Study on Rough-Surface Biaxially Oriented Polypropylene Film. III. Influence of MFI of Base Resin

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Synopsis

In Part II of this series we reported that by using a successively stretching tenter method, biaxially-oriented polypropylene films (BOPs) with a crater-like roughness have been obtained by selecting appropriate machine direction (MD) stretching conditions. In the present article, in order to select still better conditions, the influences of melt flow index (MFI) of base polypropylene (PP) and machine and transverse direction stretching conditions on the roughening of BOP have been studied. Rough-surface BOPs were obtained by casting homoisotactic PPs, having MFIs of 1.3–9.8 dg/min and containing no β-crystal nucleator, with a T-die extruder into sheets which subsequently have been stretched successively in the machine direction (MD) and the transverse direction (TD) with a roll-type stretching machine and a pantograph-type stretching machine, respectively. The level of β-crystal content in the cast sheet was maximum when a PP with MFI $\fallingdotseq$ 8 dg/min was used as a raw material and the obtained roughness state of the BOP was the best. As for the MD-stretching conditions, the roughness state of a BOP improved as the surface temperature of the heating roll was increased to a temperature range of 135–150°C, and there was the best roll rotation speed from the balance of the thermal conductivity of the sheet and the melt-recrystallization rate of the β crystals. A temperature in a range of 150–155°C was best for the TD-stretching.

INTRODUCTION

In Part II of this series[1] we reported that biaxially oriented polypropylene films (BOPs) with a crater-like roughness were obtained by stretching cast sheets containing β crystals in the machine direction (MD) under appropriate conditions, and subsequently stretching them in the transverse direction (TD). It is supposed that the roughness of BOP is strongly influenced by the amount and morphology of the β crystals formed in the cast sheet. In the present article, in order to select the best base polypropylene (PP) for the roughening of BOP, the influence of melt-flow index (MFI) of PP on the roughness of BOP has been studied. The MD- and TD-stretching conditions have also been studied. Yamane et al.[2] state in their patent that the lower the intrinsic viscosity of raw resin, the higher the crystallinity of the sheet cast from it, and hence the more preferable for roughening, but the more difficult to stretch.

Journal of Applied Polymer Science, Vol. 36, 1011–1023 (1988)
 CCC 0021-8995/88/051011-13$04.00

EXPERIMENTAL

Raw Resin

Tokuyama Polypro Grade FB110 (MFI = 1.3 dg/min), YE130 (MFI = 3.4 dg/min), FC140 (MFI = 7.8 dg/min), and FA150 (MFI = 9.8 dg/min) which were homoisotactic PPs produced by Tokuyama Soda Co., Ltd. and did not contain β crystal nucleator.

Casting of Sheet

Sheets 700 μm (800 μm for FB110) thick were cast with a 65 mmϕ extruder equipped with a 500 mm wide T-die from the PPs at an extrusion temperature of 220°C and a chill roll temperature of 90°C.

X-ray diffractions were measured on the cast sheets with a Rigaku Denki RU-200 diffractometer with Ni-filtered Cu-Kα radiation using a rotating specimen table, and β-crystal content (K value) was calculated from the diffraction curves according to Turner-Jones et al.[3,4]

The spherulitic morphologies were observed with a polarizing microscope (Olympus PM-6) on thin pieces ~ 10 μm thick sliced from the cast sheets normal to the MD with a microtome. The melting process of the β crystals was observed with the polarizing microscope on a thin piece ~ 10 μm thick sliced from the FC140 cast sheet normal to the MD with the microtome on a hot stage at a heating rate of 6°C/min.

MD Stretching

The cast sheets were stretched in the MD five times with a roll-type stretching machine shown in Figure 1 of Part II of this series[1] under the following conditions: preheating oven temperature: 140°C; setting temperature of heating rolls: 160°C, 170°C, and 180°C; roll rotation speed[1]: 1/5, 3/15, and 5/25. The cast sheets were passed so that the surfaces opposite to the chill roll touched surfaces touched the 2′ roll. The average roughnesses, R_a, in the MD on the surfaces touching the 2′ roll were measured with a Surfcorder Model TDF-3A manufactured by Kosaka Laboratory Ltd. according to the JIS B0601 for the MD stretched sheets (UOP).

TD Stretching

The UOPs were stretched in the TD 10 times with a pantograph-type stretching machine[1] under conditions of a preheating time of 1 min, a stretching temperature of 150°C, and a stretching rate of 3000%/min. Furthermore, for the FC-UOP MD-stretched under conditions of a heating roll setting temperature of 180°C and a roll rotation speed of 3/15, the TD stretchings were carried out at a temperature range of 140–165°C, and the influence of the TD stretching temperature on the roughness of BOP was studied.

The roughness states of the BOPs obtained were studied by observation of the surfaces with a reflection-type differential interference microscope and the measurements of R_a and haze.[1]

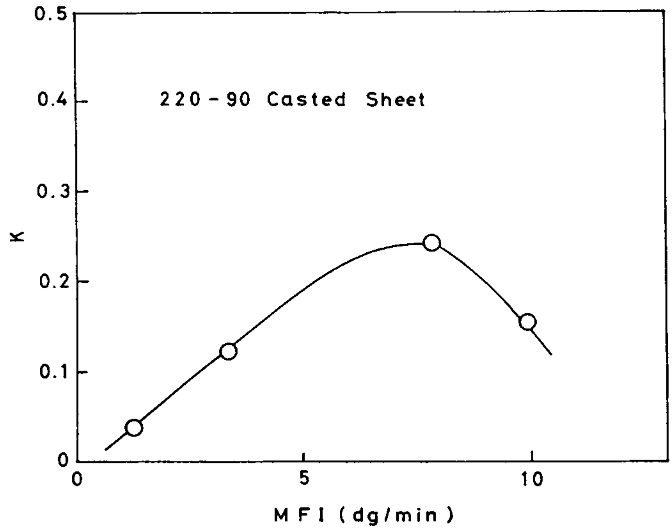

Fig. 1. Dependence of K value of cast sheet on MFI of resin.

RESULTS AND DISCUSSION

Analysis of Cast Sheet

Figure 1 shows the influence of MFI of the base PP on the β-crystal content (K value) of the cast sheet. As in the case of PPs with a β-crystal nucleator,[4] the K value shows a maximum at MFI ≒ 8 dg/min. Although the casting machines and the chill roll temperatures differ from each other, the overlap of Figure 1 on Figure 5 of Part II of this series[4] where PPs contained a β nucleator at levels of 1 and 3 ppm indicates that the dependence of the K value on the MFI of the base PP shows similar tendencies in the case of PPs both with and without the β nucleator and the K values increase in parallel with increasing the β-nucleator content.

Figure 2 shows polarized micrographs of the cast sheets. Since these micrographs are printed in black and white, a tone of color is not clear. However, the large spherulites formed with yellow and blue are observed in original color photographs. These large spherulites which are seen as bright areas in Figure 2 are supposed to represent the β spherulites. The number of the large spherulites increases with increasing MFI of the base PP, whose tendency almost agrees with that of the K value in Figure 1.

MD Stretching

As mentioned in the Experimental section, MD stretching of each cast sheet was carried out at 3 heating-roll temperatures × 3 roll-rotation speeds = 9 conditions.

Figure 3 shows the dependence of the average roughness, R_a, of the UOP obtained from each cast sheet on the surface temperature of the heating roll using the roll-rotation speed as a parameter. In all cases, R_a tends to increase with increasing roll temperature and good rough surfaces were obtained in a

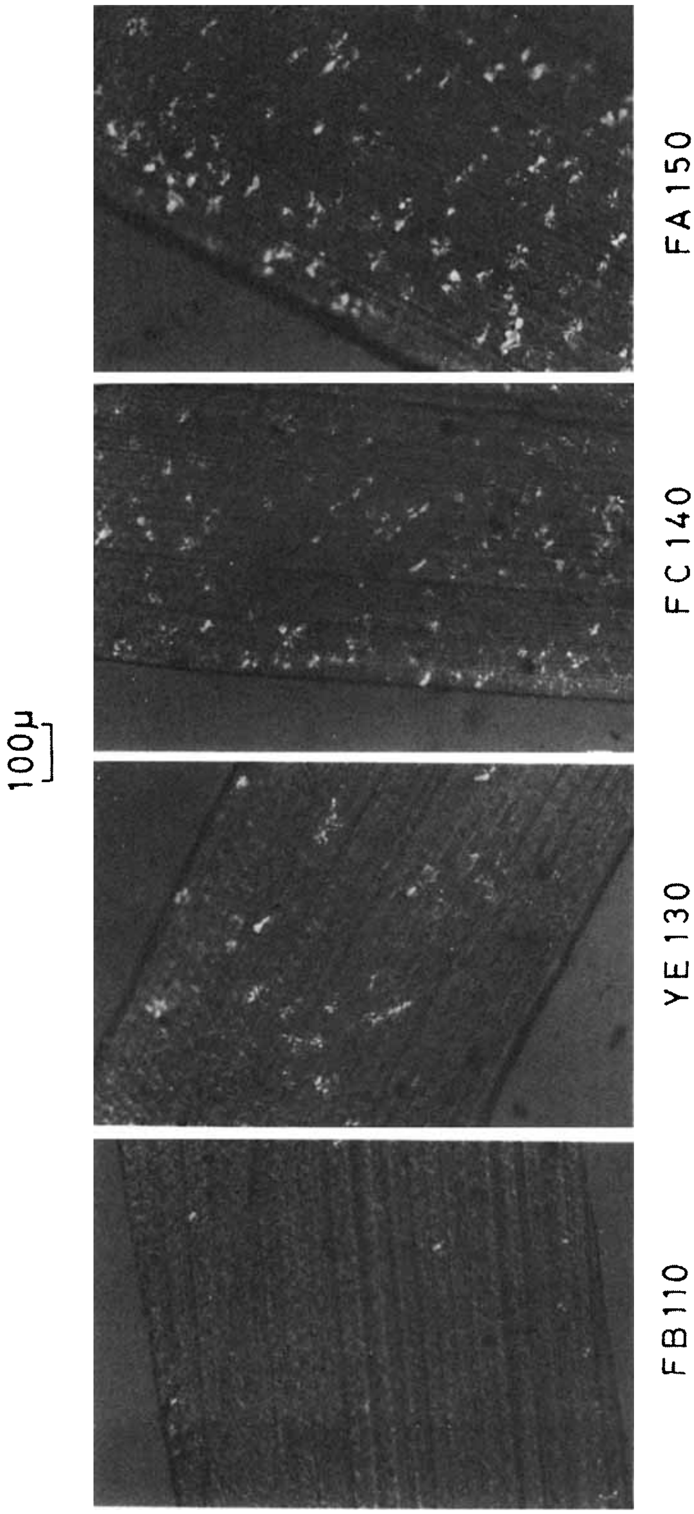

Fig. 2. Polarized micrographs of cast sheets.

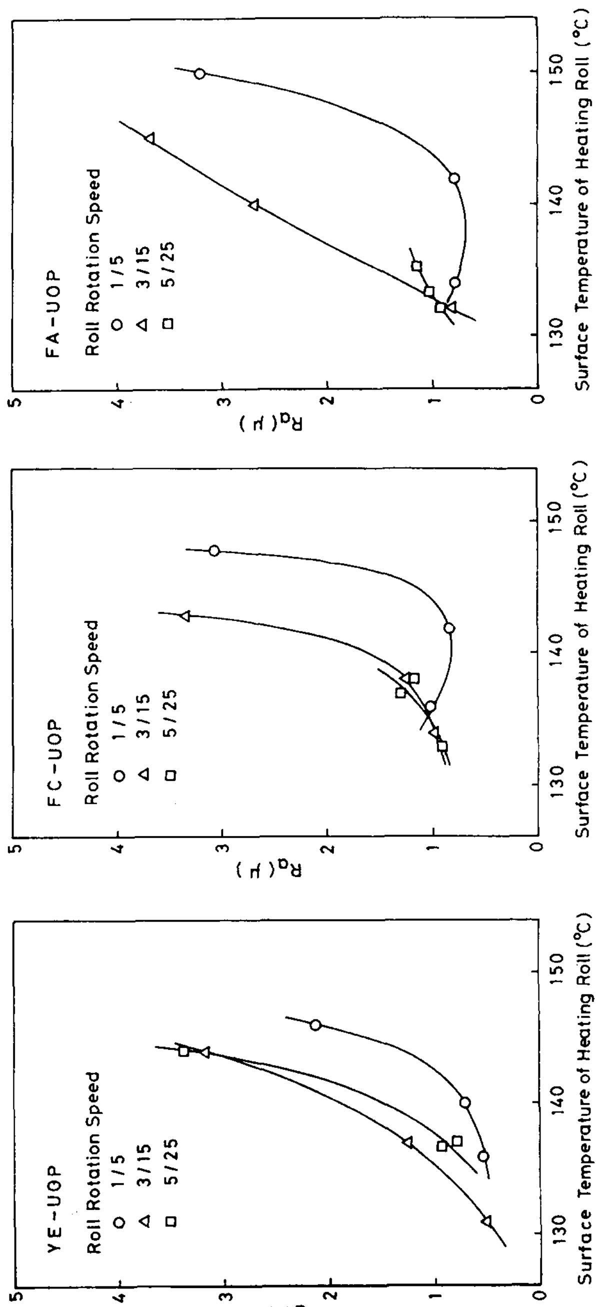

Fig. 3. Dependences of average roughness, R_a, of MD-stretched sheet (UOP) on heating roll temperature. Roll-rotation speed: (○) 1/5; (△) 3/15; (□) 5/25.

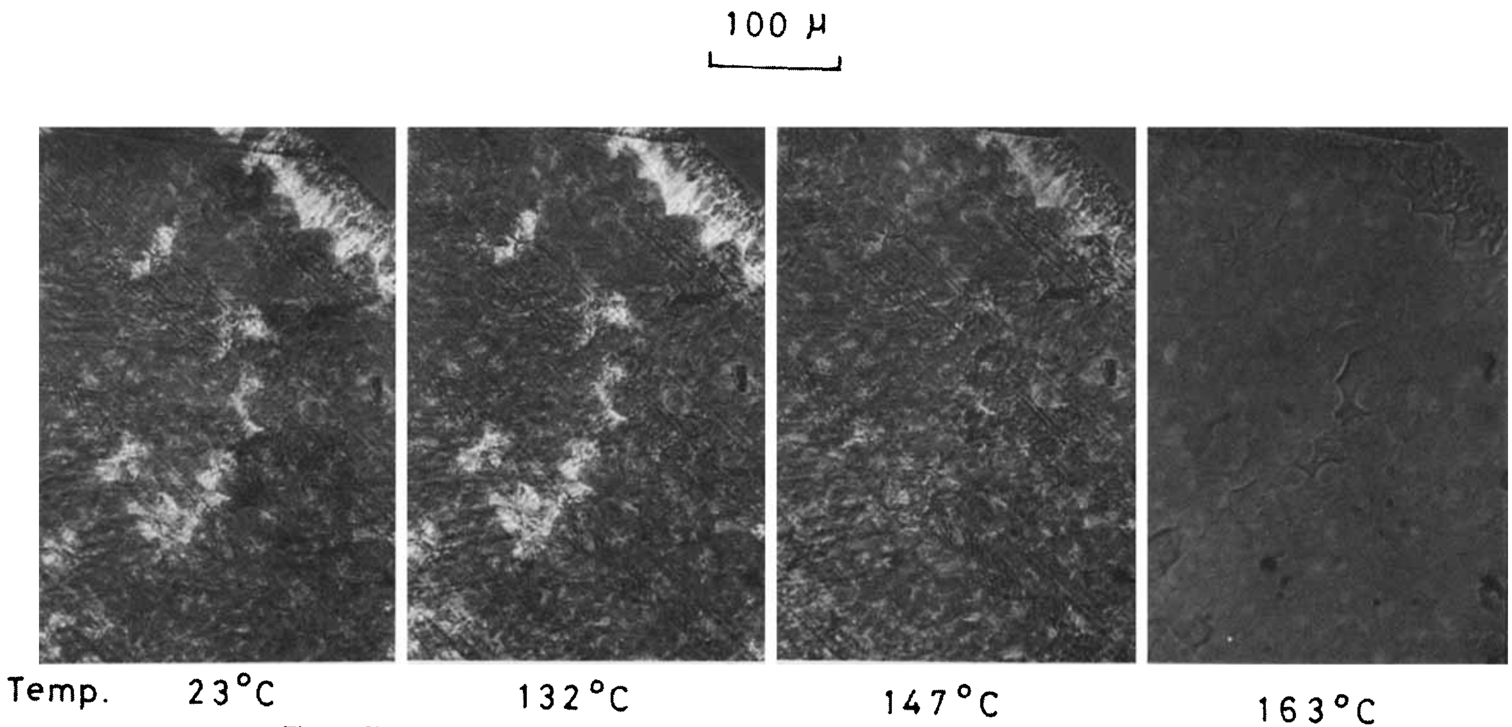

Fig. 4. Variation of spherulitic texture with temperature. FC-cast sheet. Heating rate: 6°C/min.

temperature range of 135–150°C. These facts agree with the experimental result that the recrystallization after melting of the β crystals was more suppressed when the heating temperature was higher in the study on melt-recrystallization of the β crystals by DSC measurements in a previous article.[1] The partial melting of the β crystals was insufficient below 135°C and the probability of sheet breakage was high above 150°C. The higher the roll rotation speed, the lower the temperature at which the sheet breaks. This is assumed to be due to the fact that in the case of high roll rotation speeds, a number of melted parts remain at the time of stretching since the sheet is rapidly heated and recrystallization after melting of the β crystals is suppressed. Figure 4 shows the temperature change of the spherulitic morphology of the FC140 cast sheet heated on a hot stage. The morphology shows almost no change at 132°C from that at room temperature, the β crystals are considerably melted at 147°C, and almost completely melted at 163°C.

Compared with the same surface temperature of the heating roll, R_a is low in the case of a low roll rotation speed (1/5) and high in the case of high roll rotation speeds (3/15 and 5/25). However, there is little difference between R_a in the case of 3/15 and that of 5/25. These behaviors can be interpreted from the viewpoints of the thermal conduction from the heating rolls to the sheet and the melt-recrystallization rate of the β crystals. Figure 5 shows an example of an approximate calculation of the temperature change of a cast sheet heated by the 2′ roll (second heating roll) of the MD-stretching machine. The sheet which has been heated up to 126°C by the 2 roll (first heating roll) is heated as shown in Figure 5 by the 2′ roll. The temperature rise is nearly completed within 6–7 s after the sheet touches the 2′ roll. The sheet/roll

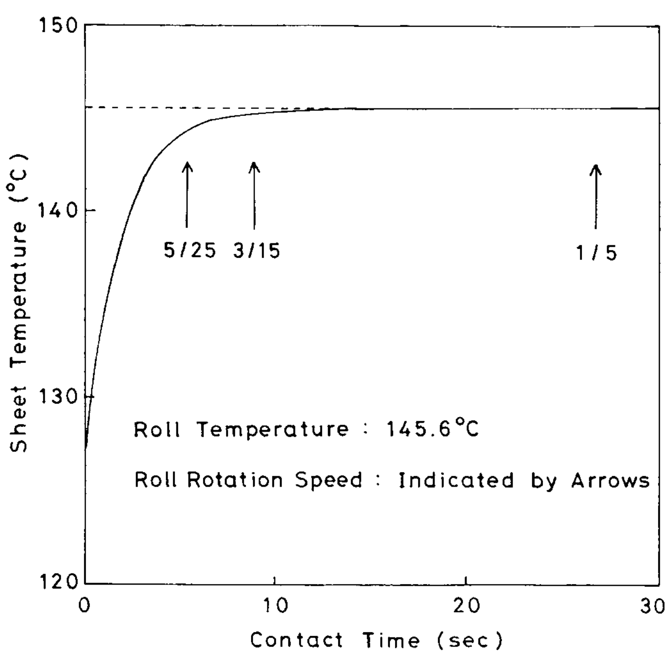

Fig. 5. Variation of sheet temperature with roll contact time. Roll temperature: 145.6°C; roll-rotation speed: indicated by arrows.

touching time for each roll rotation speed is shown by arrows in Figure 5. In the case of the roll rotation speed of 1/5, the sheet is held at the roll temperature for about 20 s after it has been heated to sufficient temperature. In the case of 3/15, the sheet is stretched just as the temperature rise is nearly completed. In the case of 5/25, the time to heat the sheet is slightly insufficient. In the case of 1/5, since the sheet is held at the roll temperature for about 20 s after it has been heated to the temperature, large segments of the β crystals have been melt-recrystallized, the molten part has grown small, and the little collapse of the molten part has occurred at the time of stretching, hence the surface roughness of the stretched sheet is low. When the heating temperature is high, a high roughness can be obtained even in the case of a low roll rotation speed since the recrystallization after melting of the β crystals is slow[1].

TD Stretching

Figure 6 shows the variations, with the MD-stretching roll-setting temperature, of the surface states of the BOPs obtained from various PPs under conditions of a MD-stretching roll-rotation speed of 3/15 and a TD-stretching temperature of 150°C. At each MD-stretching temperature, the surface roughness of BOP tends to increase with increasing MFI of the base PP. However, for the BOP obtained from FA150 (MFI = 9.8 dg/min), many voids are observed on the surface and the surface state is not good. From the viewpoints of the β-crystal content in the cast sheet and the surface state of the BOP, the base PP with MFI ≒ 8 dg/min (ex. FC140) is the best for the roughening of BOP.

Figure 7 shows an example of the variation, with the MD-stretching roll-rotation speed of the surface state of the BOP obtained by MD stretching the FC140 cast sheet at a heating roll setting temperature of 180°C and then by TD stretching the MD-stretched sheet at a temperature of 150°C. The roughnesses of the BOPs obtained at a roll-rotation speed of 1/5 are low. Comparison of the photographs in Figure 7 with those of FC140 in Figure 6 indicates that the MD-stretching at a high roll rotation speed (ex. 5/25) corresponds to that at a low roll-setting temperature.

Figure 8 shows the dependences, on the MD-stretching heating roll temperature, of R_a's and hazes of the BOPs obtained by MD stretching the cast sheets at various roll-rotation speeds and then by TD stretching the MD-stretched sheets at 150°C. R_a and haze show similar tendencies and increase with increasing surface temperature of the MD-stretching roll. Although R_a and haze also tend to increase with decreasing roll temperature in the low temperature range, these increases in R_a and haze are supposed to originate from the ground roughness other than craters and the roughness state is not very good. Compared at the same MD-stretching temperature, for the most part, R_a and haze are higher when the MFI of the base PP is higher. As for the influence of the roll-rotation speed, R_a and haze of a BOP stretched at a high rotation speed shift to low temperature side and the roughness of the UOP (Fig. 3) is inherited intact. This behavior has been discussed in the previous section from the viewpoint of the melt-recrystallization of the β crystals.

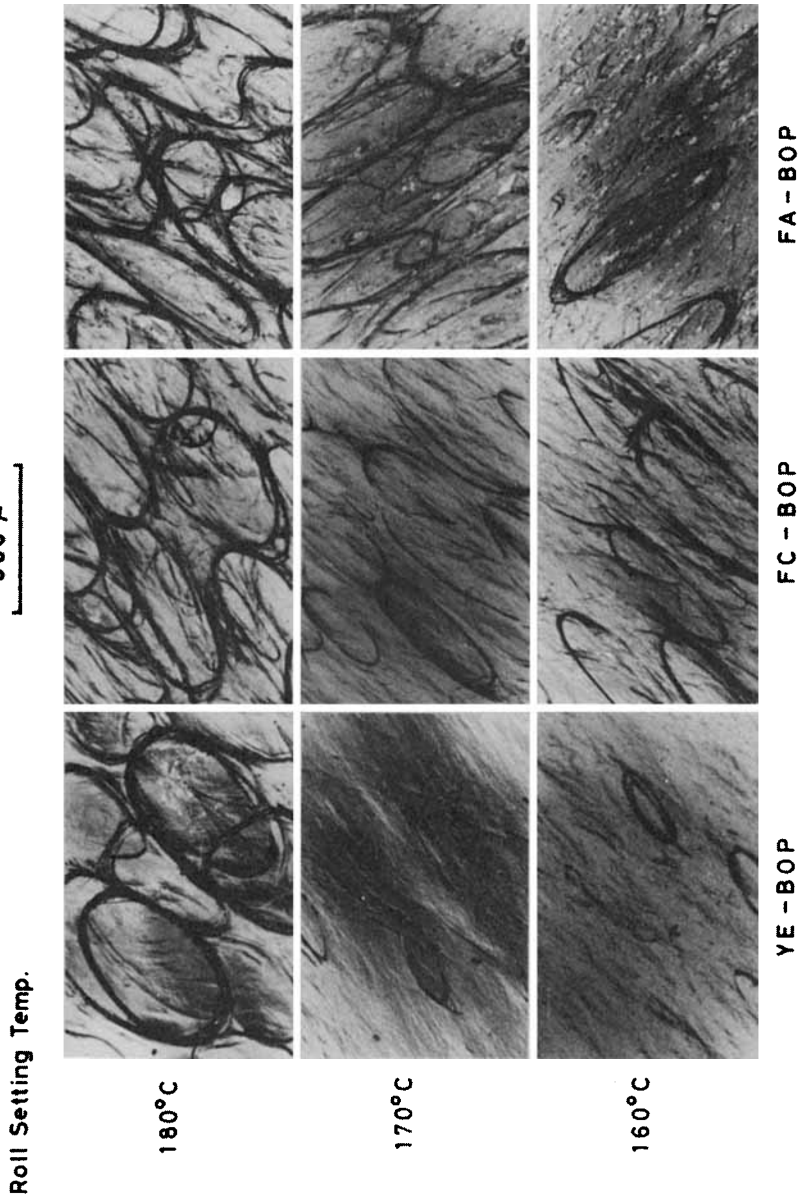

Fig. 6. Variations of surface roughness of BOP with MD-stretching roll-setting temperature. Roll-rotation speed: 3/15, TD-stretching temperature: 150°C.

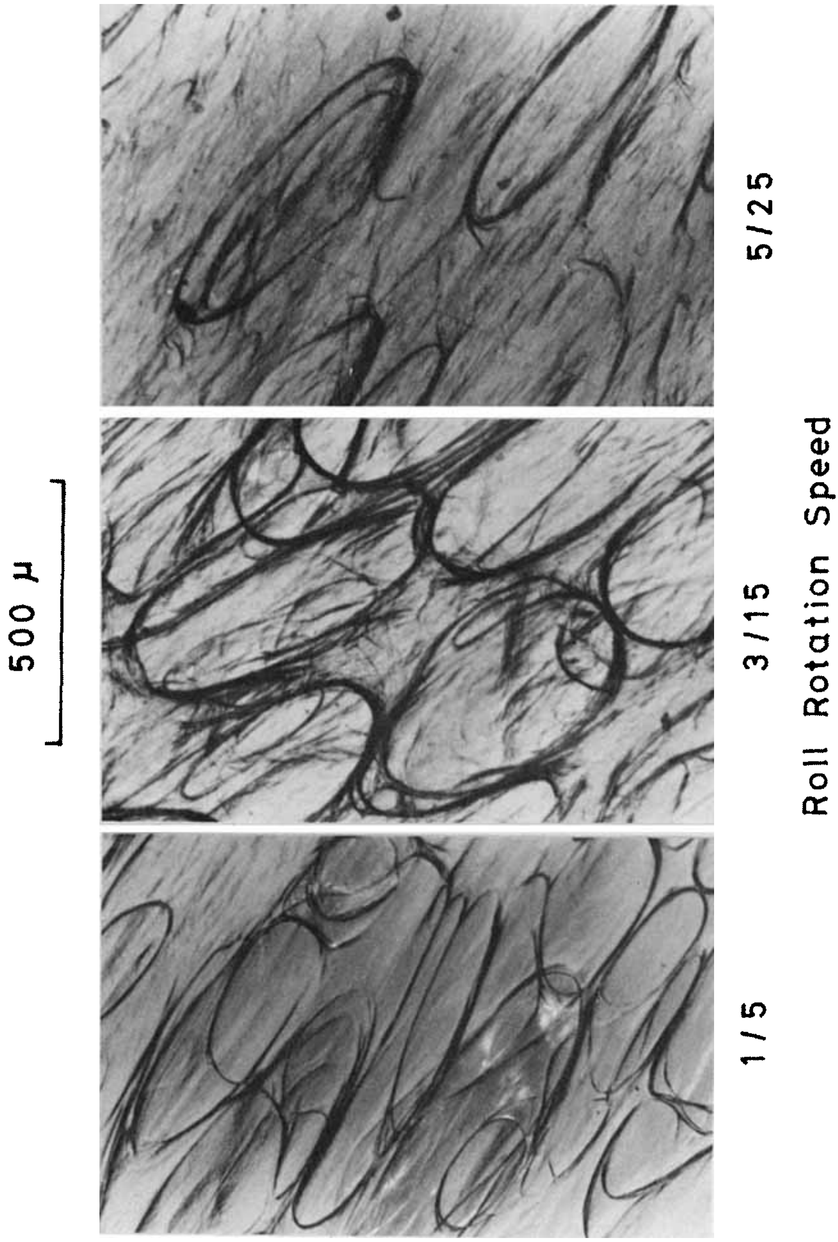

Fig. 7. Variation of surface roughness of BOP with roll-rotation speed. FC-UOP, roll-setting temperature: 180°C, TD-stretching temperature: 150°C.

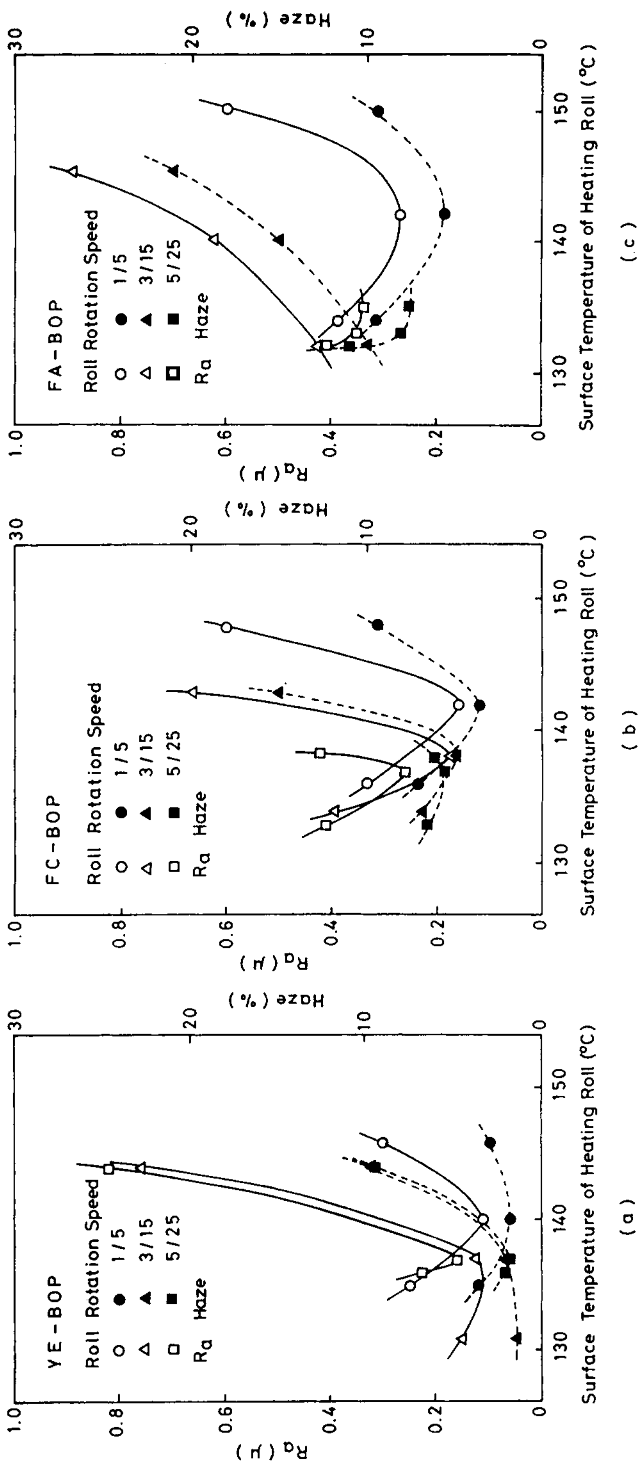

Fig. 8. Dependences of average roughness, R_a, and haze on heating roll temperature. Roll rotation speed: (○, ●) 1/5; (△, ▲) 3/15; (□, ■) 5/25.

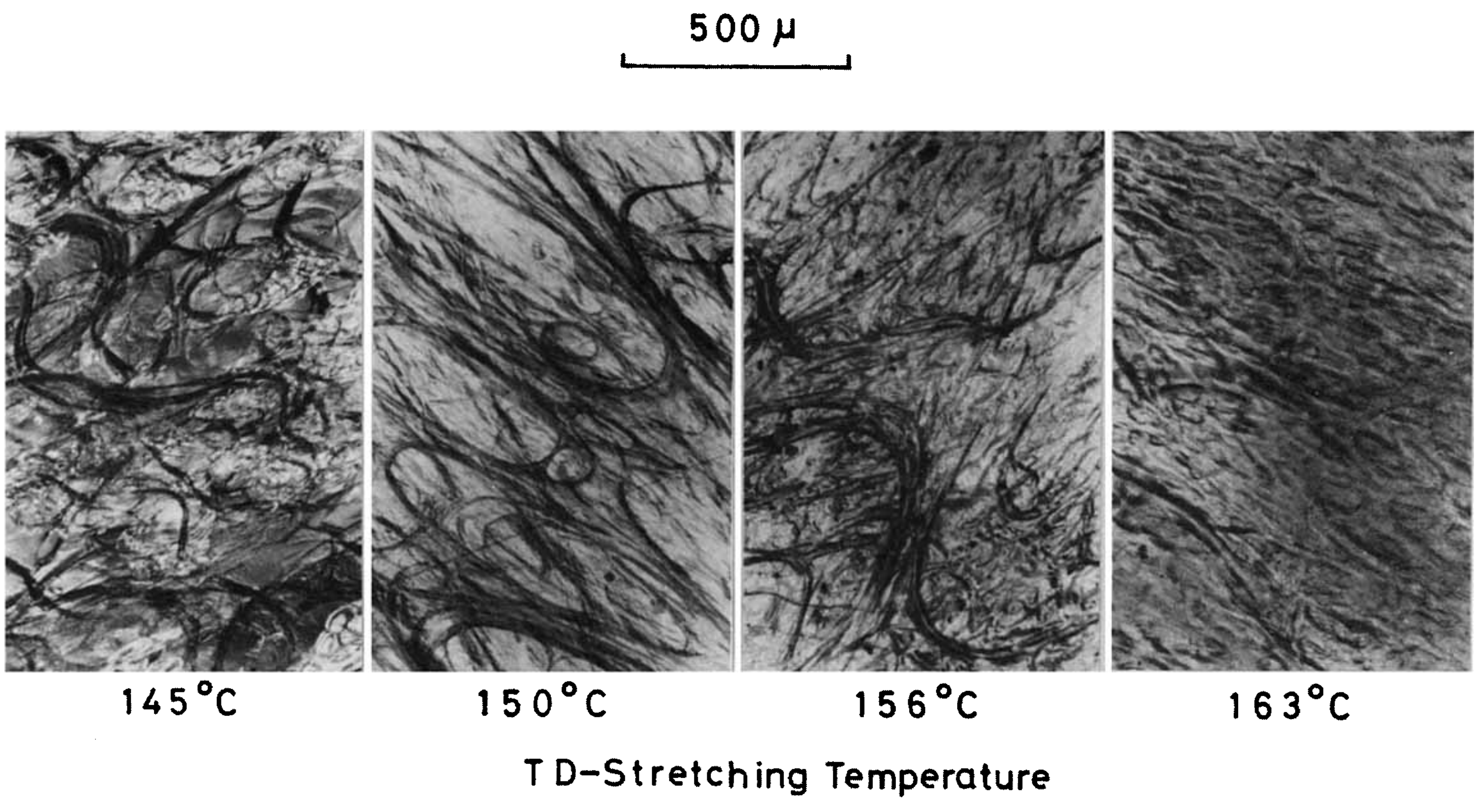

Fig. 9. Variation of surface roughness of BOP with TD-stretching temperature. FC-UOP, roll setting temperature; 180°C, roll rotation speed; 3/15.

Figure 9 shows examples of the surface photographs of the BOPs obtained by MD stretching the FC140 cast sheet at a heating roll-setting temperature of 180°C and a roll-rotation speed of 3/15 and then by TD stretching the MD-stretched sheet at various temperatures. The film broke at TD-stretching temperatures below 145°C. The surface state was not uniform, clear craters were not formed, and the ground became rough at a TD-stretching temperature of 145°C. Good rough-surface BOPs with a clear crater-like roughness were obtained at 150–155°C. A sign of melting was observed at 156°C and the film melted and broke at 163°C. The photograph of 163°C was taken on a section of a melt-broken film.

CONCLUSIONS

The influence of MFI of the base PP on the roughening of BOP by a successively stretching tenter method has been studied. The amount of the β crystals formed in the cast sheet was maximum when a PP with MFI $\risingdotseq$ 8 dg/min was used as a raw material and the BOP obtained from it was a good rough-surface film with the clearest crater-like roughness. As for the MD-stretching conditions, the roughness of the BOP increased with increasing the MD-stretching temperature, but the probability of melt-breaking of the sheet at the time of the MD stretching became higher at MD-stretching temperatures above 150°C. As for the roll-rotation speed, the best speed was obtained from the balance of the thermal conductivity of the sheet and the melt-recrystallization rate of the β crystals. As for the TD-stretching conditions, stretching temperatures of 150–155°C were suitable. When the stretching temperature was too low, although the roughness of the BOP became considerably high, the craters on it became unclear, the ground became rough, and hence a BOP with a good surface state could not be obtained. When the stretching temperature was still lower, the film broke at the time of stretching. When the stretching temperature was too high, the craters on the film melted, and hence a good film could not be obtained. When the stretching temperature was still higher, the film melted and broke. Highly roughened BOPs with a crater-like roughness can be obtained by appropriately selecting the MFI of the base PP and MD- and TD-stretching conditions.

References

1. M. Fujiyama, Y. Kawamura, T. Wakino, and T. Okamoto, *J. Appl. Polym. Sci.*, **36**, 995 (1988).
2. T. Yamane, M. Tanaka, and E. Wada, Japan Kokai Tokkyo Koho, 80-9806 (1980).
3. A. Turner-Jones, J. M. Aizlewood, and D. R. Beckert, *Makromol. Chem.*, **75**, 134 (1964).
4. M. Fujiyama, Y. Kawamura, T. Wakino, and T. Okamoto, *J. Appl. Polym. Sci.*, **36**, 985 (1988).

Received August 11, 1987
Accepted November 21, 1987