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Studies on Melt Spinning. IV. Spinning Through a Ribbon Die

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Synopsis

An experimental study has been carried out to investigate effects of stretch ratio on molecular orientation in polypropylene monofilaments which are melt spun from a ribbon die into a water bath with an adjustable air gap distance between the two. By varying the air gap distance and the rate of stretching, a variety of filaments of different molecular orientations were obtained. Measurements were taken of fiber birefringence of finished filaments under a polarizing microscope with camera attachment and mercury lamp. It has been found, according to the already established relationship between the molecular orientation and birefringence, that the molecular orientation in polypropylene filaments is increased with the rate of stretching. Two other interesting observations were made. One was that the filaments form crimps whose frequency increases with the rate of stretching. The other was that the phenomenon of draw resonance was observed when the rate of stretching was increased beyond a certain critical value.

INTRODUCTION

It has been a well-established fact that the mechanical properties of a finished fiber are strongly influenced by the orientation of molecules in the fiber. The extent of molecular orientation in the finished fiber may be controlled in two steps. One is in the spinning process itself, during which the molten threadline is stretched and cooled. The other is in the after-treatment process, where the already formed thread is heated again and stretched.

The former is of greater importance, because in the fiber-forming step two structuring processes take place simultaneously: crystallization and molecular orientation. The orientation of molecules occurs in the direction of the axial velocity gradient, and hence the extent of molecular orientation depends on the elongation rate and also on the temperature profile along the spinning way. Because of the cooling effect, crystallization and orientation in melt spinning proceed in a highly complicated manner. When the crystallization rate is considerably lower than the rate of orientation, the macromolecules orient along the fiber axis, but form little crystalline regions. Hence, fibers spun from such polymers consist of amorphous, though axially oriented molecules (e.g., polystyrene).

More than a decade ago, Ziabicki and his co-workers[1-4] made a pioneering investigation of the effect of spinning conditions on the orientation of molecules in melt spinning, both theoretically and experimentally. For the theoretical study, these authors considered the orientation of rigid ellipsoids and flexible coiled chains for the molecular segments in the elongational flow field and defined a "coefficient of orientation" which was suggested for use in determining the extent of orientation of macromolecules in melt spinning. For the experimental study, measurements of fiber birefringence and x-ray patterns were made to correlate the orientation of macromolecules with spinning conditions and the molecular structure of the materials investigated.

On the other hand, Katayama[5] has measured the birefringence of a molten threadline along the spinline and has shown how much the stretching affects the orientation of molecules, which ultimately controls the mechanical properties of the finished filament. Other similar attempts are reported in a few recent contributions to the literature.[6,7]

In the present paper, which is the fourth of this series, we shall discuss our recent measurements of fiber birefringence in melt spinning, with particular emphasis on orientation of macromolecules under various spinning conditions. For the study, a ribbon die (1 mm × 10 mm) was used to produce polypropylene monofilaments. During the spinning experiments the phenomenon of draw resonance was observed, also, when the rate of stretching was increased beyond a certain critical value.

EXPERIMENTAL

The spinning apparatus and experimental procedures used for the present study have been described in a previous paper by Han and Lamonte.[8] In the present study, however, a ribbon die was used, having 1 mm on the short side and 10 mm on the long side of the rectangle.

The material used was polypropylene (Enjay Resin E115), and the extrusion temperature was 180°C. The extrudate upon exiting from the die traveled first through the ambient air and then through a quenching water bath. The traveling distance through the air (air gap distance) was varied by controlling the level of water in the quench bath. A take-up device was placed after the water bath, and different stretch rates were applied by varying the speed of the motor attached to the take-up roller. Table I gives a summary of the spinning conditions used in the study.

Measurements were taken of the throughput rate, the take-up velocity, and the air gap distance. At each take-up speed, extrudate samples were collected. During the course of experiment, the phenomenon of draw resonance was also observed, which was extensively discussed in a previous paper, part III of this series.[9] When draw resonance occurred, motion pictures were taken, using a movie camera (Bolex Super 8), of the pulsing extrudate near the spinnerette.

Three other interesting observations were made with the extrudate samples collected. In the first, extrudate samples were placed under a polariz-

TABLE I
A Summary of the Operating Conditions for Ribbon Die Extrusion[a]

Sample code	Take-up velocity, m/min	Apparent stretch ratio
a	105.5	69
b	182.0	119
c	243.0	159
d	441.0	289

[a] Die dimensions; width = 10 mm; thickness = 1 mm; material = polypropylene (Enjay Resin E115); throughput rate = 11.0 g/min; average velocity in the die (V_0) = 1.53 m/min; melt temperature = 180°C.

ing microscope and the stress patterns were examined by photographing the fiber birefringence in a filament. This was done in order to investigate a change in molecular orientation as the stretch rate is changed. According to the literature,[1-4,10,11] the molecular orientation is correlatable with the phenomenon of birefringence. In the second, the fiber-crimping phenomenon was examined by taking pictures of extrudate samples. This was done in order to find any correlation between the crimp frequency and stretch ratio. In the third, the filament samples collected were cross sectioned, and pictures were taken of the filament cross section in order to investigate if variations in the stretch rate would affect the fiber shape.

RESULTS AND DISCUSSION

Effect of Stretch Rate on Fiber Birefringence

Figure 1 shows some representative pictures taken of birefringence in samples of ribbon fiber, which were collected under various stretching conditions. More precisely speaking, what is shown in Figure 1 is the retardation. In order to quantitatively determine fiber birefringence, both retardation and thickness of the samples had to be measured, together with the stress optical coefficient. Measurement of birefringence has long been used as a means of investigating orientation of molecules. Analysis of birefringence measurement with crystalline polymers, like the polypropylene used in the present study, is much more complicated than with amorphous polymers, such as polystyrene. This is because, in crystalline polymers, the crystalline phase becomes much more oriented than the amorphous phase when the polymer is stretched. This is one of the main reasons why much of the fundamental study has been made using polystyrene.[10,11]

However, the primary objective of the present study was to qualitatively investigate the effect of stretching on the degree of molecular orientation in the extrusion of ribbon fiber. Use of a ribbon die, instead of a circular die, has an advantage in that, when polarized light is passed through the sample, there is very little edge effect on the birefringence measured with a very thin flat film. It is now seen from Figure 1 that fringes (or retardation) increases as the stretch rate is increased, indicating that the degree of

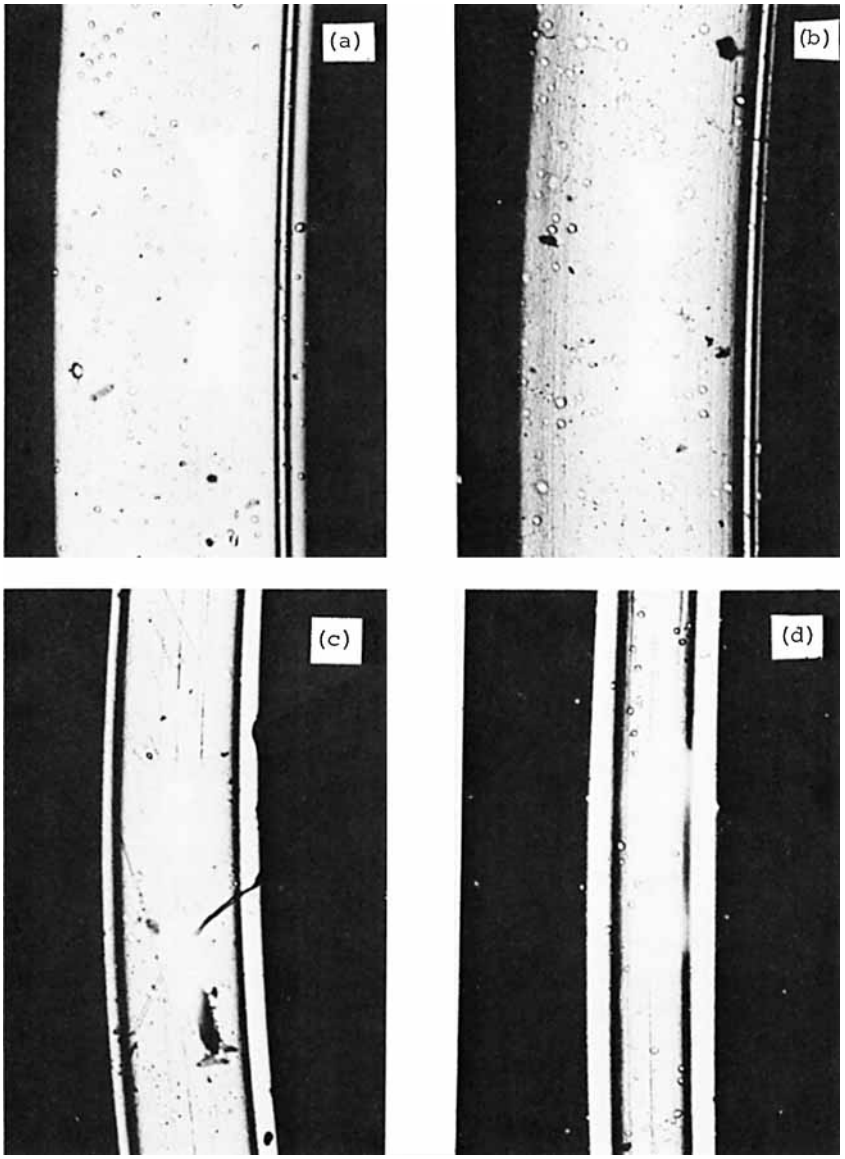

Fig. 1. Representative pictures of fiber birefringence (retardation), with constant values of throughput rate and hole size, of polypropylene monofilaments as seen through crossed polarizers: (a) $V_L/V_0 = 69$; (b) $V_L/V_0 = 119$; (c) $V_L/V_0 = 159$; (d) $V_L/V_0 = 289$.

orientation increases with stretch ratio. It seems important to point out that the stretch ratio, V_L/V_0, includes the take-up velocity, V_L, hole size (long and short side of rectangular hole), and throughput rate, Q. One may then surmise that the results shown in Figure 1 could have been different if the same V_L/V_0 value had been obtained by varying the hole size

or throughput rate. It should be noted therefore that the results shown in Figure 1 were obtained with constant values of hole size and throughput rate, but with increasing values of V_L.

Since the stretching was uniaxial in the present study, orientation of molecules would have occurred mostly in the direction of stretching. This is seen clearly from Figure 1, where interference fringes run parallel to the axis of the filament. Note also in Figure 1 that fringes are more closely spaced at the edges of the filament than at the center. This is attributable to the fact that cooling is faster at the edges of the filament than at the center, giving rise to different fringe orders (i.e., different orientations) across the filament width. That is, qualitatively speaking, fringe order increases regularly from zero to some maximum value going from the edge of the filament to the center, implying that more orientation of molecules would occur at the center than at the edge of the filament.

Although the relations between birefringence, orientation, and stress become much more complicated for crystalline polymers (e.g., polypropylene, polyethylene) compared to those for amorphous polymers, we can still qualitatively state that the degree of orientation in the ribbon samples of polypropylene tested is proportional to the amount of birefringence. Then we can say from Figure 1 that the sample showing more fringes (or retardation) has a higher degree of orientation than the one showing fewer fringes. That is, the degree of molecular orientation increases with stretch ratio, which is as expected.

Of course, a quantitative investigation requires measurements of fringe orders, which is then to be related to stress distributions in a sample. This was beyond the scope of the present study, and future study will be concerned with this investigation.

Finally, it should be mentioned that although the air gap distance was varied in the experiment, there was no appreciable effect observed of this variable on birefringence. This was mainly because in the experiment the air gap distance was varied somewhere between 3 to 5 feet below the ribbon die surface. This implies that the filaments were cooled off considerably before they actually reached the water bath. However, the situation could have been quite different if the air gap distance had been shortened considerably, say, within several inches below the die surface. Future study will be concerned with the effect of air gap distance on fiber birefringence by modifying the experimental setup.

Effect of Stretch Rate on the Formation of Fiber Crimp

Thin ribbons wound up on the take-up roller were found, interestingly enough, to form crimps. Figure 2 shows pictures of some representative samples collected at different take-up ratios. It is seen from these pictures that the frequency of fiber crimps increases with stretch ratio. Based on the observations made above that orientation of molecules increases with stretch ratio, the increase in crimp frequency with stretch ratio is believed to be a consequence of the increase in molecular orientation.

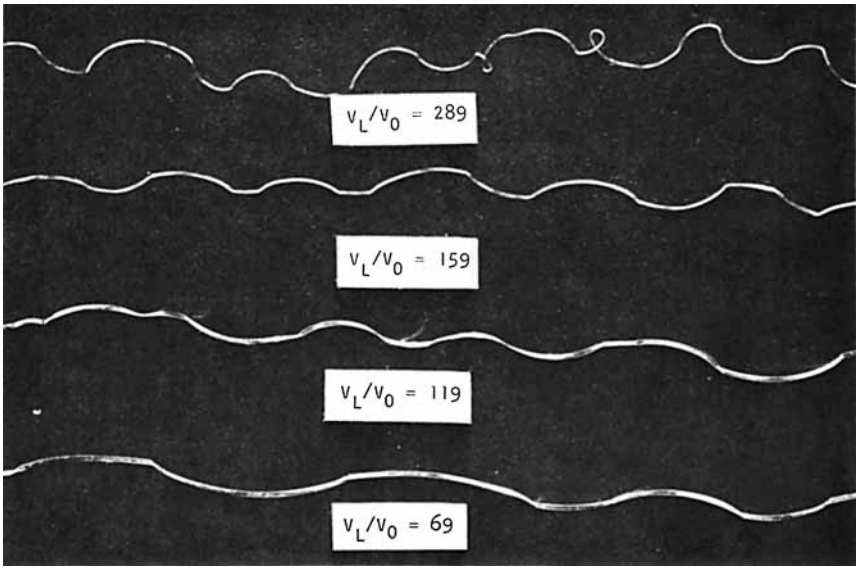

Fig. 2. Pictures of the crimps formed by polypropylene monofilaments spun at varying jet stretches.

The Phenomenon of Draw Resonance

The phenomenon of draw resonance has been observed in the present study also, as in a previous study,[9] which was concerned with spinning through circular spinnerette holes. It should be noted that draw resonance occurred above a certain critical value of stretch rate. Earlier, Bergonzoni and DiCresce[12] reported their observations made on this phenomenon, also in the extrusion through a ribbon die.

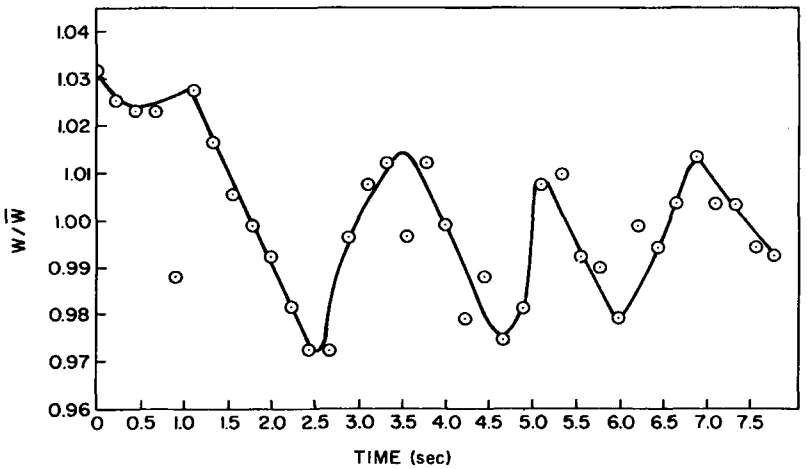

Fig. 3. Plots of the normalized ribbon width $W(t)/\bar{W}$ vs. time for a pulsing polypropylene monofilament at a high jet stretch ($V_L/V_0 = 289$).

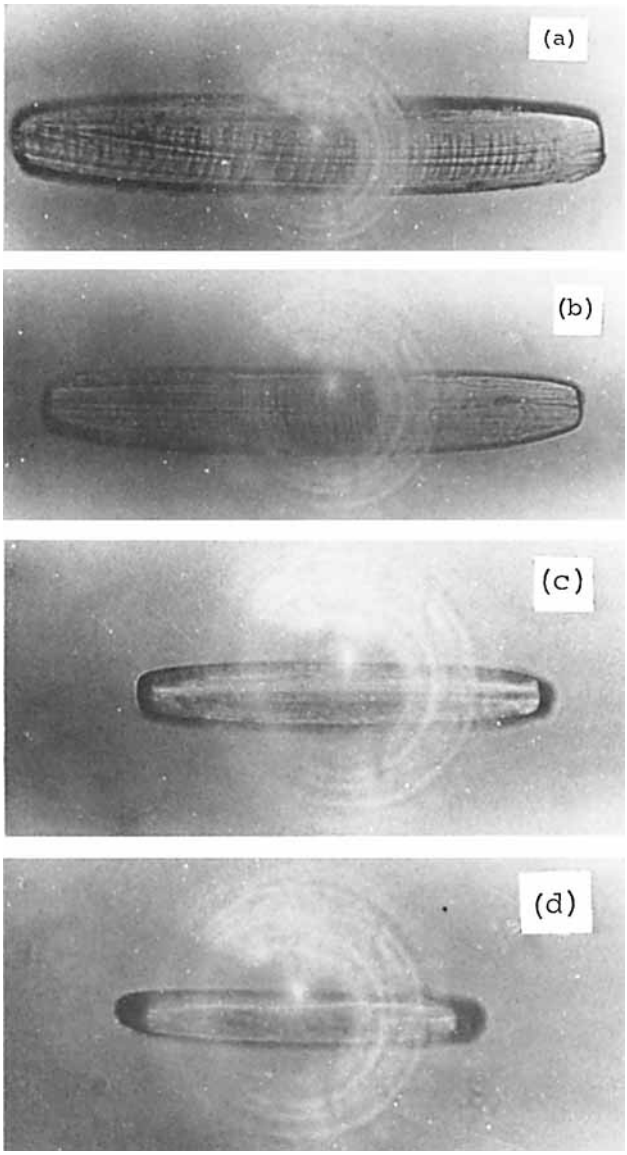

Fig. 4. Pictures (about 400×) of the cross section of polypropylene monofilaments spun at varying jet stretches: (a) $V_L/V_0 = 69$; (b) $V_L/V_0 = 119$; (c) $V_L/V_0 = 159$; (d) $V_L/V_0 = 289$.

Figure 3 shows a plot versus time of the ratio of width to average width, $W(t)/\bar{W}$, for the pulsing filament. It is seen in Figure 3 that the resonant behavior of the filament width is very similar to that reported in an earlier paper by Han et al.[9] It should be noted that the plot given in Figure 3 was prepared by reading off the projected images of the motion pictures which were taken during the extrusion experiments. Details of the procedure by means of which data were analyzed are as described before.[9]

In the present study, it has also been found that a further increase in stretch rate leads to the breakage of filament. At present, however, there is no clear-cut theory available which can explain the cause (or causes) of the onset of draw resonance. The fact that polypropylene is one of the very few polymers which are reported in the literature to have exhibited draw resonance seems worth noting. In view of this fact, a theory which makes use of more than just the phenomenological approach would seem to have a better chance for explaining the as yet unanswered question as regards the origin of the onset of draw resonance.

Shape of Extruded Filaments

The ribbon die used for the present study has a rectangular spinnerette hole with an aspect ratio of 10. Flow of polymer melts through rectangular holes is an interesting problem in itself. Recently, Han[13] has reported some interesting observations on the swelling behavior of extrudates exiting from rectangular dies.

In the present study, extrudate samples were collected which were extruded at varying stretch rates. Figure 4 gives pictures of filament cross section at four different stretch rates; it is seen that swelling at (or near) the center of the long side of the rectangle is more pronounced than at the short side, and yet the shape itself is little influenced by jet stretch, V_L/V_0. In an earlier paper,[13] Han has offered an explanation for this by measuring normal stresses at the die wall on both long and short sides of the rectangle.

In a more recent study, Han and Park[14] have examined further the problem of making fibers having noncircular cross sections.

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