Study on Rough-Surface Biaxially Oriented Polypropylene Film. VII. Roughening by Blending Low Melting Point Polymers

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Synopsis

The roughening of biaxially oriented polypropylene film using polypropylene (PP) blended with low melting point polymers such as polybutene-1 and high-density polyethylene as raw resin in a successively stretching tenter method has been studied for the same reasons as has the roughening of BOP utilizing β crystals. Rough-surface BOPs with a low depth and a high density of roughness were obtained by using the blended PPs as raw resins. By combining this roughening method with the roughening method utilizing the β crystals, rough-surface BOPs with a crater-like large roughness and a fine roughness on the ground were obtained. These rough-surface BOPs showed excellent impregnation properties of insulating oils.

INTRODUCTION

Roughening of biaxially oriented polypropylene film (BOP) utilizing β -form crystals in a successively stretching tenter method has been studied previously.¹⁻⁶ The principle is as follows: By utilizing the difference of the melting points of the α -form crystals, which is the usual crystal form of polypropylene (PP) and the β -form crystal, which is formed under particular conditions, both of which constitute a casted sheet, the sheet is stretched at a temperature between the melting point of the β -form crystals (147°C) and that of the α -form crystals (164°C) and the β crystal parts are melt-collapsed. Accordingly, it is considered that a rough-surface BOP can also be obtained by stretching a PP sheet which contains low melting point polymers such as polybutene-1 (PB-1) and high-density polyethylene (HDPE). Then, with regard to the roughening of BOP by blending low melting point polymers, the following two methods have been studied.

First, a low-melt flow index (MFI) PP which contained PB-1 or HDPE was cast into a sheet under conditions where no β crystal was formed, which was subsequently biaxially stretched.

Next, a high-MFI PP which contained PB-1 or HDPE was cast into a sheet under conditions where β crystals were formed, which was subsequently biaxially stretched.

As for the roughening method of BOP by stretching sheets which contain different polymers, several patents have been applied for,⁷⁻¹³ but no scientific paper has appeared as far as the authors know.

EXPERIMENTAL

Roughening Using Low-MFI PP Blends

The low-MFI PP used was Tokuyama Polypro Grade FB110 which was a homoisotactic PP produced in the form of pellets by Tokuyama Soda Co., Ltd. PB-1 and HDPE were also in the form of pellets. The recipes and properties of the raw materials are shown in Table I. These raw materials were mixed in a tumbler, and were cast into sheets 800 μ m thick with a 65 mm extruder equipped with a 500 mm wide T die at an extrusion temperature of 250°C and a chill roll temperature of 60°C. The cast sheets were stretched in the machine direction (MD) 5 times with a roll-type stretching machine (Fig. 1, ref. 2; Part II of this series) under the following conditions: preheating oven temperature, 140°C; setting temperature of heating rolls, 140°C and 170°C; roll rotation speed,² heating rolls (low speed)/cooling rolls (high speed) = 3 m/min/15 m/min (3/15). The MDstretched sheets were stretched in the transverse direction (TD) 10 times with a pantograph-type stretching machine² under conditions of a preheating time of 1 min, a stretching temperature of 150°C, and a stretching rate of 3000% / min.

The amount of β crystals formed in the cast sheets was measured by X-ray diffraction. Roughness states of the BOPs obtained were studied by the observation of the surfaces with a reflection-type differential interference microscope and by measurements of haze with a haze meter and of the average roughness, R_a and the roughness period, L, with a Surfcorder.⁵

Roughening Using High-MFI PP Blends

The high-MFI PP used was Tokuyama Polypro Grade FC 140 which was a homoisotactic PP produced in the form of pellets by Tokuyama Soda Co., Ltd. The recipes and properties of raw materials are shown in Table I. These raw materials were mixed in a tumbler, and were cast into sheets 800 µm thick with a 65 mm extruder equipped with a 500 mm wide T die at an extrusion temperature of 220°C and a chill roll temperature of 90°C. The cast

Recipes of Raw Materials									
Sample code	Base PP		Blended resin						
	Grade	MFI (dg/min)	Kind	Melting point (°C)	MFI (dg/min)	Content (wt%)	K value		
FB-PB-1-20	FB110	1.5	PB-1	129	0.4	20	0		
FB-PB-1-40	FB110	1.5	PB-1	129	0.4	40	0		
FB-PB-1-60	FB110	1.5	PB-1	129	0.4	60	0		
FB-HDPE-20	FB110	1.5	HDPE	132	1.0	20	0		
FB-HDPE-40	FB110	1.5	HDPE	132	1.0	40	0		
FC-PB-1-20	FC140	7.8	PB-1	129	0.4	20	0.05		
FC-HDPE-20	FC140	7.8	HDPE	132	1.0	20	0.02		

TABLEI

sheets were stretched in the MD 5 times with a roll-type stretching machine (Fig 1, ref. 2; Part II of this series) under the following conditions: preheating oven temperature, 140°C; setting temperature of heating rolls, 175°C; roll rotation speed,² 3/15. The MD-stretched sheets were stretched in the TD 10 times with a pantograph-type stretching machine² under conditions of a preheating time of 1 min, a stretching temperature of 150°C, and a stretching rate of 3000%/min.

Analysis of the cast sheets and evaluation of the roughness states of the BOPs were carried out in the same manner as in the case of the low-MFI PP blends.

RESULTS AND DISCUSSION

Analysis of Cast Sheet

The results of the X-ray diffractions of the cast sheets are shown in Table I. No β crystal is formed in the sheets cast from the blends of low-MFI PP(FB110) and PB-1 or HDPE. Slight β crystals are formed in the sheets cast from the blends of the high-MFI PP(FC140) and PB-1 or HDPE. The difference in the amount of the β crystals formed between the two cast sheets originates from the difference in the MFI of the base PP³ and the casting conditions.^{1,2}

Roughening Using Low-MFI PP Blends

Figure 1 shows the dependences of the average roughnesses, R_a , and hazes of the BOPs obtained from the blends of the low-MFI PP and PB-1 or HDPE



Fig. 1. Dependences of average roughness, R_a , and haze of BOP on content of resin blended to low-MFI PP. (- \bigcirc -, -- \bigcirc --) PB-1, MD 170°C; (- \triangle --, -- \bigcirc --) HDPE, MD 170°C; (- \blacksquare --), -- \blacksquare ---) PB-1, MD 140°C; (- \blacksquare --, -- \blacksquare ---) HDPE, MD 140°C.





upon the blended resin content. In both the cases of PB-1 and HDPE, although R_a and haze tend to increase with increasing the content, all R_a 's are lower than 0.2 μ m and the degree of the roughness is not so high.

The surface photographs of the BOPs obtained from the low-MFI PP blended with 20 wt% PB-1 or HDPE are shown in the left side of Figure 2. In the case of the PB-1-blended PP, a very fine roughness is formed and its roughness frequency, L, is 60 μ m. In the case of the HDPE-blended PP, a roughness of slightly lower density is formed. In the roughening method of utilizing the β crystals, generally $L = 100 - 500 \ \mu$ m, and it is difficult to obtain a BOP with L lower than 100 μ m even if a crystallization nucleator is used.⁵ Accordingly, the rough-surface BOP obtained from the PB-1-blended low-MFI PP has a characteristic that R_a and L are low: very fine roughness.

The roughness of PB-1- or HDPE-blended PP forms differently, which is assumed to originate from the difference of the microdomain structure of each blend system.

Roughening Using High-MFI PP Blends

In the roughening of BOP utilizing the β crystals, BOPs with crater-like roughness are obtained.²⁻⁶ However, when the craters are large and the degree of roughness of the BOP is high, there is a case where voids remain during the



Fig. 3. Schematic representation of rough-surface BOP made from PB-1 or HDPE-blended high-MFI PP. 1; Ground roughness; 2, crater; D, crater diameter; H, height of crater flange; W, width of crater flange; X, period of ground roughness; h, height of ground roughness.

Sample	Average roughness $R_a(\mu m)$	Roughness period L(µm)	Impregnation rate (cm/h ^{1/2})	Impregnation state ^a
FC-PB-1-20	0.39	240	5.3	A
ibid	0.18	150	4.7	А
FC-HDPE-20	0.20	85	4.7	А
ibid	0.22	100	4.7	Α
FC-β-1 ^b	0.31	180	3.8	В
$FC-\alpha-1^{b}$	0.38	450	5.3	С

TABLE II Characterization and Impregnation Properties of Rough-Surface BOPs

^aA; Excellent, B; Good, C; Poor

^bRough-surface BOP made by utilizing β -form crystals.⁴

impregnation process of insulating oils. In order to improve this point, roughening utilizing the β crystals was combined with the roughening by the polymer blend.

The surface photographs of BOPs obtained from the high-MFI PP blended with 20 wt% PB-1 or HDPE are shown in the right side of Figure 2. Rough surfaces with a crater-like large roughness and a fine roughness on the ground are observed. Figure 3 shows a schematic representation of such BOP.

Table II shows the values of the average roughness, R_a , and the roughness period, L. L of the BOP obtained from the PB-1-blended PP is higher than that obtained from the HDPE-blended PP, which is due to the fact that the crater diameter of the former is larger than that of the latter. The impregnation properties of an insulating oil of these BOPs were measured⁵ and the results are shown in Table II. Comparing at an equivalent roughness, the impregnation properties of the BOPs obtained by combining with the polymer blend method are better than that obtained by utilizing only the β crystals.

CONCLUSIONS

Rough-surface BOPs with a low depth and a high density of roughness were obtained by using PB-1 or HDPE-blended PP as raw resin. Rough-surface BOPs with a crater-like large roughness and a fine roughness on the ground were obtained by combining this method with the method utilizing the β

FUJIYAMA ET AL.

crystals. The impregnation properties of an insulating oil of these BOPs were better than that of BOPs obtained utilizing only the β crystals.

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