Highly Transparent Polypropylene Sheet by using Optimum Screw Design and Multi-Layered Structure

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ABSTRACT

In order to improve the transparency of isotactic polypropylene sheet, extrusion screw geometry and a multilayer extrusion were conducted.

The transparency of melted resin sheet was obtained by screw geometry so that the specific energy consumption was small and the melted temperature was low. The screw geometry was designed to satisfy the target qualities, which were both high transparency and extrusion stability, by optimizing the geometry of the gently tapered compression screw with a torpedo type barrier and a gear wheel type mixer.

A multilayer extrusion was also conducted using a resin with lower melt viscosity for surface layers than the resin for the core layer. As a result, the number of spherulites near the surface decreased dramatically, and furthermore the internal haze of the sheet was improved. The number of spherulites in the sheet cross section and the internal haze correlated to each other. The flow velocity distribution, shear rate distribution, and shear stress distribution in the die lip section were calculated using the finite element method. The shear stress in the sheet thickness direction was reduced by laminating resin with low melt viscosity on both surfaces, and therefore it can be surmised that stress induced crystallization was restrained. Furthermore, the cooling simulation in the belt quenching process was conducted. The density of the number of spherulites was set as the response variable, and then the plateau area time of the temperature in each position of the thickness direction and the shear stress were chosen as the independent variables, and the prediction formula was derived by multiple regression analysis. Moreover, using the obtained formula, the density of several numbers of spherulites in various surface layer ratios were predicted, and optimization of multilayer composition was proposed.

1. INTRODUCTION

Polypropylene is an excellent resin from the view point of it having a reasonable price, good physical properties and good recycling features etc. However, because it is a crystalline resin, it is rather difficult to obtain a highly transparent sheet, and has had limited development with regards to usage where high transparency is required. In order to obtain a highly transparent polypropylene sheet, a random polypropylene as a base material and various kinds of nuclear agents were used. Also, a quick quenching system and a rolling process were adopted for the sheeting process.

In order to improve the internal haze of the sheet, the control of the degree of crystallization is very important. A quick quench process by using a water-bath is one of the most effective methods[1)]. To obtain a highly transparent appearance with an excellent gloss, it is necessary to decrease the surface roughness of the melted sheet before solidification.

There has been a lot of research in the past regarding the unstable flow phenomenon of melted polymer that obstructs transparency[2~6)]. In the latest research, it is reported that "Shark skin" is a phenomenon in which only the surface of melted resin film becomes rough. The cause of it is a rapid change from the shear stress at the wall vicinity in the die land to extension stress in the die exit section[7)]. However, even in the state that the critical range of shear stress is not exceeded, the transparency of melt web depends on the screw geometry of the extruder.

Therefore, transparency should be discussed from another viewpoint.

In this paper, an experimental analysis of the extrusion screw geometry for an external haze of the homo polypropylene melted sheet was conducted. Furthermore, in order to improve the transparency of isotactic PP sheets, multilayer extrusion was conducted using a resin with lower melt viscosity for the surface layers than the one for the core layer.

2. EXPERIMENTAL APPARATUS

Primepolymer polypropylene F-200S (density 910 kg/m^3 and MFR=2.0) was used as the material. Figure 1 shows φ50 mm screw geometry (L/D=26) used for the preliminary evaluation. The extrusion characteristics, such as extruder throughput capacities, melt resin pressure distribution, pressure fluctuation, and melt temperature of the resin, were measured by using the φ50 mm extruder with the adaptor and die (500 mm in width and 2 mm lip openings) for different angles of the extrusion screw.

Here the same extrusion temperature condition was adopted regardless of the screw geometry and the screw rotational speed. Pictures were taken by fixing a camera at the 1500 mm position from the die to judge the transparency of the melted resin sheet in the die exit.

To examine the melt plasticizing performance, the extruder was stopped using the emergency stop button when a pigment appeared from the die. After it cooled, the extruder was opened and the melt plasticizing phenomena along the screw were investigated.

3. RESULTS AND DISCUSSION

3.1 The influence of the screw geometries

The results of extrusion performance with the different screw geometry are shown in Fig. 2 and Table 1. The screw geometry was selected in order to enable melt plasticization in the early stage of the extruder even under low shear stress conditions. Screw No. 6 of a straight channel depth screw with a torpedo type barrier section at the middle part of screw was examined. It was excellent in respect to high transparency and as a result the extrusion stability.

In Fig. 3, the relationship between average melt temperature at the center of the die exit and specific energy consumption at screw speed 150 rpm were plotted. Additionally, the transparency order of melt web is also shown in Fig. 3. It was found from the results obtained by experiments of the screw geometry change that high transparency is obtained under specific low energy consumption and low melted temperature. In the case of screw No. 5 which was a straight screw of 5 mm in the channel depth, stable melt plasticizing did not occur.

To obtain highly transparent melt web, it is very important not to generate excessive heat owing to the shear stress or the shear rate on the assumption that the unmelted resin particles are not left behind. This transparent mechanism is fundamentally different from the “shark skin” generated at the die exit. That is, the “shark skin” is generated at more than a certain limit of shear stress. It means that the lower the melt temperature, the easier the “shark skin” is generated. On the other hand, the lower melt temperature causes a more transparent appearance of melt web depending on the screw geometry.

The straight channel depth screw with a torpedo type barrier achieved the most favorable results for high transparency melt web, and also for the comprehensive evaluation of the extrusion stability, throughput and melt resin temperature. However, in the case of a large size extruder for the production machine, the melt plasticization problem was predicted, and the gently tapered compression screws with a torpedo type barrier were examined in more detail.

In order to secure extrusion stability even under high throughput conditions, the plasticization capacity was improved by the optimization of the screw geometry. Also the low melt temperature was pursued as much as possible. The optimization of the lengths of feed, compression and metering zone were attempted, and also the segment which compulsorily destroyed a solid bed in the entrance section of the torpedo was newly investigated. As a result, the screw geometry to satisfy the demand for quality was obtained using these optimizations for high transparency and extrusion stability.

3.2 Effect on transparency of PP sheets by the multilayer sheet extrusion process

In order to improve the transparency of isotactic polypropylene sheet, a multilayer extrusion was conducted using a resin with lower melt viscosity for surface layers than the resin for the core layer shown in Table 2. As a result, the number of spherulites near the surface decreased dramatically as shown in Fig.4 and 5. Furthermore, Table 2 shows the internal haze of the sheet was improved. The number of spherulites in the sheet cross section are shown in Fig.5 and the internal haze correlated to each other. The flow velocity distribution (Fig.6), shear rate distribution, and shear stress distribution (Fig.7) in a die lip section were calculated using the finite element method. The shear stress in the sheet thickness direction was reduced by laminating resin with low melt viscosity on both surfaces, and therefore it can be surmised that the stress induced crystallization was restrained. Furthermore, the cooling simulation in the belt quenching process was conducted and is shown in Fig.8. The density of the number of spherulites was set as the response variable, and then the plateau area time of the temperature in each position of the thickness direction and the shear stress were chosen as the independent variables, and the prediction formula was derived by multiple regression analysis. Moreover, using the obtained formula shown in Table 3, some densities of the number of spherulites in various surface layer ratios were predicted, and optimization of multilayer composition was proposed.

4. CONCLUSIONS

The transparency of melted resin sheet was obtained by screw geometry so that the specific energy consumption was small and the melted temperature was low. The screw geometry was found to satisfy the target qualities, which were both high transparency and extrusion stability, by optimizing the geometry of the gently tapered compression screw with a torpedo type barrier.

A multilayer extrusion was also conducted using a resin with a lower melt viscosity for the surface layers than the resin for the core layer. As a result, the number of spherulites near the surface decreased dramatically, and furthermore the internal haze of the sheet was improved. Fig.9 shows the stress induced crystallization was reduced by multi-layer flow.

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Table 1. Summary of extrusion performance for each screw geometry

Screw No.	Screw Geometry	Extrusion Stability Fluctuation range at screw tip (MPa)	Transparency	Throughput at 150rpm (Kg/H)	Specific Energy Consumption at 150rpm (WH/Kg)	Melt Temperature at 150rpm (℃)
1	Low compression, deep metering channel depth	± 18.5	4	54	0.242	248
2	Low compression, shallow metering channel depth	± 0.3	2	39	0.314	252
3	Low compression, metering with mixer	± 0.3	1	43	0.292	261
4	Straight channel depth (4mm)	± 5.3	5	43	0.169	237
5	Straight channel depth (5mm)	± 9.5	3	57	0.170	228
6	Straight channel depth (4mm) with topido type barrier (δ=1.0mm, L=50mm)	± 0.3	5	43	0.178	245
7	Straight channel depth (4mm) with topido type barrier (δ=1.0mm, L=150mm)	± 0.3	1	43	0.241	256
8	Straight channel depth (4mm) with topido type barrier (δ=0.5mm, L=50mm)	± 0.4	1	45	0.217	254

Definition of transparency : Excellent=5, Very Good=4, Good=3, Fair=2, Poor=1

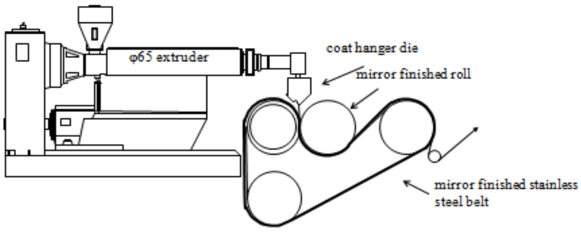

FIG. 1. Schematic diagram of extrusion sheeting system (single belt quick quenching process).

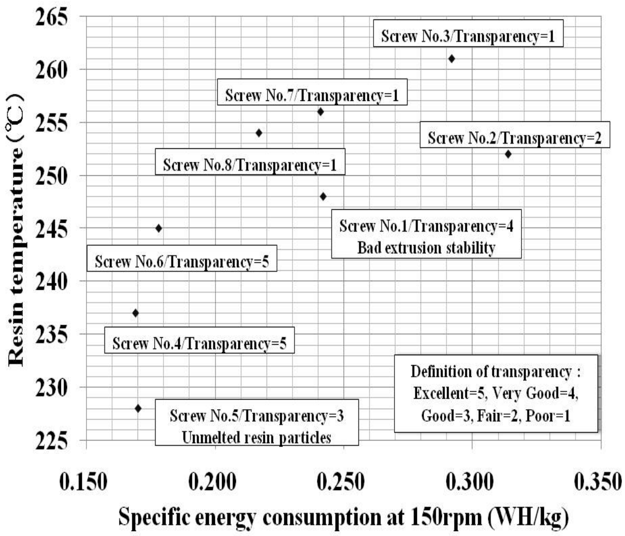

Figure 3. The relationship between average melt temperature and specific energy consumption at 150 rpm for each screw

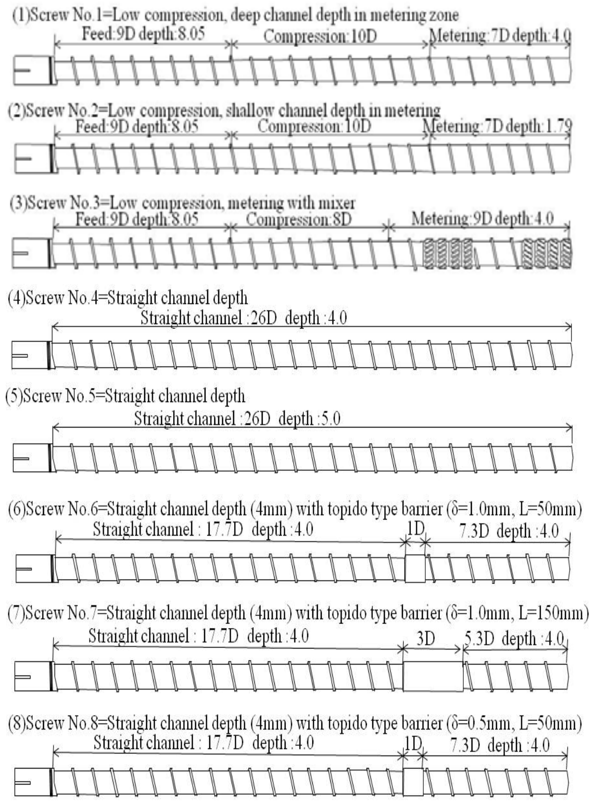

Figure 2. Schemactic representation of screw geometries, lengths expressed in extruder

Table 2. Optical Properties of Multi-Layer Sheets

Sheet structure	MFR (g/10min)	Meso pentad fraction (mol%)	Normalized to 350 μm		
			Total haze (%)	Internal haze (%)	Outer haze (%)
A	3	92.5	14.0	13.5	0.5
B/A/B	7/3/7	92/92.5/92	9.1	8.6	0.5
C/A/C	20/3/20	92/92.5/92	8.3	7.6	0.7
D/A/D	4.5/3/4.5	70/92.5/70	12.2	11.7	0.5

Sheet Thickness: 340 μm, Each Multi-Layer Thickness: 20/300/20 μm

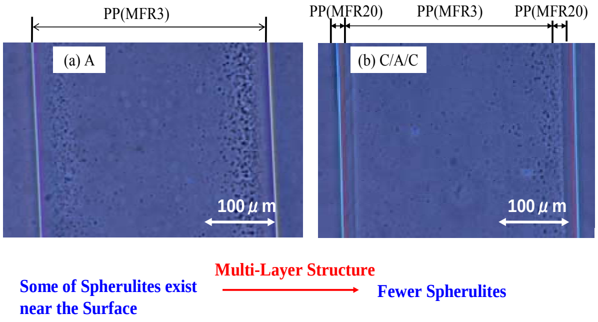

Figure 4. Micrographs obtained by phase-contrast microscopy of cross sections of (a) A and (b) C/A/C structure sheets

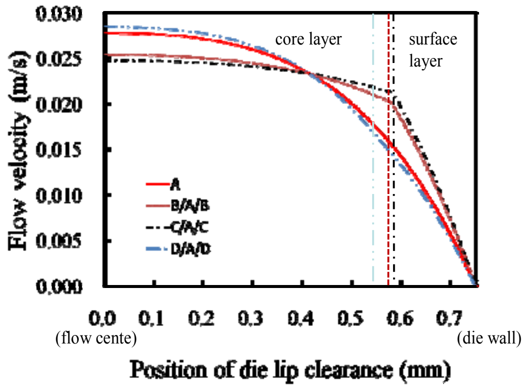

Figure 6. Flow velocity distribution at die lip section in the case of A, B/A/B, C/A/C and D/A/D flow

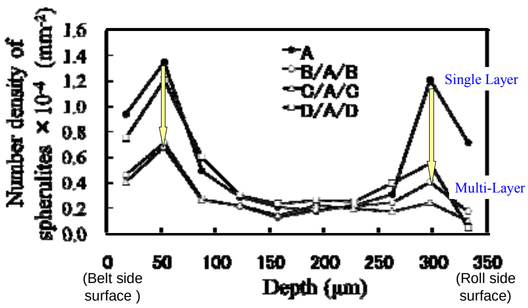

Figure 5. Depth profiles of the number density of spherulites for A, B/A/B, C/A/C and D/A/D sheets

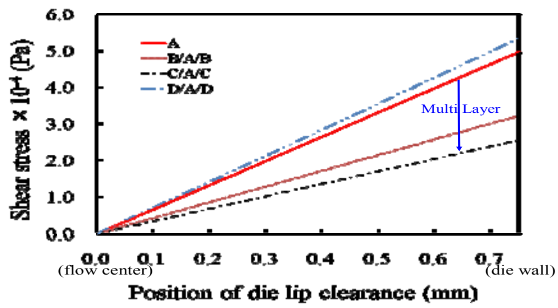

Figure 7. Shear stress distribution at die lip section in the case of A, B/A/B, C/A/C and D/A/D flow

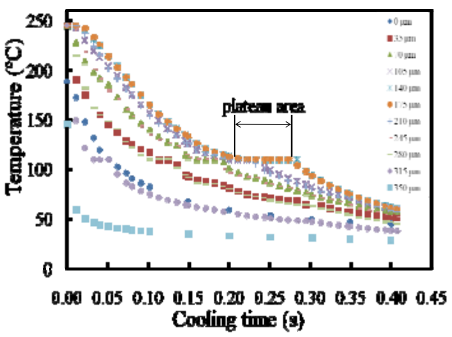

Figure 8. Temperature change in the sheet thickness direction by the cooling single belt process

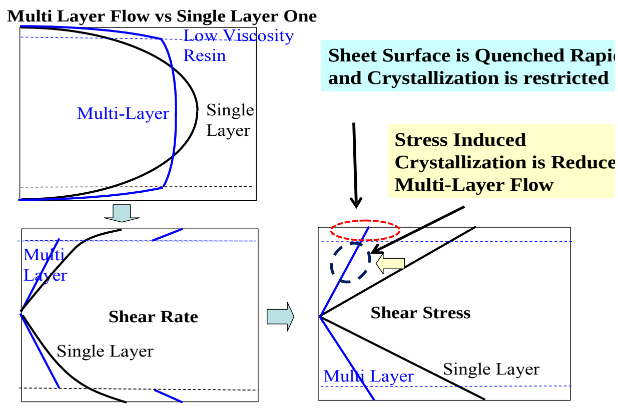

Figure 9. Reduction of Stress Induced Crystallization by Multi-Layer Flow

$$Y = -1.12 \times 10^4 + 2.96 \times 10^{-1} X_1 - 1.61 \times 10 X_1 X_2$$
$$+ 9.42 \times 10^{-3} (X_1 X_2)^2 + 1.22 \times 10^6 X_2 - 2.05 \times 10^7 X_2^2$$
$$R_f^2 = 0.90$$

Y : Spherulites Density, X_1 : Shear Stress at the Die Lip,
X_2 : Plateau Time during the Crystallization
R_f^2: Multiple Correlation Coefficient adjusted for the Degree of Freedom

Table 3. Number Density of Spherulites predicted by Multiple Regression Analysis