Effect of Processing Conditions on the Gloss of Polypropylene Films

G. W. SCHAEL,* *Esso Research and Engineering Company, Linden, New Jersey*

Synopsis

A study of the effect of three processing variables, melt temperature, quench temperature, and air pressure, on the gloss of polypropylene films shows that although these processing parameters are used to control this property they do not constitute the primary cause of gloss variations.   The main causes of gloss variations were found to be changes in the polymer melt index, i.e., melt roughness, and film crystallinity, both of which can be controlled by changing the processing conditions.

Introduction

One of the most important properties of extruded packaging films is surface gloss.   This importance arises first from the esthetic value of gloss in giving an attractive film-wrapped package and second from the effect gloss has on the coefficient of friction (COF) of the film.   Since the COF depends in part on surface roughness, it can be controlled by regulating the gloss of the film.   In Figure 1 is shown an example of the increase of COF as gloss increases and the film surface becomes smoother.   Because gloss is a significant factor in determining film quality and because of the relative ease and low cost of measuring this property it is always used as a quality-control property during film manufacture.

Experience both commercially and in the laboratory has shown that lot-to-lot variations of a given polymer or variations among different grades of polymers requires that extrusion conditions be adjusted in order to obtain a film having a surface gloss within a desired range.   Three rules of thumb for regulating the surface gloss of extruded polypropylene film which is quenched on a chill roll are (*1*) increasing melt temperature increases gloss, (*2*) decreasing the quench temperature, i.e., chill-roll temperature, increases gloss, and (*3*) increasing the contact pressure of the film on the chill roll, i.e., increased air pressure in the air knife, increases gloss.   Frequently the application of these rules is ineffective in regulating gloss.   This report describes the results of a study of the effect of melt temperature, quench temperature, and air pressure on the gloss of polypropylene films.

* Present address: Carwin Research and Development Division, The Upjohn Company, North Haven, Connecticut.
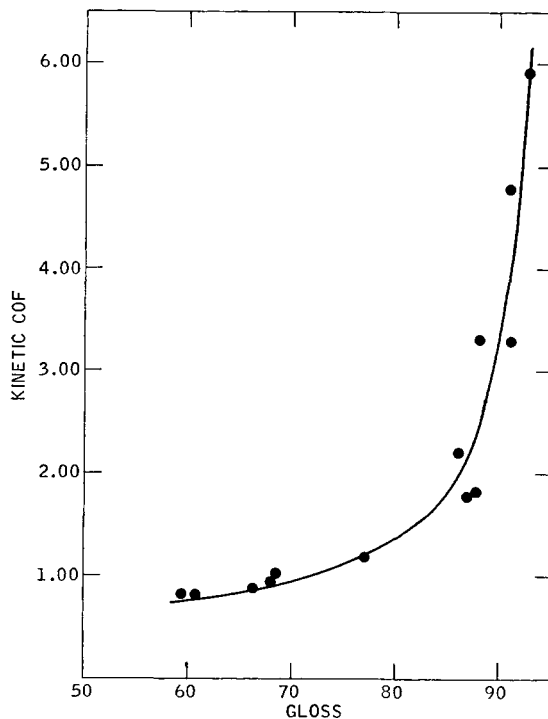
Fig. 1. Increase of COF with increasing surface gloss.

Experimental

Films having a nominal thickness of 0.87 mil were prepared from Escon polypropylene by extruding and quenching on a chrome-plated chill roll having a No. 2 high mirror finish. The films were extruded at a speed of 160 ft./min. The gloss was regulated by varying the melt temperature, quench temperature, air knife pressure (on or off). Additional control of gloss was obtained by adding a nucleating agent to some of the polymers used.

The x-ray crystallinities were determined by using a procedure described by Wilchinsky.[1] The diffraction pattern of the films was recorded by a symmetrical transmission technique. From the diffraction pattern the percentage crystallinity was determined by combining the amorphous and paracrystalline components into a noncrystalline component and calculating the crystallinity as the ratio of the area of the crystalline pattern to the total area of the diffraction pattern. Crystallinities not measured by the x-ray procedure were determined by use of the equation

$$d_f = V_c d_c + V_a d_a + (1 - V_c - V_a)d_p \qquad (1)$$

where d_f is the film density, V_c and V_a the volume fractions of the crystalline and amorphous phases, respectively, and d_c, d_a, and d_p the densities of

the crystalline, amorphous and paracrystalline phases, respectively. Natta's values[2,3] were assumed for the density of completely amorphous (0.8500 g./cm.^{3}) and completely crystalline (0.9360 g./cm.^{3}) polypropylene. A density of 0.907 g./cm.^{3} reported by Gailey and Ralston[4] was used for the paracrystalline density.

The densities of the films were calculated from refractive index measurements by using a previously described technique.[5]

The kinetic coefficient of friction of the films was determined on a Thwing-Albert Egan slip tester by ASTM test D1894-61T with a modified sled consisting of a 37.3 g. circular sled (diameter 2.19 in.) wrapped with a medium-density sponge rubber. The sled weight was chosen to give an applied pressure of 10 g./in.^{2}.

Surface gloss was determined with a Gardner automatic photometric unit fitted with a 45° Gardner gloss head. The procedure is described in the ASTM test D-523-53T.

The melt indices were measured by ASTM test 1238-62T.

Molecular weights were evaluated from the intrinsic viscosity of solutions of polypropylene dissolved in Decalin. All viscosities were determined at 135°C.

Results and Discussion

Melt Temperature

The effect of melt temperature on surface gloss was studied by extruding films at three melt temperatures (506, 530, and 560°F.) and quenching at 75°F. Two groups of films were prepared. The first group was stabilized with different concentrations of an effective processing stabilizer (A) while the second group was stabilized with a less effective stabilizer (B). In Table I are tabulated data showing the gloss, film melt index, and molecular weights obtained at the three melt temperatures. For any given stabilizer concentration increasing the melt temperature decreased the polymer molecular weight, increased the melt index, and increased the glossiness of the film surface. From the gloss data graphed in Figure 2 it can be seen that, within the range studied, gloss was a linear function of film molecular weight.

Although the apparent cause of gloss improvement was the higher melt temperature the actual cause was the degradation of the polymer during the extrusion. This degradation, indicated by the increase of polymer melt index, produced a smoother melt surface which resulted in a film having a higher gloss. Photomicrographs of films having a gloss of 68.1 and 90.9 are shown in Figure 3. The dependence of gloss on degradation during extrusion relates directly to stabilizer efficiency. Comparison of the effect of stabilizer concentration on gloss shows that when the amount of A was increased, the melt index (degradation) and gloss of the film decreased, while an increase of the amount of B had little effect in stopping degradation or decreasing gloss. It is easy to be misled by these results into con-

TABLE I
Effect of Melt Temperature on Film Gloss

Processing temperature, °F.		Stabilizer		Film properties[b]			Molecular weight × 10³ (film)
				Melt index			
Melt	Quench[a]	Type	%	Polymer	Film	Gloss	
506	75	A[c]	0.04	5.1	13.3	87.0	196
530	75	A	0.04	5.1	14.9	90.9	265
560	75	A	0.04	5.1	19.9	94.3	175
506	75	A	0.08	3.5	8.5	68.6	222
530	75	A	0.08	3.5	9.3	77.4	260
560	75	A	0.08	3.5	10.8	87.8	205
506	75	A	0.12	3.6	7.4	60.9	235
530	75	A	0.12	3.6	8.1	66.6	280
506	75	A	0.16	3.5	7.2	59.6	255
530	75	A	0.16	3.5	6.7	68.1	240
506	75	B[d]	0.04	5.9	14.7	91.2	183
530	75	B	0.04	5.9	16.5	92.7	173
506	75	B	0.08	5.5	16.4	91.0	—
530	75	B	0.08	5.5	16.4	92.9	178
560	75	B	0.08	5.5	20.5	95.0	172
506	75	B	0.12	5.9	16.5	88.1	186
530	75	B	0.12	5.9	16.9	92.3	182
506	75	B	0.16	5.3	14.5	86.4	195
530	75	B	0.16	5.3	17.3	91.6	182
560	75	B	0.16	5.3	15.8	93.7	185

[a] Quench temperature is taken as equal to the measured chill roll temperature.

[b] The crystallinity of the films was zero. The phase content consisted of approximately 64% paracrystalline and 36% amorphous material.

[c] Films made from polypropylene stabilized with stabilizer A.

[d] Films made from polypropylene stabilized with stabilizer B.

cluding that the different gloss values were the result of type of stabilizer rather than stabilizer efficiency. Such is not the case. If the gloss data in
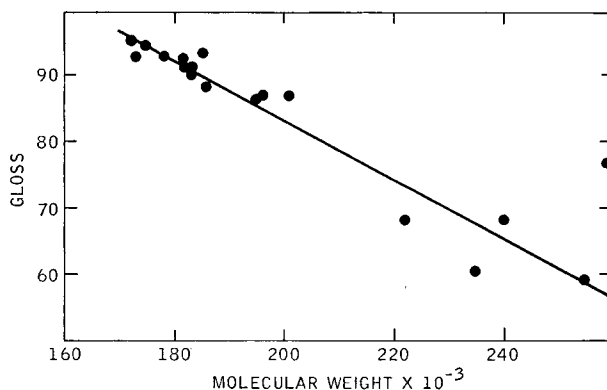

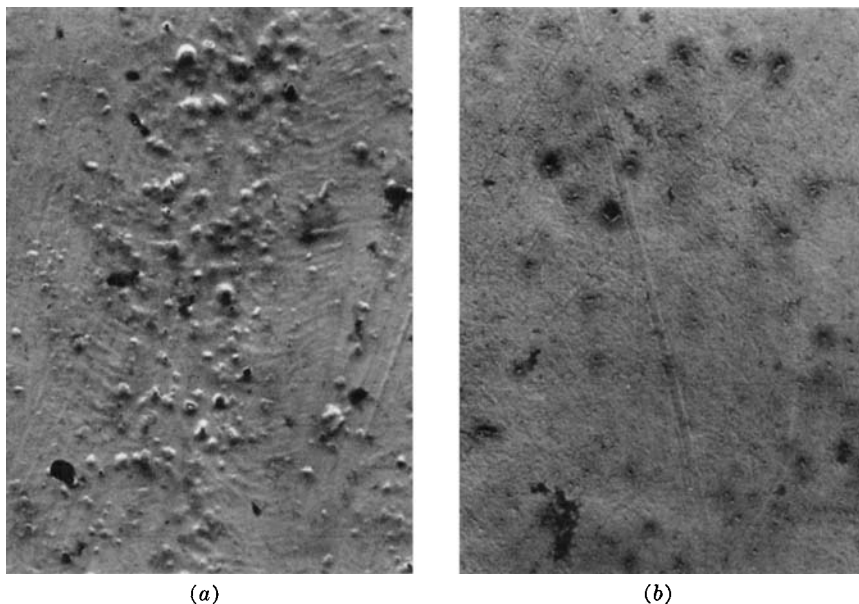Fig. 2. Decrease of gloss with increasing molecular weight.

(a)   (b)

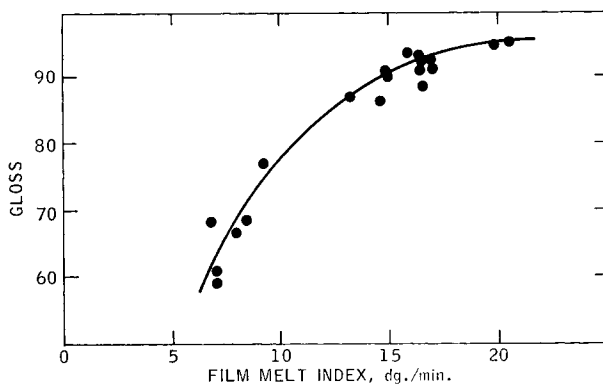Fig. 3. Photomicrographs of film surfaces: (a) gloss 68.1; (b) gloss 90.9.   10,000×.

Fig. 4. Increase of film gloss with increasing melt index.

Table I are plotted as a function of film melt index (see Fig. 4), the points for all of the films fall on the same line regardless of stabilizer type.   If a stabilizer also acts as a nucleating agent then stabilizer type would have to be considered as a gloss controlling factor.   It can also be seen from Figure 4 that, because of the greater slope of the curve for melt indices between 5 and 12, there will be greater difficulty in controlling the gloss of films made from well-stabilized polymer of low melt index.

Air Pressure

Quenching the polymer film after extrusion is facilitated by pressing the film on the chill roll with a jet of air from an air knife.   The improved

contact to the chill roll results in more efficient quenching and yields a glossier film.

To demonstrate the effect of air pressure on gloss, films were prepared which were quenched with and without pressure applied across the full film width and with pressure applied on only half the film width. All of the films were made from the same polymer containing different concentrations of nucleating agent to aid in controlling crystallinity. The melt and quench temperatures were held constant to minimize changes in film melt index. The gloss and crystallinities of the films are shown in Table II.

TABLE II
Effect of Air Pressure on Film Gloss and Crystallinity

| Processing temperatures, °F.		Nucleating Additive		Film properties			
				Gloss		Crystallinity, %	
Melt	Quench	Type	%	With air	Without air	With air	Without air
Air pressure applied to half of film width							
545	79	A	0.005	91	77	(0.0)[a]	14.0
549	80	A	0.008	88	59	(0.4)	(33.1)
549	80	A	0.015	86.5	58	(0.5)	(31.0)
Air pressure applied to full film width							
549	80	A	0.03	83.7	54.5	13.0	(27.3)
550	80	A	0.05	88.2	55.5	(0.0)	(26.5)
548	80	B	0.005	91.3	77.0	0.3	10.5
549	80	B	0.008	90.0	78.0	2.5	6.0
549	80	B	0.015	89.2	82.0	2.5	9.0
550	79	B	0.003	90.8	87.0	4.0	12.5
550	78	B	0.05	91.3	86.6	4.0	12.0
550	79	B	0.00	91.5	85.7	5.5	12.5

[a] Values in parentheses are crystallinities determined by x-ray analysis. Other crystallinities were determined by using eq. (1).

Application of air pressure to only half the film dramatically demonstrates the effect of quenching with and without air under exactly the same thermal conditions. The gloss of the film half having no applied air pressure was 14–29 units lower and crystallinity 14–33% higher than the film half to which pressure was applied. It is apparent from the gloss values obtained with application of pressure that although the polymer underwent enough degradation to give a high-gloss film, removal of the air pressure yielded a low-gloss surface. The low gloss in this instance resulted from increased surface roughness produced by the formation of spherulites on the film surface. In Figure 5 are shown photomicrographs of three film surfaces having gloss values of 78, 85, and 90. As the gloss increased the size of the spherulites decreased until they became almost indiscernible as the gloss approached 90. In comparison, the low gloss surface in Figure 3 contains no spherulites but is rough in texture. In Figure 6 are plotted data showing the change of gloss with increasing crystallinity. At zero crystallinity, i.e.,
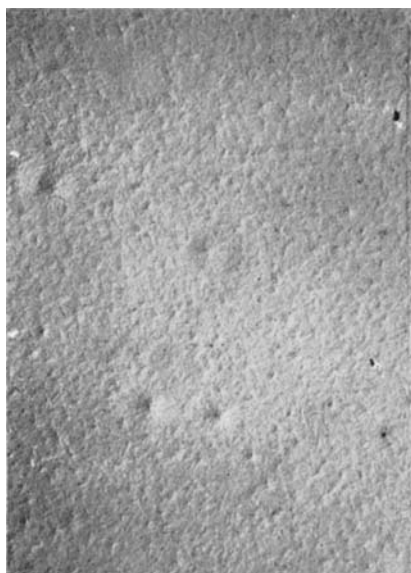
approximately 64% paracrystalline and 36% amorphous phase content, the gloss was 92. Increasing the crystallinity to 30% decreased the gloss to 56.7%.
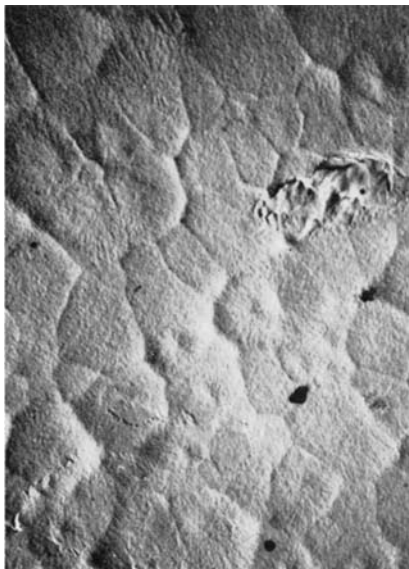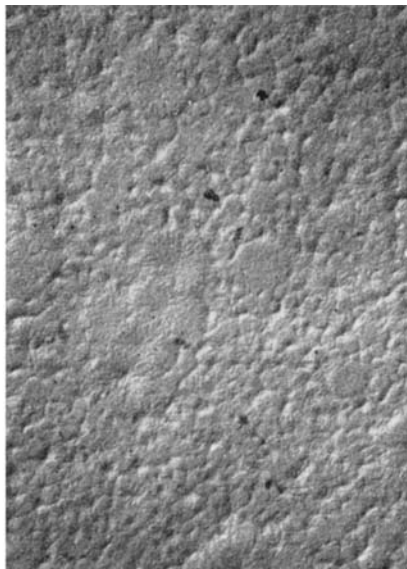
(a)   (b)

(c)

Fig. 5. Photomicrographs of film surfaces: (a) gloss 78; (b) gloss 83; (c) gloss 90. 10,000×.
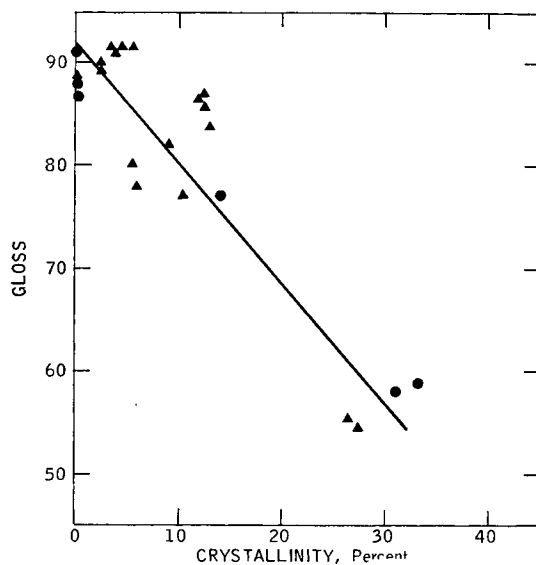
Fig. 6. Decrease of gloss with increasing crystallinity: (●) air pressure applied to only half of film.

Quench Temperature

The gloss and crystallinity of films extruded at a melt temperature between 530 and 538°F. and quenched at temperatures between 63 and 100°F. are compared in Table III. Increasing the quench temperature

TABLE III
Effect of Quench Temperature on Gloss and Crystallinity

Processing temperature, °F.		Stabilizer[b]		Film properties[c]			
Melt	Quench[a]	Type	%	Gloss	Melt index	Density, g./cm.[3]	Crystal-linity, %
538	70	A	0.12	57.1	7.3	0.8855	5.5
538	77	A	0.12	60.9	7.3	0.8849	3.0
530	75	A	0.12	66.6	8.1	0.8838	0.0
536	90	A	0.16	68.1	8.0	0.8852	3.0
536	100	A	0.16	58.4	8.0	0.8860	9.0
535	63	B	0.04	93.8	20.1	0.8844	0.0
535	70	B	0.04	92.8	20.1	0.8846	0.0
535	80	B	0.04	92.3	20.1	0.8849	2.5
536	92	B	0.08	92.6	19.6	0.8855	6.0
536	100	B	0.08	83.9	19.6	0.8857	7.0

[a] Quench temperature is equal to the measured chill roll temperature.

[b] Stabilizer A was an effective processing stabilizer while stabilizer B was relatively ineffective.

[c] The polymer used to make these films contained no nucleating additive.

from 63 and 70°F. to 100°F. increased the crystallinity by 7–9% and decreased the gloss by approximately 9–10 units. Since quench temperatures up to 90°F. had little effect on gloss, it is believed that melt roughness and crystallization tendency of the polymer are more significant in controlling gloss than is quench temperature.

Summary

From the results of this study it is seen that the processing variables of melt temperature, quench temperature, and air pressure which are used to change surface gloss produce this change by modifying the polymer melt index (melt roughness) and film crystallinity. Increasing melt temperature degrades the polymer, increases melt index and decreases melt roughness, with a resulting improvement of surface gloss.

TABLE IV
Effect of Increasing Magnitude of Processing Variables

Property	Melt temperature	Quench temperature	Increasing air knife pressure
Gloss	Increases	Decreases	Increases
Melt index	Increases	No effect	No effect
Melt roughness	Decreases	No effect	No effect
Crystallinity (surface spherulites)	No effect	Little effect except with nucleated polymer	Decreases
Molecular weight	Decreases	No effect	No effect

Application of air pressure to the extruded film during quenching was found to be critical in giving a glossy surface. Even with polymers of high melt index, which potentially give high gloss surfaces, air pressure is necessary to insure effective quenching and minimize spherulite formation.

Quench temperature is considered as having only a minor effect on gloss. Within the temperature range studied this variable did not significantly change the gloss, density, or crystallinity of the test films.

The relationship existing among the variables discussed in this paper are summarized in Table IV.

References

1. Z. W. Wilchinsky, paper presented at 13th Annual Conference for the Application of X-Ray Analysis, Denver, Colorado, Aug. 1964.

2. G. Natta, P. Corradini, and M. Cesari, *Atti Accad. Nazl. Linecei, Rend. Classe Sci. Fis. Mat. Nat.*, **22**, 11 (1957).

3. G. Natta, F. Danusso, and G. Moraclio, *Angew. Chem.*, **69**, 686 (1957).

4. J. A. Gailey, and R. H. Ralston, *SPE Trans.*, **4**, No. 1, 29 (1964).

5. G. W. Schael, *J. Appl. Polymer Sci.*, **8**, 2717 (1964).

Résumé

Une étude de l'effet des trois variables du processus à savoir, la température de fusion, la température de quenching et la pression d'air sur la transparence de films de propylène montre que si ces paramètres du processus sont utilisés pour contrôler cette propriété, elles n'en constituent pas la cause première des variations. La cause principale des variations a été trouvée être la rhéologie du polymère c'est à dire la difficulté d'écoulement, la cristallinité du film, chacune de celles-ci pouvant être contrôlées en variant les conditions de processus.

Zusammenfassung

Eine Untersuchung des Einflusses von drei Verarbeitungsvariablen, nämlich der Schmeltztemperatur, der Abschreckungstemperatur und des Luftdruckes auf den Glanz von Polypropylenfilmen zeigt, dass die angegebenen Verarbeitungsparameter zuar zur Kontrolle dieser Eigenschaft verwendet werden, dass sie aber doch nicht die primäre Ursache für die Änderungen des Glanzes sind. Als Hauptursache für die Variation des Glanzes werden Änderungen der Polymerrheologie, d.h. der Schmelzrauhigkeit und der Filmkristallinität, festgestellt, welche beide durch Änderung der Verarbeitungsbedingungen kontrolliert werden können.