# Study on Rough-Surface Biaxially Oriented Polypropylene Film. VI. Roughening by Laminated Cast Sheet

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**Synopsis**

When the roughening of biaxially oriented polypropylene film (BOP) is performed by forming $\beta$ crystals in a monolayer cast sheet, occasionally voids are formed in the BOP. In order to prevent this and to roughen both sides of the BOP, rough-surface BOPs were obtained by casting two-resin, three-layer sheets consisting of two surface layers with a high-melt flow index (MFI) polypropylene (PP) and a core layer with a low-MFI PP, by stretching the cast sheets five times in the machine direction (MD) with a roll-type stretching machine, and by stretching the MD-stretched sheets 10 times in the transverse direction (TD) with a pantograph-type stretching machine. Comparatively large spherulites were observed in the surface layers of the cast sheets and structural differences between the surface layers and core layer were recognized. Both sides of the cast sheets could be roughened. The influence of processing conditions on the roughness state of BOP was generally similar to that in the case of monolayer cast sheet.

## INTRODUCTION

The roughening of biaxially oriented polypropylene film (BOP) by utilizing $\beta$-form crystals in a successively stretching tenter method has been studied.[1-5] As a result, the relations among processing conditions, the surface states of the BOP, and the properties of the BOP in the roughening of monolayer cast sheet could systematically be analyzed.[5]

Roughening of the BOP involves the surface. Accordingly, the $\beta$ crystals formed in the cast sheet should exist in the surface region, and when they exist in the inner region, there is a possibility that they create voids. Specifically, it is practically desirable that the $\beta$ crystals exist only in the surface region of the cast sheet. Furthermore, it is considered that if a cast sheet with surface layers containing a large amount of $\beta$ crystals on both sides is used, roughening of both sides of BOP would be possible. Based on the above reasons, roughening of BOP using laminated cast sheets consisting of two surface layers with a large amount of $\beta$ crystals and a core layer with no $\beta$ crystals was studied and the roughness states of BOPs obtained were analyzed. Furthermore, considering the roughening mechanism, there is the possibility that a BOP with more uniform roughness could be obtained in such case. The roughness states of the BOPs obtained were compared with those obtained from monolayer sheets.

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TABLE I
Formulations of Raw Materials

| Sample code | Layer | Base PP Grade | Base PP MFI (dg/min) | γ-Quinacridone (ppm) | BHT (wt%) | Irganox 1010 (wt%) | CaSt (wt%) |
|---|---|---|---|---|---|---|---|
| CE-β-0 | Surface layer | FC140 | 7.8 | 0 | 0.2 | 0.1 | 0.015 |
| CE-β-1 | Surface layer | FC140 | 7.8 | 1 | 0.2 | 0.1 | 0.015 |
| — | Core layer | FB110 | 1.3 | 0 | 0.2 | 0.1 | 0.015 |

Nakamura et al.[6] and Yamane et al.[7] claim in their patents that biaxially stretching a two-layer sheet or a two-resin, three-layer sheet improves its drawability and mechanical and electric properties of the rough-surface BOP obtained from it.

## EXPERIMENTAL

### Raw Resin

Polypropylenes (PP) used for the surface layers and the core layer are shown in Table I. It has been ascertained from the dependence of the amount of the β crystals formed in the cast sheet on the melt flow index (MFI) of the base PP that the MFI most favorable for the formation of the β crystals is about 8dg/min.[1,3] Accordingly, in order to form β crystals in the surface layers only, a high-MFI PP was used for CE-β-0 and CE-β-1, and 1 ppm of γ-quinacridone, a β-crystal nucleator, was added to CE-β-1. The purpose of adding the β-nucleator to CE-β-1 was to study the amount of the β crystals formed in the cast sheet and at the same time to study the control of the roughness state.[5] BHT, Irganox 1010, and calcium stearate (CaSt) are thermo-oxidative stabilizers. Each raw material was formulated as shown in Table I, mixed with a Supermixer, and extruded with a 65 mm ϕ extruder at an extrusion temperature of 250°C into a strand which was cut into pellets of about 3 mm size with an automatic cutter.

### Casting of Sheet

Two-resin, three-layer sheets were cast from the pellets with two extruders; a 60 mm ϕ extruder for the surface layers, and a 65 mm ϕ extruder for the core layer, equipped with a 600 mm wide two-resin, three-layer coextrusion T die. Casting conditions were as follows: an extrusion temperature of the surface layers of 250°C, an extrusion temperature of core layer of 290°C, and a chill roll temperature of 70°C. The coextrusion apparatus is shown in Figure 1.

The amount of the β crystals formed in the cast sheets was measured by x-ray diffraction, and the spherulitic morphologies of the surface and core layers of the cast sheets were observed with a polarizing microscope.[4]

### MD Stretching

The cast sheets were stretched in the machine direction (MD) 5 times with a roll-type stretching machine shown in Figure 1 of Part II of this series[2]

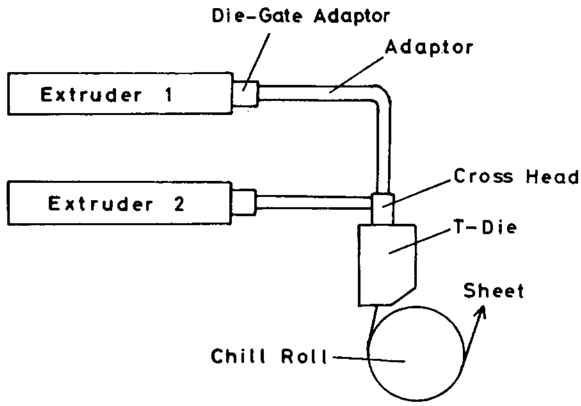


Fig. 1. Two-resin, three-layer coextrusion apparatus.

under the following conditions: preheating oven temperature, 140°C; setting temperature of heating rolls, 155°C, 160°C, and 170°C; roll rotation speed,[2] 3/15. It was already known that when the MD-stretching is carried out with this stretching machine roughening occurs on the surface of the cast sheet which touched the 2′ roll.[2] Since the laminated cast sheets in the present experiment were set so that both sides could be roughened, the MD stretchings were carried out by passing each sheet two ways in order to roughen each side: first one side of each sheet touched the 2′ roll and then the other side of the sheet touched the 2′ roll.

### TD Stretching

The MD-stretched sheets were stretched in the transverse direction (TD) 10 times with a pantograph-type stretching machine[2] under conditions of a preheating time of 1 min, a stretching temperature of 155°C, and a stretching rate of 2500%/min.

The roughness states of the BOPs obtained were analyzed with a reflection-type differential interference microscope and a Surfcorder. The factors measured were the average roughness, $R_a$, and the maximum roughness, $R_m$, which represent the depth of the roughness, and the roughness period, $L$, the crater diameter, $D$, and the crater density, $d$, which represent the density of the roughness.[5]

## RESULTS AND DISCUSSION

### Analysis of Cast Sheet

Polarized micrographs of the cast sheets obtained by the coextrusion are shown in Figure 2 and their analytical results are shown in Table II.

It can be seen from Figure 2 that comparatively large spherulites are formed in the surface layers of the cast sheets. This is assumed to be due to the fact that the MFI of the base PP of the surface layers was comparatively high. The diameter of the $\beta$ spherulites in the surface layers is large for CE-$\beta$-0, which does not contain the $\beta$ nucleator, and small for CE-$\beta$-1 which does contain the $\beta$ nucleator. Furthermore, the diameter of the $\beta$ spherulites

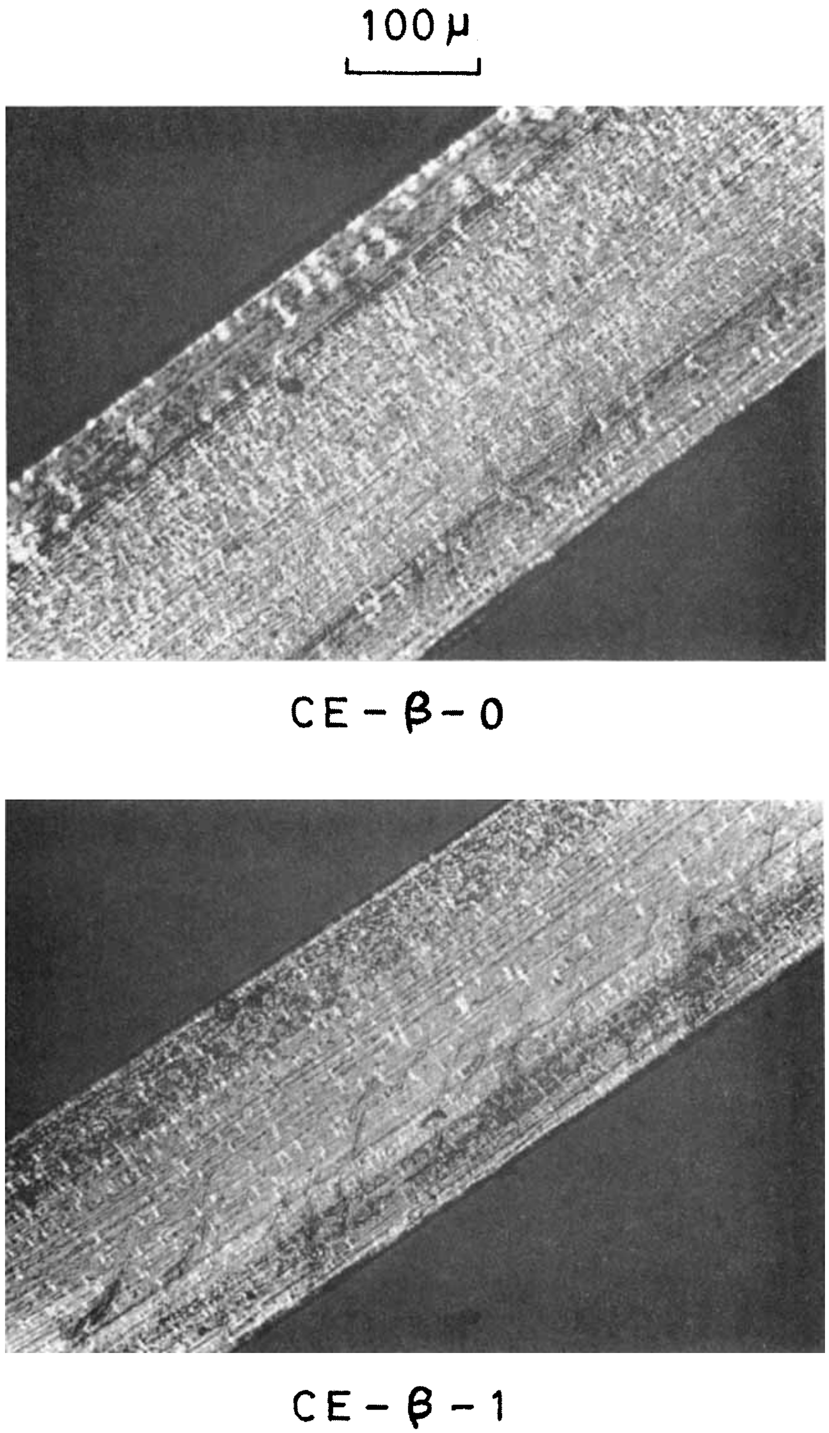


Fig. 2. Polarized micrographs of cast sheet. Lower layers are chill roll side.

TABLE II
Analysis of Cast Sheets

| Sample | β-Nucleator content in surface layer (ppm) | Layer construction (μm) FC140/FB110/FC140 (Total) | β-Spherulite diameter in surface layer (μm) | | K value |
|---|---|---|---|---|---|
| | | | Chill roll side | Opposite side | |
| CE-β-0 | 0 | 83/215/83 (381) | ca. 7 | 20–35 | 0.19 |
| CE-β-1 | 1 | 69/146/69 (284) | < 4 | < 7 | 0.20 |

in the chill roll-touched surface layer is smaller than that in the opposite layer. This is because the chill roll-touched surface layer was cooled more rapidly than the opposite layer.

The K value (β-crystal content) measured by the x-ray diffraction is about 0.20 for both sheets and the increase in the β-crystal content by the addition of the β nucleator is not seen. The β-crystal content is influenced most by the extrusion temperature.[1,2] Since the extrusion temperature of the surface layers was set slightly higher in the present experiment, the influence of the addition of the β nucleator on the β-crystal content did not markedly appear.

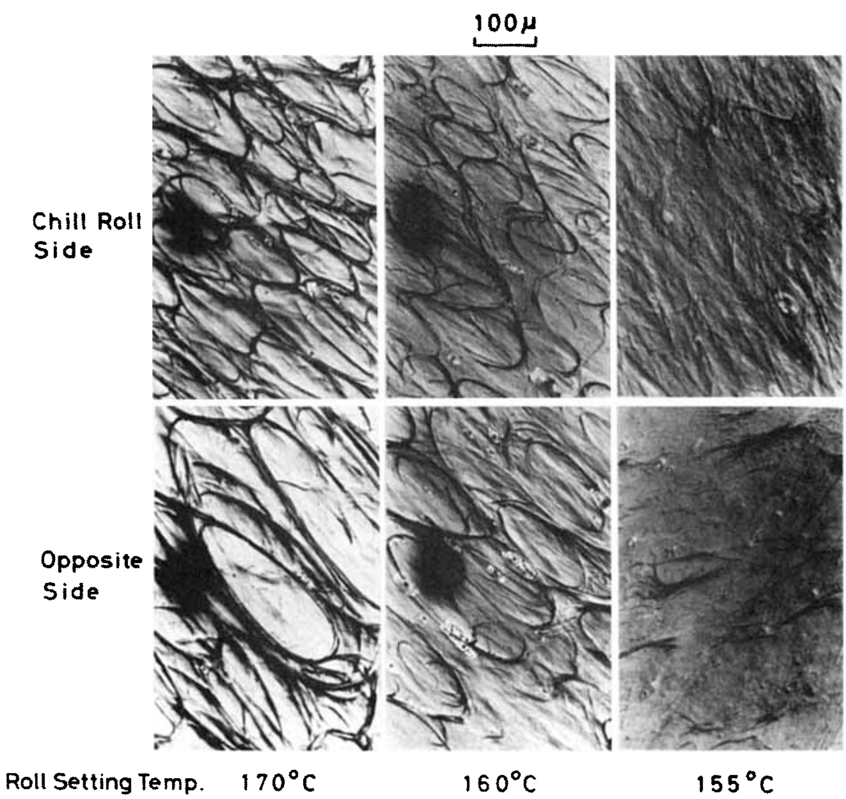


Fig. 3. Photographs of surfaces of BOPs made from CE-β-0.

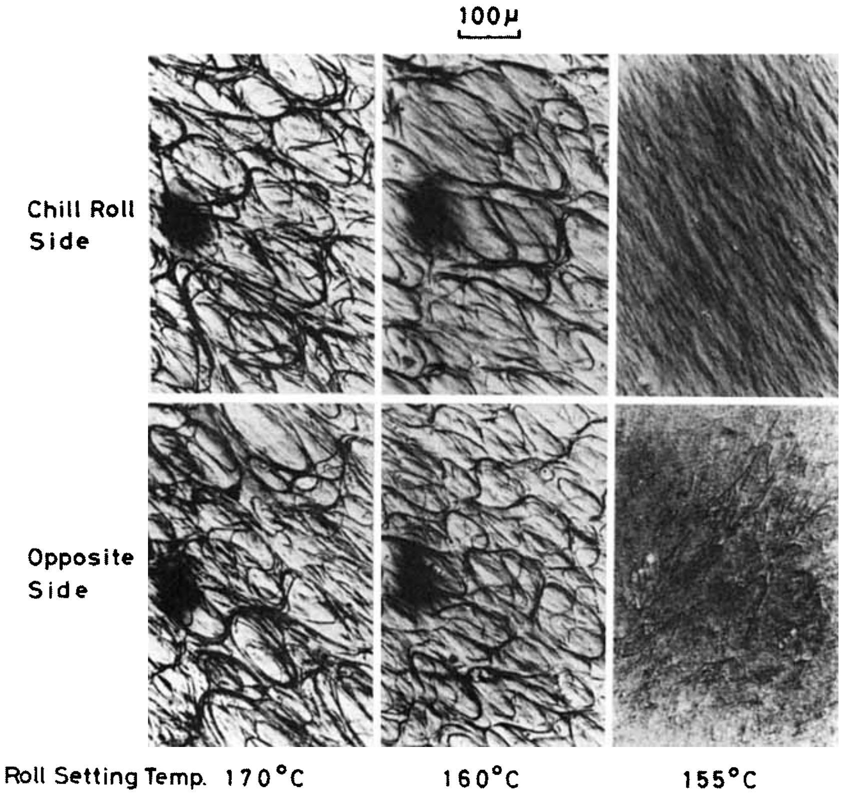


Fig. 4. Photographs of surfaces of BOPs made from CE-β-1.

## Analysis of Roughness State of BOP

### *Observation of Surfaces of BOPs*

Figure 3 shows the surface micrographs of the rough-surface BOPs obtained from the CE-β-0 cast sheet. Both sides of the sheet could be roughened and the diameter of the craters on the chill roll-touched surface is smaller than that on the opposite surface. This is due to the fact that the diameter of the β spherulites in the chill roll-touched layer is smaller than that in the opposite layer. The roughness of the BOP is higher as the MD-stretching temperature is higher: the same tendency is seen in the case of the monolayer cast sheet.[3]

Figure 4 shows the surface micrographs of the rough-surface BOPs obtained from the CE-β-1 cast sheet. Both sides of the sheet could be roughened in this case too. However, it differs from the case of the CE-β-0 cast sheet in that the diameter of the craters on the chill roll-touched surface is nearly equal to that on the opposite surface. The diameter of the craters formed on the BOP is proportional to the diameter of the β spherulites in the cast sheet and the diameter of the β spherulites in the cast sheet can be controlled by the amount of the β nucleator added to the base PP.[4,5] Since CE-β-1 was added to the β nucleator, the diameters of the β spherulites in both the chill roll-touched surface layer and the opposite layer in the cast sheet are small and nearly equal as shown in Figure 2 and Table II. Accordingly, it is assumed that the diameters of the craters on both the surfaces of the BOPs obtained from it are small and nearly equal. The roughness of the BOP is higher as the MD-stretching temperature is higher, which is similar to that in the case of monolayer cast sheet.

### *Analysis of Roughness State*

The data which represent the roughness states of the BOPs obtained are shown in Table III. The dependence of these data on the MD-stretching temperature, which most affects the roughness state of the BOP, is shown in Figure 5.

Figure 5(a) indicates that the average roughness, $R_a$, increases with increasing MD-stretching temperature, which agrees well with the results of the monolayer sheets.[2-5] On the other hand, the roughness period, $L$, decreases with increasing MD-stretching temperature, which differs from the result of the monolayer sheet.[5] Since the extrusion and chill roll temperatures deviated from the best conditions in the present experiment, both the amount and diameter of the $\beta$ spherulites formed in the surface layers of the cast sheet were small. Accordingly, in the case of low MD-stretching temperature, the recrystallization after melting of the $\beta$ crystals at the time of the MD stretching was easy to proceed and easy to be affected by the MD-stretching temperature, and hence the above mentioned result was obtained.

Figure 5(b) indicates that the crater diameter, $D$, increases with increasing MD-stretching temperature independent of the addition of the $\beta$ nucleator and of the sheet surfaces. The comparison between the samples indicates that while there is almost no difference in $D$ between CE-$\beta$-0 and CE-$\beta$-1 on the chill roll-touched surface, $D$ of CE-$\beta$-1 is smaller than that of CE-$\beta$-0 on the opposite surface. While the crater density, $d$, increases with increasing MD-stretching temperature on the chill roll-touched surface, it decreases with increasing MD-stretching temperature on the opposite surface. The comparison between samples indicates that $d$ of CE-$\beta$-1 is lower than that of CE-$\beta$-0 on both the surfaces.

### *Comparison with Rough-Surface BOP Obtained from Monolayer Sheet*

Figure 6 shows the relation between the maximum roughness, $R_m$, and the average roughness, $R_a$, of the BOPs obtained from the monolayer and laminated cast sheets. A good correlation exists between $R_m$ and $R_a$ of the rough-surface BOPs obtained from the monolayer cast sheet,[5] and the points of the rough-surface BOPs obtained from the laminated cast sheet are also almost on the regression line. Accordingly, it is assumed that the roughness state of the rough-surface BOP obtained from the laminated cast sheet is almost the same as that obtained from the monolayer cast sheet; and, contrary to the expectation, it cannot be said that the former has a more uniform roughness than the latter.

TABLE III
Analysis of Roughness of BOPs

| Sample | Roughened surface | MD-stretching roll temperature (°C) | | Crater diameter $D$ (μm) | Crater density $d$ (number/cm$^2$) | Average roughness $R_a$ (μm) | Maximum roughness $R_m$ (μm) | Roughness period $L$ (μm) |
|---|---|---|---|---|---|---|---|---|
| | | Setting | Surface | | | | | |
| CE-β-0 | C[a] | 170 | 144 | 100 | 11,000 | 0.41 | 4.5 | 160 |
| | O[a] | 170 | 146 | 210 | 4,500 | 0.58 | 6.0 | 230 |
| | C | 160 | 139 | 90 | 8,500 | 0.18 | 2.1 | 380 |
| | O | 160 | 139 | 160 | 6,100 | 0.23 | 2.1 | 260 |
| | C | 155 | 135 | — | — | 0.09 | 1.7 | 560 |
| | O | 155 | 134 | — | — | 0.08 | 1.9 | 560 |
| CE-β-1 | C | 170 | 145 | 100 | 20,000 | 0.37 | 5.1 | 180 |
| | O | 170 | 147 | 100 | 18,000 | 0.41 | 4.4 | 200 |
| | C | 160 | 140 | 90 | 11,000 | 0.12 | 0.9 | 290 |
| | O | 160 | 139 | 90 | 24,000 | 0.17 | 1.8 | 250 |
| | C | 155 | 134 | — | — | 0.07 | 1.3 | 690 |
| | O | 155 | 135 | — | — | 0.11 | 1.3 | 560 |

[a] C, chill roll side; O, opposite side.

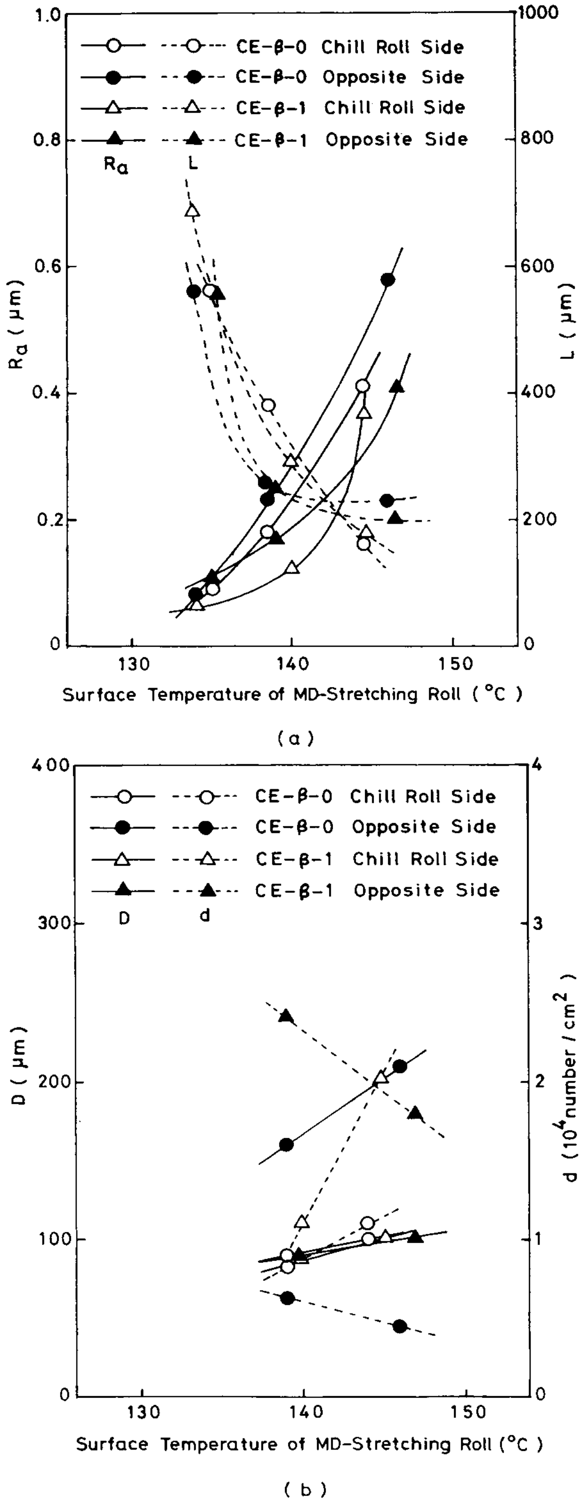


Fig. 5. Dependence of (a) average roughness, $R_a$, and roughness period, $L$, and (b) crater diameter, $D$, and crater density, $d$, on surface temperature of MD-stretching roll. (○) CE-β-0, chill roll side; (●) CE-β-0, opposite side; (△) CE-β-1, chill roll side; (▲) CE-β-1, opposite side.

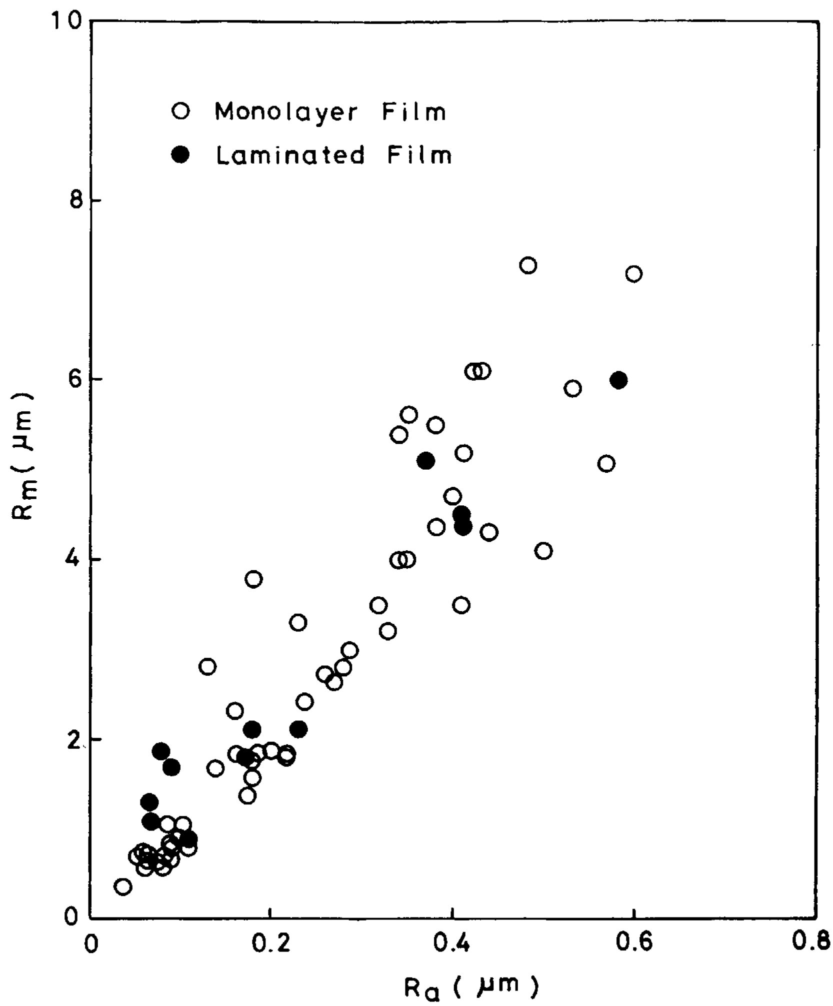


Fig. 6. Relation between maximum roughness, $R_m$, and average roughness, $R_a$. (○) Monolayer film; (●) laminated film.

## CONCLUSIONS

The roughening of BOPs by using laminated cast sheets obtained by coextrusion has been studied and rough-surface BOPs with a good roughness state were obtained. Analysis of the cast sheets indicated that comparatively large $\beta$ spherulites were formed in surface layers and the amount of the $\beta$ spherulites formed was about 0.2 in K value. The influence of stretching conditions on the roughness state of the BOP was generally the same as in the case of the monolayer cast sheet.

In the present experiment, the roughening of BOP was carried out successively on each surface, and a sufficient roughening could be achieved at both surfaces. Accordingly, it might be possible to roughen simultaneously both surfaces if appropriate conditions are selected: by increasing the number of the preheating rolls and by raising the line speed.

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