

A Study of the Morphology and Physical Properties of Polypropylene Films

GEORGE W. SCHAEEL,* *Esso Research and Engineering Company, Linden, New Jersey*

Synopsis

Data are presented showing that rapidly quenched polypropylene films contain a paracrystalline phase in addition to crystalline and amorphous phases. The density of the paracrystalline phase was found to vary between 0.8890 and 0.9080 g./cm.³. Other data show that the density of quenched polypropylene films increases during aging, the increase being a linear function of the logarithm of the film age. The physical properties of the films, secant modulus, impact strength, and coefficient of friction, were found to vary not only with crystallinity, i.e., composition-dependent density, but also to vary during aging when the density was increasing with little change in crystallinity. An example of this dependence is the increase in secant modulus of 10.9×10^6 psi/g./cm.³ during aging and only 3.7×10^6 psi/g./cm.³ as the crystallinity increased. Because of the three phases and the density-time change, it is possible to have films with the same density and different physical properties and, conversely, films with the same physical properties and different densities.

INTRODUCTION

Although the properties and morphology of polypropylene have been studied extensively,¹⁻⁶ much of the work has centered on fibers and samples prepared in the laboratory under ideal conditions leading to an equilibrated internal structure. When the same polymers are fabricated in high-speed commercial operations, the rate of production does not permit the formation of an equilibrated sample. Because processing conditions have such a profound effect on polymer properties, it is essential, both to the polymer manufacturer and the product fabricator, that a knowledge of the nonequilibrium polymer properties be understood. This paper attempts to answer part of this need by describing the results of a study of the morphology existing in polypropylene films prepared by commercial extrusion techniques and the effect of the resulting morphology on the modulus, impact strength, and coefficient of friction of the films.

EXPERIMENTAL PROCEDURES

Films having a nominal thickness of 1 mil and different crystallinities were prepared from Escon polypropylene by extruding and quenching on a

* Present address: Carwin Research and Development Division, The Upjohn Company, North Haven, Connecticut.

chill roll at a speed of 160 ft./min. By making films of different crystallinity from one resin the molecular composition of the films was kept constant. The crystallinity of the films was varied by regulating the resin melt temperature between 500 and 550°F. and the chill-roll temperature between 65 and 90°F. Varying concentrations of a nucleating agent (succinic acid) were added to the polymer to facilitate control of crystallinity. Densities, secant moduli, impact strengths, and coefficients of friction of the films were determined as a function of time.

The densities of the films were calculated from refractive index measurements by using a technique which has been previously described.⁹ Natta's values^{10,11} were assumed for the density of amorphous (0.8500 g./cm.³) and crystalline (0.9360 g./cm.³) polypropylene.

The secant moduli were measured on an Instron tensile tester by using ASTM test D-1530-58T with a sample 0.625 in. in width which was elongated at a strain rate of 12%/min.

The impact strengths were measured with an Elmendorf tearing tester fitted with a Spencer impact attachment.

The static coefficient of friction of the films was determined on a Thwing-Albert Egan slip tester by ASTM test D-1894-61T with a modified sled consisting of a metal block, 1 × 2 in., weighing 100 g. and wrapped with a medium-density sponge rubber.

X-ray determinations of per cent crystallinity were carried out by using a procedure described by Wilchinsky.¹² The diffraction pattern of the films was recorded by a symmetrical-transmission technique. From the diffraction pattern the per cent 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.

DISCUSSION

Polymer Composition

Polypropylene can exist in at least four morphological forms,¹³⁻¹⁵ including the monoclinic, hexagonal, and paracrystalline structures which are formed by isotactic molecules and the amorphous structure which may be formed from atactic or isotactic molecules. Examination of x-ray diffraction patterns (Figs. 1A and 1B) obtained on films used in this work showed that the monoclinic, paracrystalline, and amorphous forms were present. The presence of a third phase is also indicated by the position of the experimental curve of density versus crystallinity shown in Figure 2. It can be seen that the densities of films having crystallinities between 0 and 50% were considerably greater than would be predicted from the straight line relating density and crystallinity of the two-phase system.^{7,8} The greater than predicted density of the experimental samples indicates that the third phase has a density intermediate between the amorphous and crystalline densities. It is also worthy of note that the experimental curve intersects

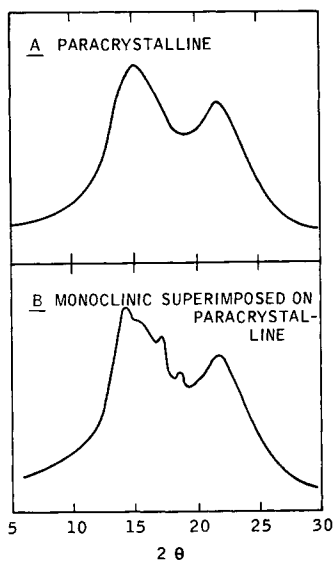


Fig. 1. Typical x-ray diffraction patterns measured on polypropylene films.

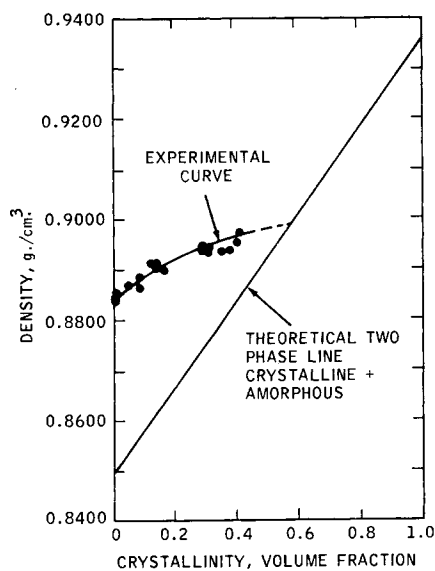


Fig. 2. Increase of density with crystallinity.

the straight line at a crystallinity of approximately 60%. At this point the paracrystalline phase should be completely converted to crystalline. Films of greater than 60% crystallinity should therefore contain only two phases.

In order to properly define the composition of the samples it was necessary to calculate the volume fraction of each form by using x-ray crystallinity

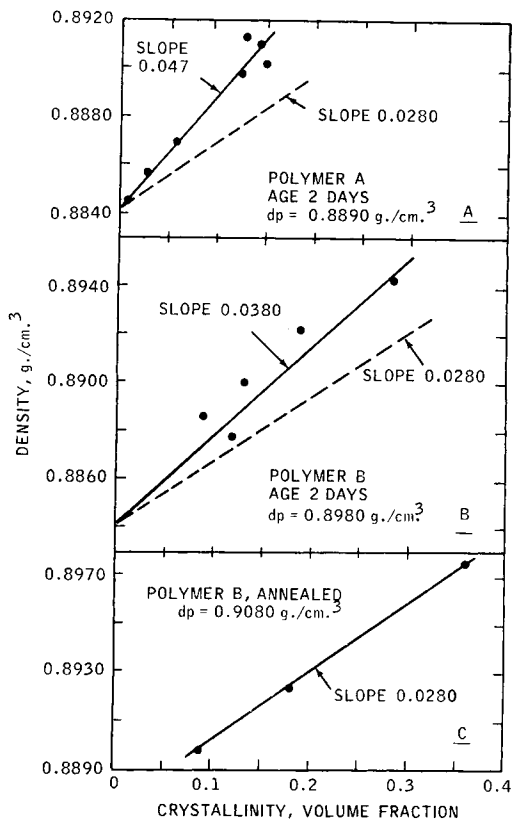


Fig. 3. Increase of film density with increasing crystallinity.

and density data. If a polymer contains only crystalline and amorphous material the volume fractions (V_c and V_a) are related to the bulk density d by eq. (1)

$$d = V_c d_c + V_a d_a \quad (1)$$

which can be used to calculate the volume fraction–density line shown in Figure 2. If a paracrystalline form is present, a term, $V_p d_p$, is added to eq. (1)

$$d = V_c d_c + V_a d_a + V_p d_p \quad (2)$$

to include the contribution of this phase to the bulk density. To calculate the volume fraction of the three forms present in a film requires that the paracrystalline density be known.

By writing eq. (2) in the form

$$d = V_c d_c + V_a d_a + (1 - V_c - V_a) d_p \quad (3)$$

and rearranging to

$$d = V_c (d_c - d_p) + V_a (d_a - d_p) + d_p \quad (4)$$

TABLE I
Paracrystalline Density of Films

Polypropylene film	Film age, days	Slope	Paracrystalline density ($d_c - \text{slope}$)
A	2	0.047	0.8890
B	2	0.038	0.8980
B, annealed	—	0.028	0.9080

it can be seen that if V_a , d_a , and d_p are constant, a plot of d versus V_c will be linear with a slope equal to $(d_c - d_p)$. The density of the paracrystalline phase can then be determined from the slope of the line. In Figure 3 are plotted density-crystallinity data for two sets of films aged 2 days and a set of annealed films. From the slopes of these lines the paracrystalline densities in Table I were calculated. Contrary to expectation, the paracrystalline density determined from each slope was different.

Although the paracrystalline density is not a constant, the density in a given set of films at a given age appeared to be constant enough to satisfy the conditions required in eq. (4).

By changing the resin type or annealing the film it was possible to obtain paracrystalline densities between 0.8890 and 0.9080 g./cm.³. The annealed paracrystalline density of 0.9080 g./cm.³ agrees well with the results of Gailey and Ralston,¹⁵ who obtained a density of 0.9070 g./cm.³ using an infrared technique. In Figure 4 are plotted density-crystallinity data for the films used in this work. In these films, which were at least 7 days old,

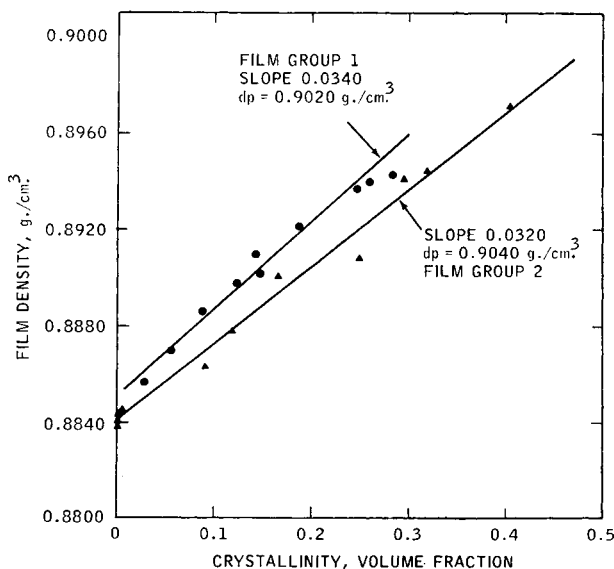


Fig. 4. Density-crystallinity relation for films 7 days old.

the rate of increase of density with time was extremely slow. The two lines in the figure represent samples with similar amounts of the amorphous phase calculated by using the Gailey and Ralston paracrystalline density of 0.9070 g./cm.³. From the slopes of the line for the two sets of data, paracrystalline densities of 0.9020 and 0.9040 g./cm.³ were determined.

In Table II are shown the phase composition of the two film sets calculated using the experimental paracrystalline densities. The average

TABLE II
Phase Content of Films Aged Seven Days or Longer^a

Film	Polymer	Film density, g./cm. ³	Crystal- line, % ^b	Paracrystal- line, %	Amor- phous, %
Film group 1	A	0.8972	40.9	23.1	36.0
	A	0.8945	31.6	33.3	35.1
	A	0.8941	29.4	36.2	34.4
	B	0.8908	24.8	37.4	37.8
	A	0.8900	16.5	49.6	33.9
	B	0.8878	11.8	53.2	35.0
	A	0.8863	9.0	54.9	36.1
	A	0.8845	0.6	65.4	34.0
	B	0.8843	0.1	65.8	34.1
	B	0.8841	0	65.6	34.4
	B	0.8839	0.1	65.0	34.4
	B	0.8838	0	65.0	35.0
	Average				
Film group 2	B	0.8943	28.3	36.9	34.8
	A	0.8941	25.9	40.4	33.7
	B	0.8937	24.9	41.4	33.7
	B	0.8922	18.7	48.3	33.0
	A	0.8910	14.2	53.4	32.4
	A	0.8902	14.8	50.9	34.3
	A	0.8898	12.4	53.9	33.7
	B	0.8886	8.7	57.6	33.7
	A	0.8870	5.6	62.9	31.5
	A	0.8857	2.8	61.6	35.6
	Average				

^a The phase contents were calculated on the basis of $d_c = 0.9360$ g./cm.³, $d_a = 0.8500$ g./cm.³, and $d_p = 0.9020$ g./cm.³ (group 1), 0.9030 g./cm.³ (group 2).

^b X-ray data.

amorphous content of film group 1 was 35.0% and that of film group 2 was 33.6%. Wyckoff⁶ has reported an amorphous content between 25 and 30% compared to 5.8% heptane extractables in the polymer. The samples of Gailey and Ralston¹⁵ contained about 40% of the amorphous phase. The amorphous content (33.6% and 35%) listed in Table II were measured on polymers containing 5–7% heptane extractables. Similar measurements on a polymer containing 10–12% heptane extractables resulted in an amorphous content of 45%.

Time Variation of Film Density and Properties

When polypropylene film is extruded it is subjected to extremely rapid quenching which leaves the film in a metastable state and produces physical properties different from those of molded or preannealed samples. The metastability not only alters the physical properties but also causes a time change in film density and other density-dependent properties such as modulus, impact strength, and coefficient of friction (COF). Comparison of the density-time change of a number of polypropylene films prepared at different melt and chill-roll temperatures as well as films which were annealed immediately after quenching showed that the magnitude of the change in density during aging is very nearly the same for all films. In Figure 5 are shown density-time curves for two films (1 and 2) made from

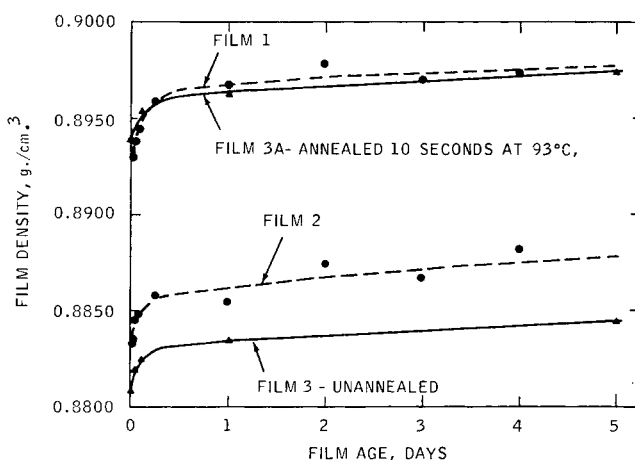


Fig. 5. Increase of polypropylene film density during aging.

the same resin at different thermal conditions and one film before and after annealing. The average increase in density of these films during 5 days of aging was 0.0038 g./cm.³. The same type of density-time change was found to occur in polyethylene and a propylene-ethylene copolymer. In Figure 6 the density change for the three types of polymers is shown to be a linear function of the logarithm of the film age.

Among the possible ways in which the film density can increase are (1) conversion of the paracrystalline phase to crystalline during annealing or aging, (2) an increase in paracrystalline density during aging, (3) a conversion of amorphous isotactic material to the paracrystalline phase, or (4) a general contraction of the film during aging.

In Table III are compared the measured crystallinity increase during the first 2-7 days of film aging and the crystallinity increase calculated on the assumption that the density change is a conversion of the paracrystalline material to crystalline. The measured crystallinity increase was much less than the calculated value, indicating that only a small portion of the para-

TABLE III
Comparison of Measured and Calculated Crystallinity Changes Occurring during Film Aging

Film ^a	Film age, hr.	Density, g./cm. ³	Crystallinity, %	
			Measured	Calculated ^b
A	0.5	0.8879	13.1	(13.1)
	48	0.8902	15.6	22.6
A1	0.5	0.8827	0.3	(0.3)
	48	0.8845	0.7	6.2
B	0.5	0.8946	39.8	(39.8)
	168	0.8972	42.7	56.8
B1	0.5	0.8841	7.6	(7.6)
	168	0.8855	9.5	11.8

^a Films A and B were both prepared from the same resin. The crystallinities were controlled by varying the extrusion and quenching temperatures.

^b The crystallinities were calculated by using an average paracrystalline density of 0.9030 g./cm.³.

crystalline phase was converted to crystalline during aging at room temperature. The average crystallinity increase for all of the films except A1 was 2%. This increase would only change the density 0.0006 g./cm.³ or 15.8% of the average total change.

Conversion of amorphous isotactic material to either the paracrystalline or crystalline phases did not occur to a significant degree during aging or annealing. In Table IV are shown data comparing the change in phase content of a film before and after annealing at 24, 76, and 93°C.

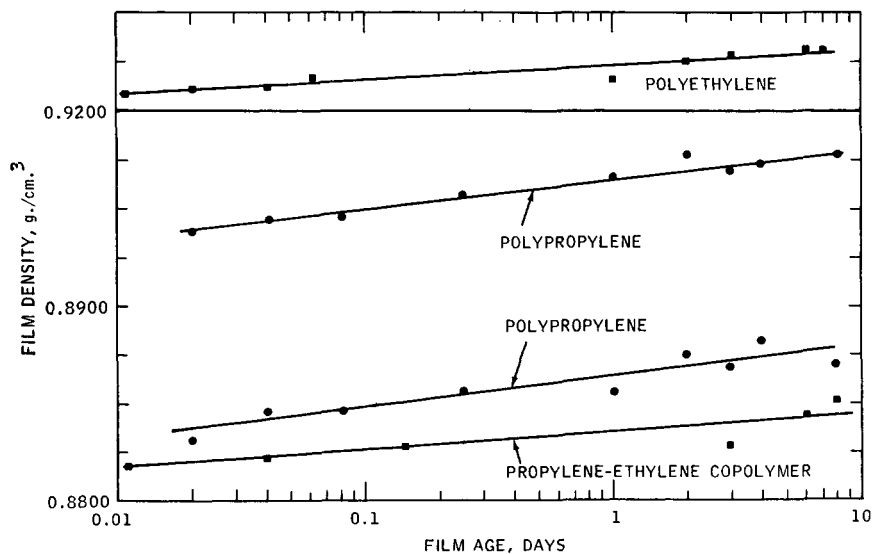


Fig. 6. Increase of film density during aging.

TABLE IV
Phase Content of Annealed Film (Polymer B)^a

Annealing temperature, °C.	Density, g./cm. ³	Crystalline, %	Paracrystalline, %	Amorphous, %
Unannealed	0.8886	8.7	57.6	33.7
24	0.8898	8.7	55.1	36.2
76	0.8924	18.0	45.8	36.2
90	0.8976	36.0	28.1	35.9

^a The phase content of the unannealed film was calculated on the basis of $d_p = 0.9040$ g./cm.³. For the annealed films, d_p was taken as 0.9080 g./cm.³ (see Fig. 3C).

The amorphous content was essentially unchanged in the annealed samples and only 2.5% lower in the unannealed sample. The main result of annealing was to increase the paracrystalline density and convert part of this phase to the crystalline phase.

Consideration of the data presented indicates that the density-time change can only be explained in terms of a variable having the same concentration in all films. This assumption is necessitated by the unique parallelism of the data shown in Figures 5-7 which were obtained on films of different morphological content.

In Figures 5 and 6 are shown density-time curves during aging of films at room temperature. All of the curves are approximately parallel, with the curve for the more crystalline film displaced to a higher density value. This same result was obtained with an unannealed film and the same film

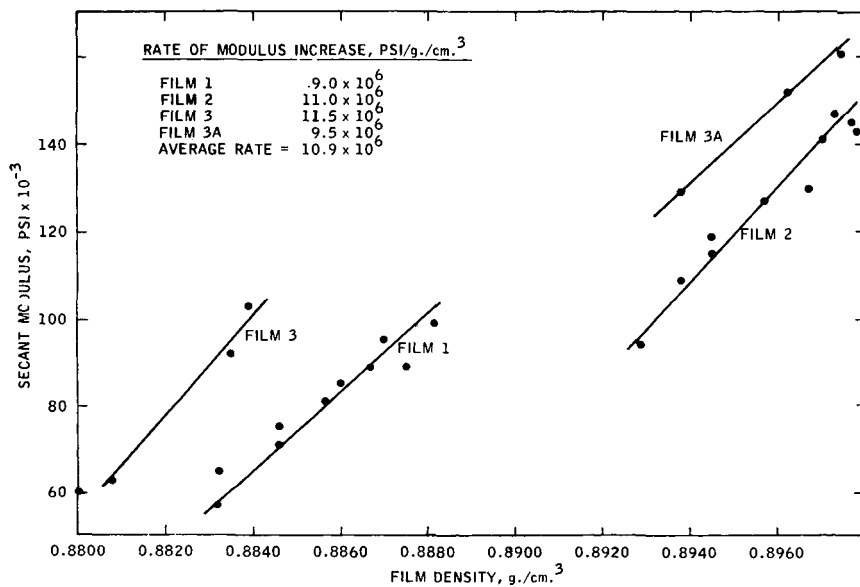


Fig. 7. Increase of secant modulus vs. film density increase during aging.

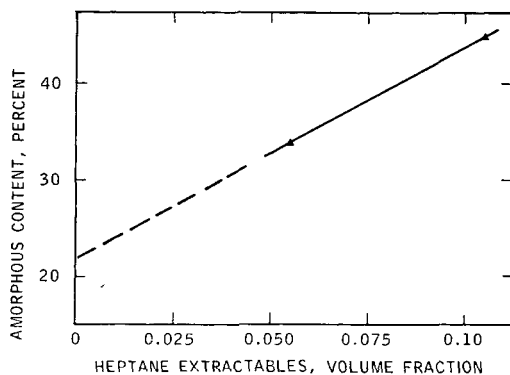


Fig. 8. Change of amorphous content vs. heptane-extractables.

annealed in-line during fabrication. The annealing time was 10 sec. at 93°C. Although annealing increased the crystallinity, the density-time change still resulted and was still parallel with the change measured on the unannealed film. In Figure 7 are shown data relating the film modulus to the density measured during aging. The lines are again parallel indicating that the rate of modulus increase during aging is independent of phase content. If the time change of film density were dependent on a density change in any one or more phases in the film, the parallelism noted would not occur because of the necessary dependence on the concentration of each phase.

A satisfactory explanation of the data can be obtained if it is assumed that the density-time change in the films occurs because of a change in concentration of randomly distributed voids^{16,17} resulting from chain entanglements, crystal imperfections, and other irregularities. The initial concentration of voids is assumed to be the same in all of the films. As the films age, the density increases because of a decrease in void concentration or volume, thus resulting in all of the lines of Figures 5 and 6 being parallel and independent of phase content. This explanation is also compatible with the results obtained by in-line annealing. Annealing did increase the film crystallinity but did not eliminate the density-time change. It would be expected that annealing would have a lesser influence on the void concentration than on the phase concentration. The void concept is also satisfactory for explaining the density-time change measured on polyethylene (Fig. 6) which does not contain a paracrystalline phase. Similar considerations would be applicable to the parallel modulus-density curves shown in Figure 7.

An indication of the contribution of voids to the apparent amorphous phase content is obtained by comparing the heptane extractable and amorphous content. If one assumes that the heptane-soluble molecules in the polymer constitute most of the uncrystallizable material it becomes apparent that the high value of the calculated amorphous phase results from the small quantity of heptane-solubles plus the contribution of the

voids.¹⁷ If the volume fraction of calculated amorphous material is assumed to change linearly with heptane-extractables it can be seen from Figure 8 that complete elimination of heptane-extractables from the polymer would still result in films having an apparent amorphous content of 22%.

Physical Properties of Polypropylene Films

Changes in density during aging and film crystallinity during processing will affect many of the film physical properties. Among the properties studied in this work were secant modulus, burst strength, and coefficient of friction (COF).

The secant modulus was found not only to increase as the film density increased but also to follow two paths. If the density increase occurred during aging (see Fig. 7), the modulus increased linearly at an average rate of 10.9×10^6 psi/g./cm.³ In Figure 9 are shown data for the increase of modulus with increasing crystallinity (plotted as density) after 7 days aging (Fig. 9, line A), and for annealed films and films aged 200 days (Fig. 9, line B). The dotted lines (films 1 and 2) represent the change of modulus during film aging. The rate of modulus increase along lines A and B was 3.7×10^6 psi/g./cm.³. The average calculated paracrystalline density of the films falling on line A was 0.9030 g./cm.³ and 0.9080 g./cm.³ for those films falling on line B. Because of the presence of three phases and the changing sample density it is possible to have films with the same density and different moduli and, conversely, films with the different densities and the same moduli. To compare film properties properly, care should be taken to establish the correct morphological definition of the sample.

It has been shown¹⁸ that as the degree of crystallinity of a polymer increases, the impact strength decreases. In Figure 10 are plotted data showing the rapid loss of impact strength as crystallinity increases. The impact strength also decreases during film aging when there is no appreciable

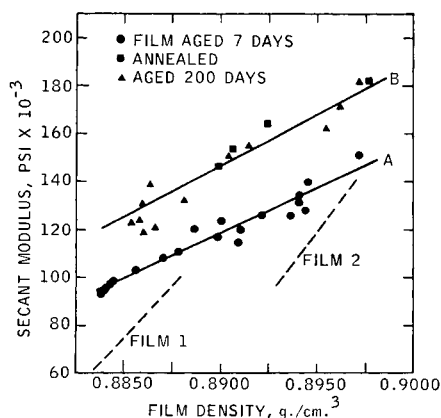


Fig. 9. Increase of film modulus with increasing density: (--) films from Fig. 7 in which the modulus increased during aging.

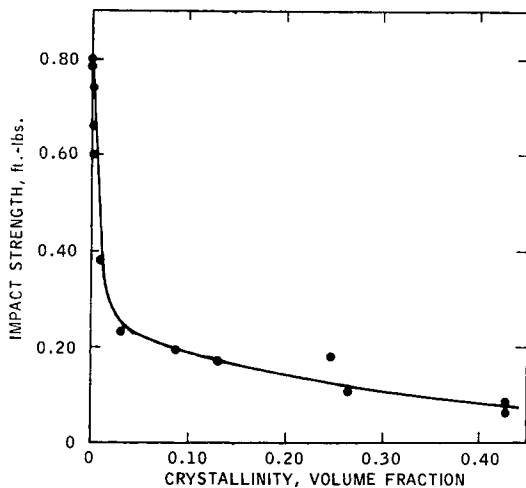


Fig. 10. Decrease of impact strength with increasing crystallinity.

change in crystallinity. This change of impact strength is probably due to embrittlement of the film caused by the density increase. Data comparing the change of impact strength and modulus with time are shown in Table V. The impact strength decreased rapidly during the first day of aging, with a gradual decrease occurring thereafter.

Examination of the literature on friction phenomena shows that the frictional force F between two plastic surfaces is proportional to the load W raised to a power, i.e., $F = KW^n$.¹⁹ The exponent n , which varies between the limits of 1 for a plastic deformation and $2/3$ for an elastic deformation, depends on the viscoelastic character of the plastic. It has also been shown²⁰ that the coefficient of friction (COF) μ is related to the viscoelastic properties of specific shear strength S and yield pressure P of the plastic by the equation $\mu = S/P$, with the shear strength, yield pressure, and ratio of these quantities decreasing as the temperature of the plastic is in-

TABLE V
Change of Impact Strength and Modulus during Film Aging^a

Film age, hr.	Density, g./cm. ³	Impact strength, ft.-lb.	Secant modulus, psi $\times 10^{-3}$
0	0.8814	1.59	68
0.5	0.8820	1.01	76
1	0.8828	1.16	80
2	0.8833	0.87	82
6	0.8841	0.29	90
24	0.8841	0.35	94
48	0.8846	0.17	102
120	0.8826	0.48	105
168	0.8863	0.22	103

^a Polymer: Escon polypropylene, CD26M4.

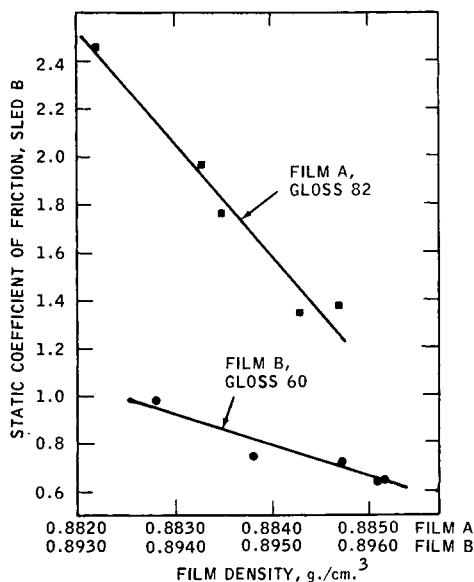


Fig. 11. Decrease of COF with increasing density of polypropylene films containing no additives.

creasing. Since the viscoelastic character of polypropylene films in part depends on the density, it would be expected that a change in density will produce a change in COF. From measurement of the COF and density changes in polypropylene films containing no additives, it was found that the COF decreased as the film density increased. The change of density and COF of two polypropylene films (film A, gloss 82; film B, gloss 60) containing no slip additive are compared in Table VI. The greatest changes in density and COF occurred during the first 24 hr. of aging. In Figure 11 it is shown that the COF decreased linearly as the density increased during aging.

In a manner similar to the modulus and impact strength, the COF not only depends on the density time change but also decreases with increasing

TABLE VI
Change of Density and COF during Aging of
Polypropylene Film Containing No Amide

Film age	Film A, gloss 82, sled B		Film B, gloss 60, sled B	
	Static COF	Density, g./cm. ³	Static COF	Density, g./cm. ³
5 min.	2.45	0.8822	0.97	0.8938
6 hr.	1.95	0.8833	0.74	0.8948
1 day	1.76	0.8835	0.71	0.8957
3 days	1.37	0.8847	0.63	0.8961
7 days	1.34	0.8843	0.63	0.8961

TABLE VII
Decrease of COF with Increasing Crystallinity

Film 1, gloss 60.9		Film 2, gloss 87.0	
Static COF	Crystallinity, %	Static COF	Crystallinity, %
0.73	2.0	1.48	1.0
0.70	2.5	1.37	1.0
0.73	3.0	1.28	2.5
0.62	17.0	1.10	11.0
0.50	32.0	0.63	27.0

crystallinity. In Table VII are shown COF-crystallinity data measured on two films annealed to different crystallinities at temperatures between 37 and 93°C. By annealing a given film the surface roughness, i.e., gloss, was held constant and therefore eliminated as a factor affecting COF.

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Résumé

Les résultats présentés montrent que des films de polypropylène rapidement refroidis contiennent des phases paracrystallines outre les phases cristallines et amorphes. La densité de la phase paracrystalline varie de 0.8890 à 0.9080 g/cm³. D'autres données montrent que la densité de films de polypropylène refroidis croît avec le vieillissement, cet accroissement étant une fonction linéaire du logarithme de l'âge du film. Les propriétés physiques du film, le module de séquence, la force d'impact, le coefficient de friction, varient non seulement en fonction de la cristallinité c.à.d. de la densité en fonction de la composition, mais également au cours du vieillissement lorsque la densité croissait avec

une légère variation de cristallinité. Un exemple de cette dépendance est l'accroissement du module de séquence de 10.9×10^6 Psi/g/cm³ au cours du vieillissement et uniquement de 3.7×10^6 Psi/g/cm³ lorsque la cristallinité croît. Par suite de l'existence de ces trois phases et du changement de densité en fonction du temps, il est possible d'avoir des films avec la même densité et des propriétés physiques différentes et inversement des films avec les mêmes propriétés physiques et densités différentes.

Zusammenfassung

Rasch abgeschreckte Polypropylenfilme enthalten neben der kristallinen und der amorphen Phase eine parakristalline Phase. Die Dichte der parakristallinen Phase bewegt sich zwischen 0,8890 und 0,9080 g/cm³. Weitere Ergebnisse zeigen, dass die Dichte abgeschreckter Polypropylenfilme während der Alterung zunimmt, wobei die Zunahme eine lineare Funktion des Logarithmus des Filmalters ist. Die physikalischen Eigenschaften der Filme, Sekantenmodul, Schlagfestigkeit und Reibungskoeffizient zeigen nicht nur eine Abhängigkeit von der Kristallinität, d.h. von der Zusammensetzungs-abhängigen Dichte, sondern auch von der Alterung, während welcher eine Dichtezunahme mit nur geringer Kristallinitätsänderung stattfand. Ein Beispiel für diese Abhängigkeit bildet die Zunahme des Sekantenmoduls um $10,9 \cdot 10^6$ PSI/g/cm³ während der Alterung und nur um $3,7 \cdot 10^6$ PSI/g/cm³ bei Kristallinitätszunahme. Wegen dieser drei Phasen und wegen der Abhängigkeit der Dichte von der Zeit kann man Filme herstellen, welche die gleiche Dichte, aber verschiedene physikalische Eigenschaften, und umgekehrt solche, welche die gleichen physikalischen Eigenschaften und verschiedene Dichte besitzen.

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