

The Influence of Resin Characteristics on the High Speed Melt Spinning of Isotactic Polypropylene. I. Effect of Molecular Weight and Its Distribution on Structure and Mechanical Properties of As-Spun Filaments

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Synopsis

The fine structures and tensile mechanical properties were characterized for high speed melt spun filaments prepared from three polypropylenes with melt flow indices in the range 12–300. It was found that spinnability and the resulting structure and properties are affected by both the weight average molecular weight and the polydispersity of the polymer. Higher tenacities, with consequent lower percent elongation to break, could be achieved by spinning the narrow molecular weight distribution polymers at high spinning speeds. This effect was associated with the development of higher birefringence in these samples. Modulus did not correlate with birefringence in the present study, but it was found to be controlled by the nature and level of crystalline order developed in the filaments.

INTRODUCTION

The melt spinning of isotactic polypropylene has long been a subject of both commercial and scientific importance. It continues to be of interest due to recent advances in the control of resin characteristics and in the technology of fiber spinning, leading to practical application of winding speeds as high as 5000–6000 m/min. The present paper reports the first in a series of investigations being carried out in our laboratories to examine the relationship of resin characteristics to the processing, structure, and properties of polypropylene filaments prepared by high speed melt spinning.

The early studies of melt spinning of polypropylene were those of Capuccio et al.,¹ Compostella et al.,² and Sheehan and Cole.³ These and later researchers^{4–16} have emphasized the importance of spinning conditions on fiber structure development and the resulting mechanical properties of the filaments. It was found that the structure and properties of as-spun filaments can be varied over a wide range by varying the processing conditions. The importance of resin characteristics such as polymer molecular weight and molecular weight distribution on the melt processability, and the resulting structure and mechanical properties of the filaments have also been recog-

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nized. Early observation of such effects were reported by Ross.¹⁶ He noted that extrusion temperatures above 260°C resulted in significant thermal and oxidative degradation of high molecular weight polypropylenes, and he used this effect to prepare samples with a range of molecular weights and molecular weight distributions from three starting materials. Although the molecular weight distribution was varied in this study, the spinning conditions were not. This and all of the studies referenced so far were confined to spinning conditions in which the take-up velocity was generally less than 1500 m/min.

More recently, many studies have been published describing the effect of high spinning speeds on the structure and properties of various fiber forming polymers.¹⁷⁻²⁷ Shimizu and his co-workers¹⁷⁻²⁰ have carried out an extensive investigation of high speed melt spinning of an isotactic polypropylene in the range of take-up velocity from 500 to 6000 m/min. The structure and properties of the spun filaments were studied as a function of take-up velocity and extrusion temperature. The effects of annealing these filaments were also studied. Their results show that density and birefringence initially increase rapidly with increased take-up velocity, but the rate of increase decreases substantially above a critical take-up velocity that varies with the extrusion temperature.²⁰ The filament tenacities and initial moduli were found to increase, while elongation to break decreased, with increasing spinning speed.

To date, there seem to be no published studies of the effect of molecular weight and molecular weight distribution on the spinnability and the resulting structure and properties of polypropylene filaments prepared by spinning in the high speed range, though previous research from our laboratory^{10,14} has partially examined these effects over a range of spinning conditions in the low take-up velocity range and for polypropylenes with melt flow indices in the range 0.4-12.0 g/10 min. The work of Nadella et al.¹⁰ showed that both structural characteristics and mechanical properties of polypropylene filaments can be correlated with the stress in the spinline at the point of crystallization. The effects of changing such parameters as extrusion temperature or polymer molecular weight could be traced to their effects on the spinline stress. In the present study we investigate such effects in the high take-up velocity range and for polymers with melt flow indices ranging up to 300. The latter resins have generally lower molecular weight and narrower molecular weight distributions than those studied previously.

EXPERIMENTAL

Materials

Three isotactic polypropylenes in the form of pellets supplied by the Exxon Chemical Co. were studied. Their basic characteristics are tabulated in Table I, together with a code naming system. The molecular weight and molecular weight distributions were supplied by Exxon. They were determined by gel permeation chromatography (GPC). It is noted that E-035 and E-300 have similar polydispersities but exhibit large differences in number and weight average molecular weights and in molecular weight at the peak maximum of the GPC curve. By contrast E-035 and E-012 possess similar peak maximum molecular weights but exhibit different molecular weight distribution. This

TABLE I
As-Received Polymer Properties

Exxon code	Melt index (g/10 min)	Our code name	Intrinsic viscosity (dL/g)	Weight average molecular weight M_w (by GPC)	Number average molecular weight M_n (by GPC)	Polydispersity $\alpha = M_w/M_n$	Molecular weight at peak max (by GPC)
3014, lot 40435	12	E-012	1.511	238,000	50,010	4.76	141,180
11852-011-001 (control)	35	E-035	1.213	170,450	78,940	2.16	147,180
3145, lot 40611	300	E-300	0.929	124,400	43,590	2.85	89,770

manifests itself in markedly different molecular weight averages and polydispersities. The molecular weight distribution of sample E-012 is considerably broader than that of either E-035 or E-300. The intrinsic viscosities were measured at 135°C in decalin.

Melt Spinning

The melt spinning was performed from a Fourne' Associates screw extruder and spinning head. The extruder had a 13-mm diameter screw. Constant throughput was maintained by a Zenith gear pump. The spinneret capillary was 3.81 mm long and 0.762 mm in diameter. Filaments were drawn down by a pneumatic device supplied by Rhone-Poulenc Fibres of France, which makes use of pressurized air to produce a suction force on the filament at the entrance of the device. The take-up velocity can be adjusted by changing the pressure of the air supply.

The quoted take-up velocities were computed using the expression

$$V = \frac{4Q}{\pi(D)^2\rho} \quad (1)$$

where Q is the mass flow rate and D and ρ are the diameter and density, respectively, of the as-spun filament.

The mass flow rate was kept constant at 1.68 g/min for all runs. Sample E-035 was spun at extrusion temperatures of 210, 230, and 250°C. Sample E-300 was spun only at 210°C due to difficulties with spinnability at higher temperatures. Sample E-012 was spun only at 230°C due to difficulties of achieving high take-up speeds at lower temperatures. Filaments were taken up at speeds of about 1360, 2240, 3700, and 5700 m/min, except for E-012 which could not be spun above 3700 m/min at 230°C without breakage occurring. The spinning conditions used in each case are given in Table II.

Density and Birefringence Measurement

A water-isopropyl alcohol gradient density column at 23°C was used to measure the density of the as-spun filaments. The samples were allowed approximately 48 h to reach their equilibrium level of displacement. The

crystallinities of the samples were then estimated using the expression

$$X_c = \frac{\rho_c(\rho - \rho_{am})}{\rho(\rho_c - \rho_{am})} \quad (2)$$

where ρ is sample density and ρ_c and ρ_{am} are the densities of crystalline and amorphous polypropylene, respectively. The values used for the latter two quantities were the same as those quoted by Samuels.²⁷ The birefringences of as-spun filaments were measured using an Olympus polarizing microscope with a Leitz Berek compensator. The diameter of the fiber was measured using a filar eyepiece.

Wide-Angle X-Ray Diffraction

Wide angle X-ray diffraction patterns of as-spun filaments were obtained using a Philips Norelco X-ray generator and a flat-plate type camera. The radiation used was nickel-filtered $\text{CuK}\alpha$ of wavelength 1.542 Å. These patterns were used as indicators of the crystalline form and qualitative features of the orientation of the spun fibers.

Mechanical Properties

Mechanical properties of the as-spun filaments were measured with an Instron tensile testing machine, after conditioning in a room at 70°F and 65% relative humidity. Fibers of initial gauge length 50.8 mm (2 in.) were stretched at a crosshead speed of 25.4 mm/min (1 in./min).

RESULTS AND DISCUSSION

Structure of As-Spun Filaments

Figure 1 shows the variation of the birefringence of as-spun filaments of sample E-035 as a function of take-up velocity at extrusion temperatures of 210, 230, and 250°C. The birefringence increases monotonically with take-up velocity at each extrusion temperature, while the rate of increase of birefringence with take-up velocity decreases with increasing spinning speed. In the lower take-up velocity range, the birefringence of the filaments increases with a decrease in the extrusion temperature. But the birefringence tends to approach the same value at the higher spinning speeds for each extrusion temperature. This behavior is qualitatively explicable on the basis of the fact that the melt viscosity of sample E-035 is such that a wide range of molecular orientations is generated in the take-up velocity range used in the present experiments. The lower melt viscosity of this sample compared to the samples studied by Nadella et al.,¹⁰ for example, requires the higher spinning speeds of the present investigation in order to generate sufficient spinline stress to develop appreciable orientation. The higher birefringences in the lower take-up velocity range of the samples spun with lower extrusion temperature reflects the higher spinning stresses for these samples due to the increase of melt viscosity in the upper part of the spinline with decreasing extrusion temperature. This is also consistent with the higher air pressure required by the

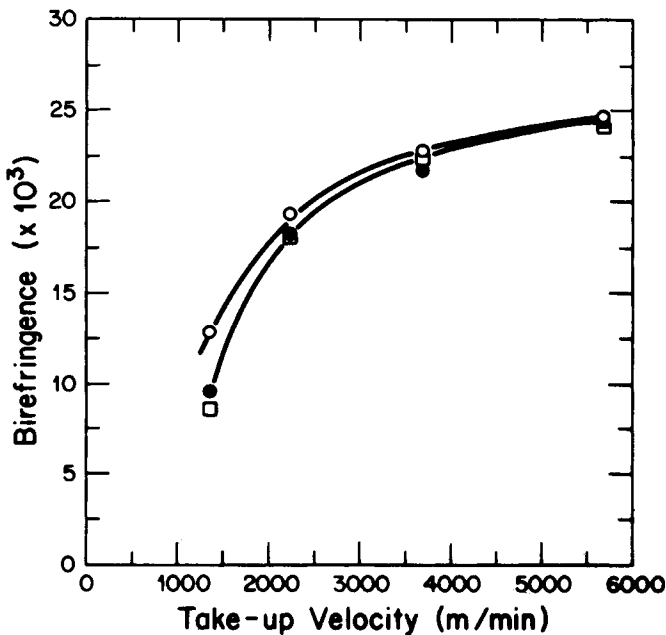


Fig. 1. Birefringence vs take-up velocity for E-035 filaments spun at three melt temperatures (°C): (○) 210; (●) 230; (□) 250.

drawdown device to produce a given take-up velocity for samples spun at lower extrusion temperature; compare runs 1-4 to 5-8 and 9-12 in Table II.

The trends shown in Figure 1 are fairly similar to those previously reported by Shimizu et al.,²⁰ except that the rapid rise in birefringences occurred at spinning speeds below 1500 m/min for Shimizu's sample. The differences between the present results and those of Shimizu et al. are readily explained by differences between the resin characteristics of the present polymer and that studied by Shimizu et al.²⁰ We will return to this discussion after presenting further experimental data.

Figure 2 shows a comparison of the way the birefringence varies with take-up velocity for the three polymers studied in this investigation. It is observed that changing the resin characteristics has a dramatic effect on the development of the birefringence in the take-up velocity range studied. The birefringence for the E-300 sample is much lower in the low take-up velocity range than for sample E-035, but the two samples appear to be approaching the same birefringence at the highest spinning speeds studied. The differences in the lower take-up velocity range again appear to reflect the lower spinline stresses developed in the lower viscosity E-300 sample. In the higher take-up velocity range the birefringence of both polymers appear to be approaching a saturation value. The high birefringence of the higher weight average molecular weight sample, E-012, in the low take-up velocity range is again consistent with higher spinline stress in that range than for the other polymers (compare the required air pressure to achieve a take-up velocity of 1360 m/min in Table II). The small variation of birefringence of sample E-012 with increase of take-up velocity, and the difficulty of spinning this sample at speeds above

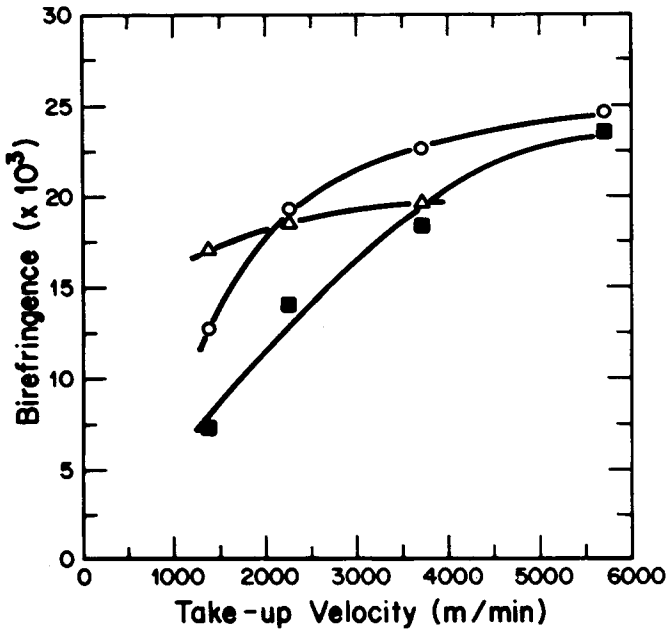


Fig. 2. Birefringence vs take-up velocity for three different resins for selected extrusion temperatures ($^{\circ}\text{C}$): (\circ) E-035, 210; (\blacksquare) E-300, 210; (\triangle) E-012, 230.

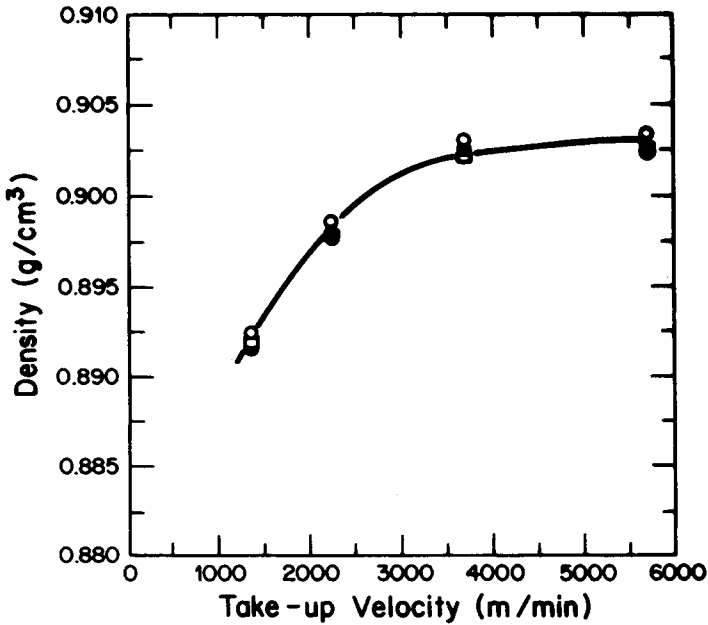


Fig. 3. Density vs take-up velocity for E-035 filaments spun at three melt temperatures ($^{\circ}\text{C}$): (\circ) 210; (\bullet) 230; (\square) 250.

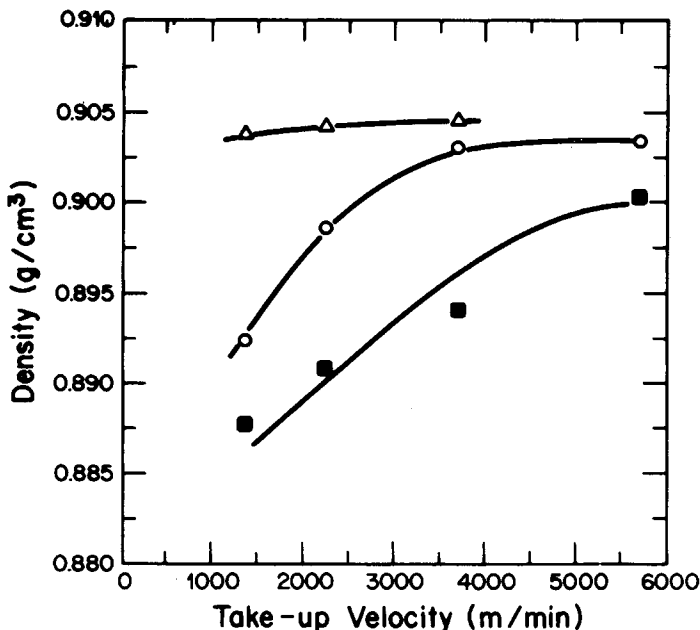


Fig. 4. Density vs take-up velocity for three different resins for selected extrusion temperatures ($^{\circ}\text{C}$): (○) E-035, 210; (■) E-300, 210; (△) E-012, 230.

3500 m/min suggests that this sample is already approaching its saturation value at speeds in the neighborhood of 1500 m/min. This is also consistent with the results on a similar, though different, polymer studied by Nadella et al.¹⁰ In that previous study the birefringence of a sample with a melt flow index of 12 was found to increase rapidly with take-up velocity in the range below 1000 m/min. The lower birefringence of filaments spun from the E-012 polymer in the high take-up velocity range, compared to filaments spun from E-035 and E-300 will be discussed in a later section.

Figure 3 shows the variation of density with take-up velocity for the filaments spun from the E-035 sample, while Figure 4 compares density data for the three different resins. The densities of the filaments are observed to increase with take-up velocity. There is little or no effect of extrusion temperature on the development of density for the E-035 resin (Fig. 3). The differences observed between resins (Fig. 4) follow trends that are similar to those followed by the birefringence values with the exception that the densities of the E-012 polymer are higher at any spinning speed than for the other polymers.

A further understanding of the structure development during high speed spinning of the present samples can be obtained from the WAXS patterns shown in Figures 5-7. These patterns clearly show that major changes in structural order accompany the changes in birefringence and density shown in Figures 1-4.

Previous studies^{3-7,10} of structure development of isotactic polypropylene during melt spinning at low take-up velocities help to provide insight into the changes occurring in the present samples. Sheehan and Cole³ and Nadella

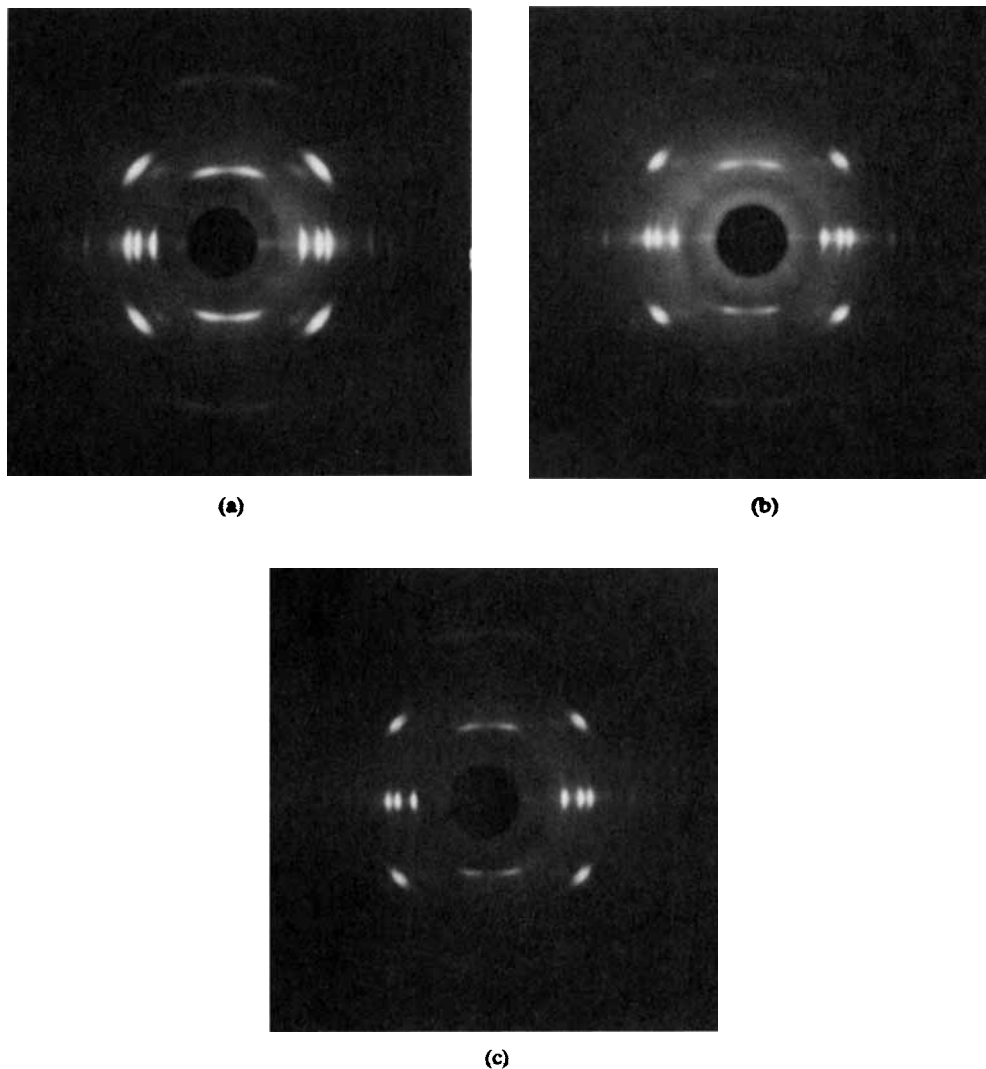


Fig. 5. WAXS patterns for the E-012 resin melt spun at an extrusion temperatures of 230°C: (a) 1360 m/min; (b) 2500 m/min; (c) 3500 m/min.

et al.,¹⁰ among others, showed that polypropylene filaments melt spun into ambient air are highly crystalline and exhibit a monoclinic crystal structure, while filaments quenched into water at low stress levels (low take-up velocities) have a paracrystalline smectic structure. The smectic structure is also known to have a lower density than the monoclinic structure.³ The smectic structure occurs under spinning conditions corresponding to high cooling rates and low spinline stresses. Decreasing the cooling rate or increasing the spinning stress at a given cooling rate tends to increase the level of stress induced crystallization and shift the structure toward the well-developed monoclinic form. We can see these effects in the WAXS patterns of Figures 5-7. The patterns for the E-012 samples show that this polymer has a monoclinic crystal structure even at the lowest spinning speed of 1360 m/min,

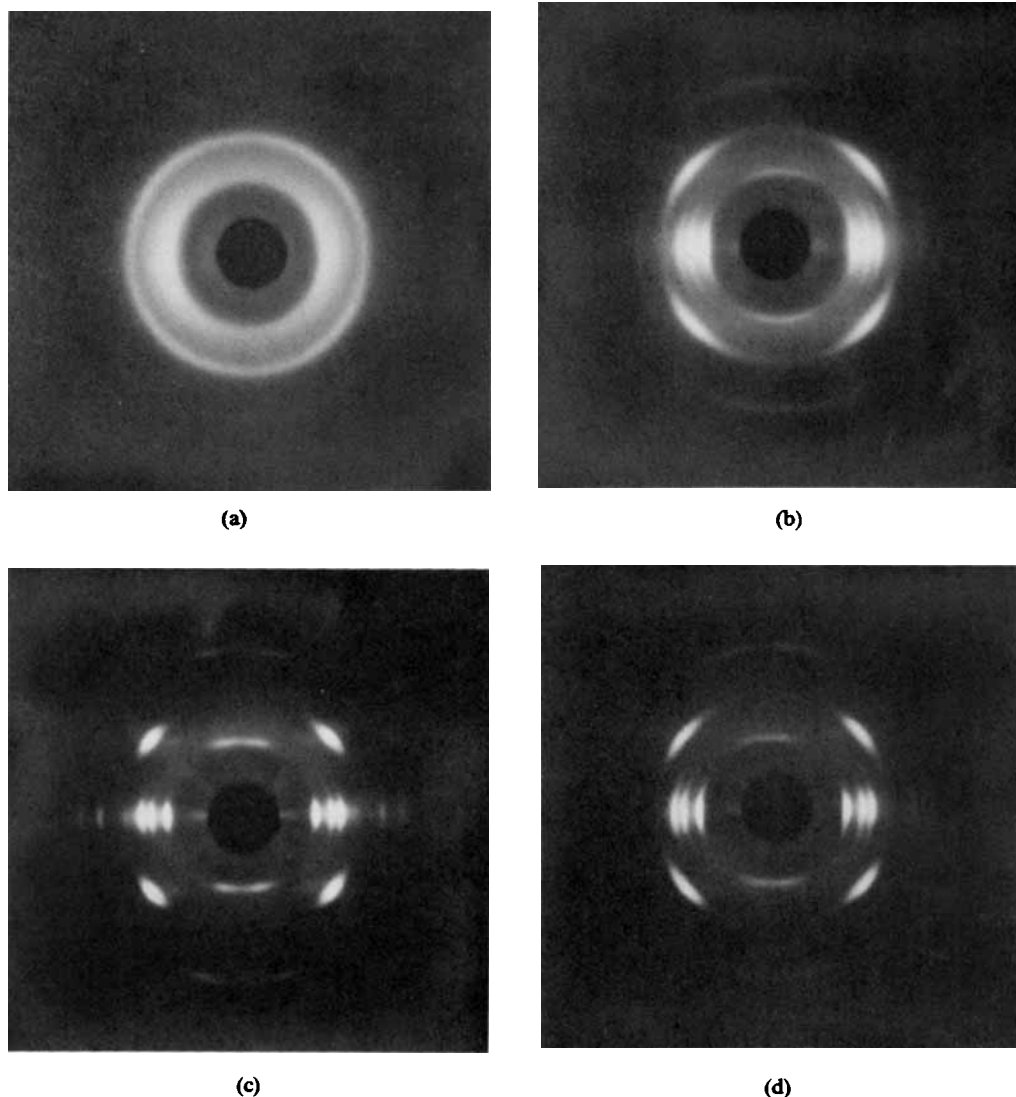


Fig. 6. WAXS patterns for the E-035 resin melt spun at an extrusion temperature of 230°C: (a) 1430 m/min; (b) 2240 m/min; (c) 3300 m/min; (d) 5700 m/min.

with the level of orientation and the perfection of the structure increasing slightly with further increases in spinning speed in spite of increased cooling rate. The filaments spun from the E-300 resin exhibit the well-developed monoclinic crystal structure only at the highest spinning speed of 5700 m/min. They exhibit paracrystalline smectic structures in the lower take-up velocity range which explains the low density of these filaments. They show a transition to monoclinic structure beginning at take-up velocities of about 3700 m/min. The behavior of the E-035 resin is intermediate between that of the E-012 and E-300 resins, with the transition to the monoclinic form occurring in a lower take-up velocity range than for the E-300 polymer, but at higher take-up velocities than for the E-012 polymer.

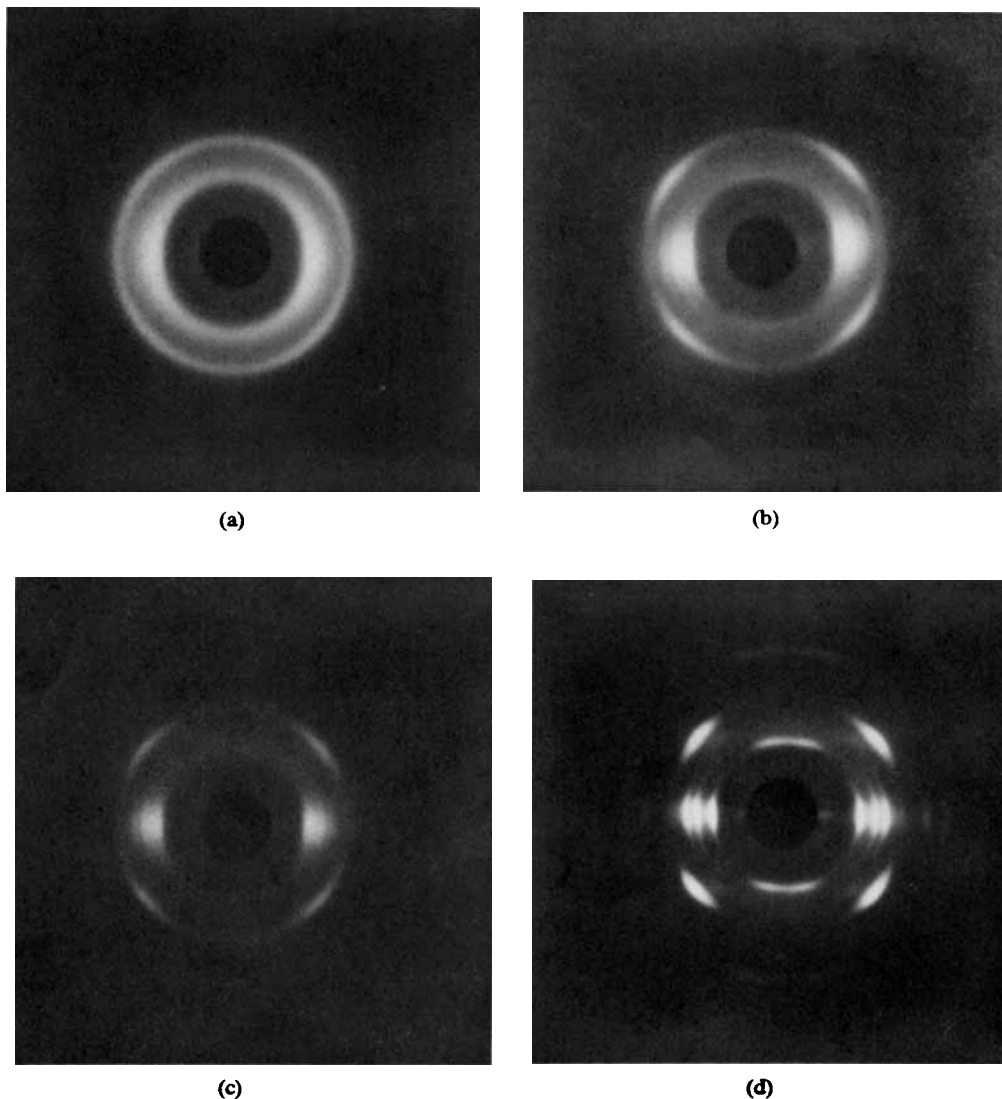


Fig. 7. WAXS patterns for the E-300 resin melt spun at an extrusion temperature of 210°C: (a) 1430 n/min; (b) 2140 m/min; (c) 3300 m/min; (d) 5600 m/min.

It is to be noted that samples spun under the same conditions (i.e., same extrusion rate and temperature, take-up velocity, and cooling medium) would have nearly identical cooling rates. The differences in structure between samples spun under the same conditions (such as run 5 and run 17) are thus due to differences in molecular weight and molecular weight distribution. These differences result in variations in the level of stress in the spinline and/or in the response of the polymer to stress. Higher stress levels occurring in the temperature range where molecular mobility is high promotes stress-induced crystallization to the monoclinic phase and a greater level of orientation and, hence, higher birefringence and density in the spun filament. It is not possible, from the present data, to ascertain whether the crystallization

kinetics are functions of temperature and stress level only or they also depend on the details of the molecular weight distribution.

Shimizu et al.²⁰ reported only the monoclinic form in polypropylene filaments spun at speeds in the range 500–6000 m/min. This was true, even though the filaments were quenched with a crossflow of -10°C air during spinning. This suggests that the balance between cooling rate and spinline stress was tipped in favor of the monoclinic structure. The polypropylene studied by Shimizu et al. had a melt flow index of 9, which might be expected to produce an even more rapid stress buildup with take-up velocity at a given spinning temperature than in our E-012 sample.

The results shown in Figures 1–7 taken together with the previous results of Nadella et al.¹⁰ and Shimizu et al.²⁰ clearly show that the molecular weight and molecular weight distribution have major effects on the development of structure during melt spinning of polypropylene. The major features of the development of orientation and morphology seem to be controlled primarily by the rheological stress in the spinline in agreement with the earlier suggestion of Nadella et al.¹⁰ This implies that the development of structure and especially such features of the spinning process as the critical take-up velocity, below which the structure of the spun filaments varies rapidly with take-up velocity and above which it varies much more slowly, are controlled by the weight average molecular weight M_w . This can be understood by considering the fact that the elongational viscosity of melts of flexible chain polymers generally increase with about the 3.5 power of M_w .^{28–31} Higher elongational viscosity associated with higher M_w produces a more rapid buildup of stress in the spinline with increasing take-up speed, which results in a more rapid buildup of orientation and crystalline order. Other features of the molecular weight distribution such as peak molecular weight or number average molecular weight do not correlate well with the results. This is easily seen by comparing the results for samples E-012 and E-035 (which have approximately the same peak molecular weight) and for samples E-012 and E-300 (which have similar number average molecular weights).

The results of the present study show that the birefringence of the lower M_w samples, E-035 and E-300, reaches higher values at high spinning speeds than the higher M_w sample, E-012. This indicates that the spinline stress or M_w is not the only variable that determines birefringence, at least in the high spinning speed range. These results suggest that molecular weight distribution plays an important role in determining the value of the birefringence.

In order to further illuminate the effect of molecular weight distribution on the development of birefringence, we plot the birefringence of our samples versus the apparent spinline stress[‡] in Figure 8. Also shown is the curve observed for several polypropylenes by Nadella et al.¹⁰ The samples studied by Nadella et al. had melt indices ranging from 0.42 to 12.0. These samples had molecular weight distributions that were broad or broader than that of sample E-012 and much broader than that of samples E-035 and E-300. The

‡In the present results no correction for air drag has been applied in the computation of the spinline stress. If this is done the discrepancy between the high speed spinning data for samples E-035 and E-300 and the previous results of Nadella et al. is even greater than shown in Figure 8.

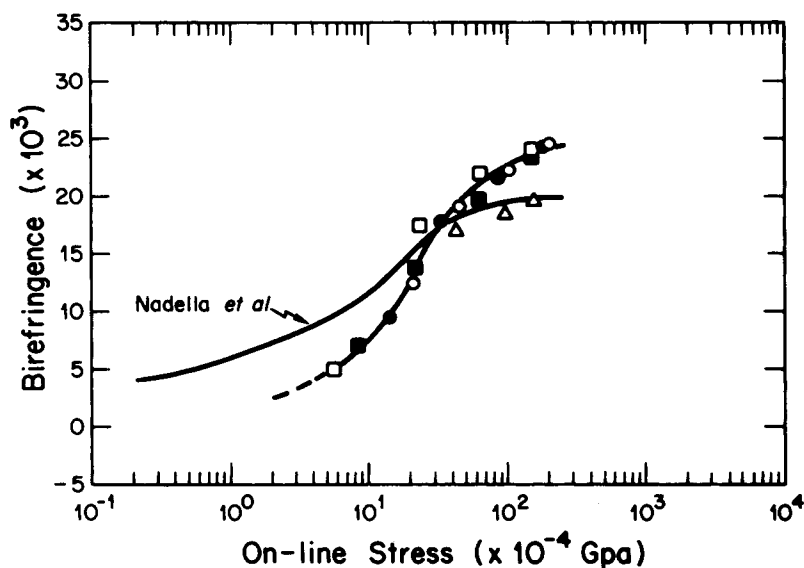


Fig. 8. Birefringence versus apparent spinline stress for the present samples compared to the previous result of Nadella *et al.*¹⁰ Resin and extrusion temp (°C): (○) E-035, 210; (●) E-035, 230; (□) E-035, 250; (■) E-300, 210; (Δ) E-012, 230.

data for the very narrow molecular weight distribution samples E-035 and E-300 form a separate curve that lies somewhat below Nadella's curve at low stresses and well above it at high stresses. It is significant that the data for the broader distribution sample E-012 lies close to Nadella's curve.

Examination of the density data of Figure 4 and comparison to the birefringence data of Figure 2 (see also Table III) indicates that the higher birefringence of samples E-035 and E-300 when spun at high speeds cannot be attributed to the development of higher crystallinity in these samples. On the contrary, the density and, hence, the crystalline order is higher in sample E-012 at about 3000 m/min than for either of the other two samples. The X-ray patterns of Figures 5-7 further show that the crystalline orientation in the E-012 sample is higher than that in the other two samples. The high birefringences of the narrow molecular weight distribution samples at high spinning speeds must be attributed to high amorphous orientation in these samples compared to the broader distribution sample. It is not clear at this time what causes the amorphous orientation of the narrow distribution polymers to reach higher values at high spinning speeds than the broader distribution polymer. We shall see in the next section that these differences in structure also cause differences in the mechanical properties.

Mechanical Properties

The tensile strength (tenacity), modulus, and percent elongation to break for selected as-spun filaments are plotted as a function of spinning take-up velocity in Figures 9-11, respectively. Data for the mechanical properties of all samples are presented in Table IV. The modulus and tensile strength of

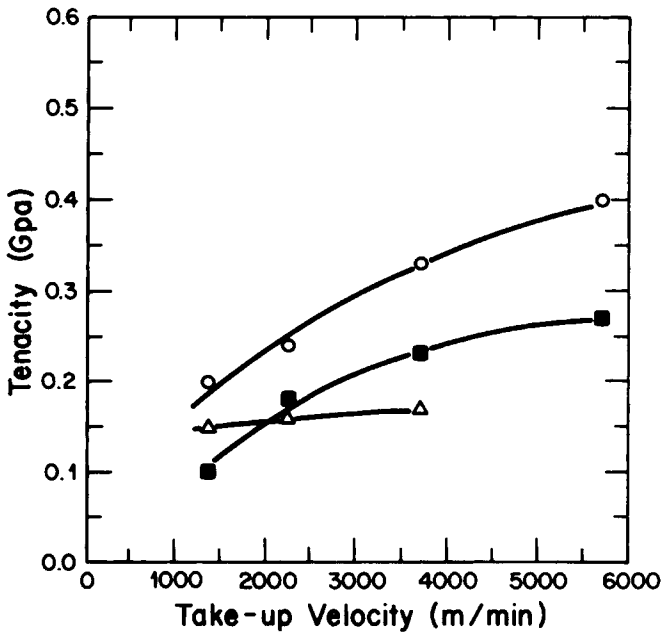


Fig. 9. Tenacity (tensile strength) vs. take-up velocity for three different resins for selected extrusion temperatures ($^{\circ}\text{C}$): (○) E-035, 210; (■) E-300, 210; (Δ) E-012, 230.

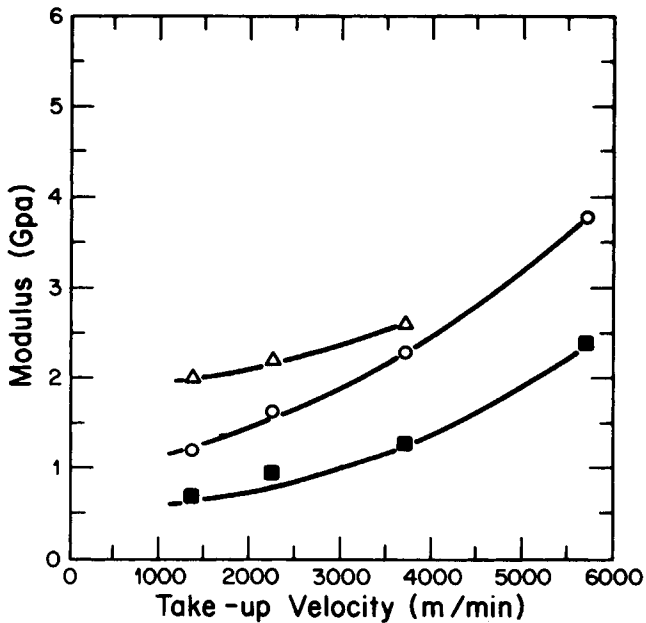


Fig. 10. Modulus vs. take-up velocity for three different resins for selected extrusion temperatures ($^{\circ}\text{C}$): (○) E-035, 210; (■) E-300, 210; (Δ) E-012, 230.

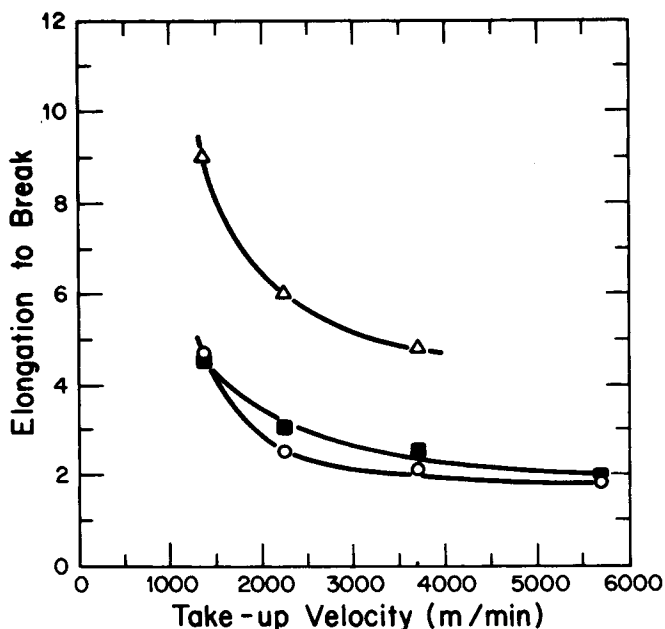


Fig. 11. Elongation to break vs. take-up velocity for three different resins for selected extrusion temperatures ($^{\circ}\text{C}$): (○) E-035, 210; (■) E-300, 210; (△) E-012, 230.

each polymer increases with increase of take-up velocity while the present elongation to break exhibits the reverse trend. It is observed from Figure 9 and Table IV that the tenacity of filaments spun from E-012 is generally lower than that of filaments spun from E-035 at equivalent spinning speeds. The difference in tenacity of the two materials increases with increase in take-up velocity. The tenacities of the filaments spun from the E-300 polymer are generally lower than those spun from the E-035 polymer, but they lie above those of the E-012 polymer in the high spinning speed range. The data plotted in Figure 11 show that the percent elongation to break of the E-012 filaments are substantially above those of the E-035 and E-300 filaments at a given spinning speed. These results appear to reflect the observed differences in orientation of the filaments as measured by the birefringence values discussed above.

Samuels^{32,33} has shown that the tenacity of drawn polypropylene filaments is controlled primarily by the orientation of the amorphous (noncrystalline) fraction of the sample and has little relationship to the crystallinity or crystalline orientation. The higher tenacity and amorphous orientation found in the filaments spun from the present narrow distribution E-035 and E-300 polymers appear to be consistent with this finding.

It should be noted, however, as shown in Figure 10, that the modulus of the E-012 filaments are much higher than that of the filaments spun from the E-035 and E-300 polymers at equivalent take-up velocities. The reason for this is due, presumably, to the higher crystalline order (i.e., the well-developed monoclinic crystal structure and higher level of crystallinity) of the filaments spun from the E-012 polymer.

TABLE IV
Mechanical Properties of As-Spun Filaments

Mechanical properties Run No.	Experimental Material																		
	E-035									E-300						E-012			
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19
Modulus (GPa)	1.2	1.64	2.3	3.8	1.08	1.33	2.25	3.32	0.96	1.27	2.1	3.12	0.68	0.95	1.24	2.4	2.0	2.2	2.6
Tenacity (GPa)	0.20	0.24	0.33	0.40	0.18	0.23	0.31	0.38	0.16	0.22	0.28	0.37	0.10	0.18	0.23	0.27	0.15	0.16	0.17
Elongation to break (%)	470	250	210	180	490	260	220	190	500	270	240	200	450	300	250	190	900	600	480

CONCLUSIONS

A study was carried out of the structure and mechanical properties of three polypropylenes that ranged in melt flow index from 12.0 to 300. These samples differed in average molecular weight and in molecular weight distribution. Two samples had narrow molecular weight distributions (similar polydispersities) but differed considerably in melt flow index and weight average molecular weights. The third sample had a broader molecular weight distribution (higher polydispersity) and higher weight average molecular weight than the other samples. The study resulted in the conclusions listed below.

(1) Sample E-035 (melt flow index 35.0) could be readily melt spun at take-up velocities up to at least 5700 m/min throughout the extrusion temperature range between 210 and 250°C. Problems were experienced in melt spinning sample E-012 (melt flow index 12.0) at high take-up velocities at an extrusion temperature of 230°C or below. The latter difficulties were due to high spinline stress values that led to cohesive fracture of the treadline. Difficulties were experienced in the melt spinning of sample E-300 (melt flow index 300) at an extrusion temperature of 230°C or above. These difficulties were caused by capillarity failure under these conditions due to the low viscosity of this polymer. It was concluded that spinnability of polypropylenes under a given set of spinning conditions is controlled by both the weight average molecular weight of the polymer, and to a lesser degree, by the polydispersity. The combination of M_w and narrow distribution exhibited by sample E-035 make this polymer have outstanding spinnability in the spinning speed range from 1500 m/min to at least 6000 m/min.

(2) The birefringence and density of all three samples increased with increased take-up velocity. For each polymer there was a critical take-up velocity below which the orientation, and density changed rapidly with take-up velocity and above which these quantities changed much more gradually. This critical take-up velocity also depended on the extrusion temperature. It was concluded that this critical take-up velocity was largely determined by the weight average molecular weight of the polymer, which in turn controls the polymer viscosity and the stress required to deform the polymer into filaments.

(3) At low spinline stresses and high cooling rates polypropylene filaments exhibit a smectic structure; at lower cooling rates or higher spinline stresses the monoclinic crystal structure is formed. In the present experiments this led to the observation of a transition in the structure of the as-spun filament from smectic to monoclinic as take-up velocity increased for a given polymer or as the weight average molecular weight of the polymer increased at a given take-up velocity.

(4) The narrow molecular weight distribution polymers achieved higher birefringence in the high take-up velocity range than did the broader molecular weight distribution sample. It was found that this result could be attributed to higher orientations in the amorphous phase for the narrow distribution polymers spun at high speeds.

(5) Under the spinning conditions studied, it was found that higher tenacities (tensile strengths) could be achieved in the narrow distribution samples than for the broad distribution sample. This appeared to be related to the

higher birefringence (orientation) of these samples which resulted from their higher amorphous phase birefringence (orientation). The percent elongation to break was generally higher for the broad distribution sample than for the narrow distribution samples and tended to decrease as birefringence increased.

(6) Initial modulus did not correlate well with birefringence for the present as-spun filaments. Instead the modulus was found to depend on the nature and level of crystalline order present.

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