the film thickness uniformity. It is supposed that bad film thickness uniformity is the cause of film breaking. Therefore, it is important to improve the film thickness uniformity to produce BOPP films at higher rate and to make thinner films.

It was concluded that film thickness uniformity obtained by sequential and biaxially oriented stretching machine had a good relationship with the stretching force parameter obtained by a table tenter. Finally, the analysis of the stretching force obtained by a table tenter with a small amount of resin was an effective technique in predicting the stretchability of BOPP film.

REFERENCES

7. V. Rauschenberger, Predicting the Processability of BOPP (Biaxially Oriented Polypropylene) Material for Film Applications on Laboratory Scale, in ANTEC Proceedings, 152 (1998).
13. T. Kanai, N. Matsuzawa, H. Yamaguchi, T. Takebe, and T. Yamada, Evaluation of structure development and stretchability during the stretching process for various polypropyl-


