

On-Line Studies of Structure Development during Melt Spinning of Poly(butylene Terephthalate)

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Synopsis

An on-line study of structure development during poly(butylene terephthalate) melt spinning was carried out. Two polymers with different molecular weights (intrinsic viscosities of 0.75 and 1.0 dL/g) were used. The range of take-up velocities studied was 1500 to 4500 m/min. On-line measurements included diameter, temperature, birefringence, and tension. The phenomenon of diameter thinning (necking) was observed for both polymers at take-up velocities of 3500 and 4500 m/min with a mass throughput of 4 g/min. At a constant mass throughput, the distance from the spinneret at which the necking occurred varied with take-up velocity and molecular weight of the polymer. Increasing the take-up velocity at constant mass throughput caused an increase in cooling rate and a slight increase in the rate at which the temperature decreased with distance from the spinneret. A small but detectable change in the rate of temperature decrease was observed at a position near or just beyond the formation of the neck. It is suggested that this effect is due to the increased heat transfer caused by the rapid increase in filament velocity and increased surface to volume ratio in the neck. Increased take-up velocity also caused necking to occur at higher temperature, as did an increase of polymer molecular weight. Birefringence increased with distance from the spinneret and indicated substantial molecular orientation was developed in the filament prior to the necking zone. A sharp increase of birefringence in the necking zone was observed for take-up velocities of 3500 and 4500 m/min. A discussion of the mechanism of neck formation is presented, and it was concluded that necking is intimately associated with stress-induced crystallization in PBT. An increase of spinline stress resulting from either an increase of take-up velocity or an increase of molecular weight can cause stress-induced crystallization and, hence, necking to occur nearer the spinneret and at higher temperature. For a given polymer this leads to filaments with higher levels of crystallinity, crystalline orientation, and crystalline perfection (greater crystal size). These changes in morphology result in changes in the filament mechanical properties. The effect of molecular weight change on the structure and properties is complicated by the fact that the development of crystallinity seems to be affected by the molecular weight independent of the spinline stress.

INTRODUCTION

Poly(butylene terephthalate) (PBT) is an important thermoplastic resin for molding, film, and synthetic fiber. As a fiber, it is being used in more and more applications because of its ability to be processed into filaments with high resilience and dyeability. These facts make basic investigation of fiber formation processes and the structure and properties of PBT fiber of considerable interest.

In a previous paper,¹ we reported a systematic study of structure and tensile mechanical properties of PBT as-spun filament prepared by melt

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spinning with take-up velocities in the range 1000–5600 m/min. It was found that PBT has a higher crystallization rate during spinning compared with poly(ethylene terephthalate) (PET). The spun fibers contain primarily α -form crystals, although a small quantity of β -form crystals existed in the samples made at the highest take-up velocity. Crystal size varied with crystallographic direction but generally increased as take-up velocity increased.

It is necessary to proceed with parameter measurements on-line in order to further explore the origin and development of structure during the spinning process. We present such on-line studies here for PBT, and this paper continues our on-line studies of fiber structure formation. Previous studies from our laboratory²⁻⁸ have made on-line diameter, birefringence, temperature, and X-ray diffraction measurements on polyethylene, polypropylene, polyamide, and PET.

EXPERIMENTAL

Materials

Two PBT resins were used; both were supplied by Celanese Plastics and Specialties Co. and are sold under the trade name Celanex 2000. One resin had an intrinsic viscosity (I.V.) of 0.75 dL/g while the second resin had an I.V. of 1.0 dL/g. According to the correlation established by Borman,⁹ this corresponds to number average molecular weights of 23,600 and 32,800, respectively.

The chips were dried in a laboratory vacuum oven for 8 h at 120°C prior to extrusion.

Melt Spinning

The filaments were spun using a screw extruder with a 13 mm diameter screw. The melt was metered to a monofilament spinneret by a gear pump; the extrusion temperature was 252°C. The single capillary die had a diameter of 0.762 mm and an L/D ratio of 5.0. The spinline was 2.0 m in length. The mass throughput of the polymer was carefully adjusted and controlled by the gear pump at 4.0 g/min. A pneumatic device (aspirator) supplied by Rhone Poulenc Fibres, Inc. was used to draw the filaments down. The take-up velocities of the as-spun filaments were adjusted by changing the pressure of the compressed air supplied to the drawdown device. Samples with take-up velocities in the range of 1500–4500 m/min could be readily prepared with this technique. Since no winding device was used, the samples were collected in a relaxed state rather than being wound on a bobbin at constant tension.

On-Line Measurements

We describe here on-line measurements of filament diameter profiles using noncontact electrooptical technology, temperature profiles by means of IR microscopy, and birefringence profiles using an optical microscopy technique. Take-up force was also measured via a sensitive tensiometer measuring head connected to a digital computer.

Diameter Profile

In the past, most measurements of diameter profiles along the spinline were done by direct observation via microscope. Vibration of the filament and vision error increased measuring deviation, especially for the case that a pneumatic device is used to provide the drawdown.

In the present investigation, a Zimmer diameter monitor was used, permitting error free measurements even at filament linear velocities of 10,000 m/min and at lateral speeds of up to 500 m/min. This diameter monitor is based on a noncontact electrooptic back illumination principle. It was connected to a digital computer that allowed very rapid data acquisition (> 2000 readings within 3 min) and statistical analysis of the resulting values. The accuracy of diameter measurement is much improved with this technique compared to use of microscope methods.

Temperature Profile

A noncontact infrared microscope system, the Barnes RM-2B, was used. The system was modified by mounting a small heater in front of the microscope lens. The temperature of the heater could be adjusted and was controlled and measured independently of the IR microscope. The filament temperature measurement was based on a detectable temperature difference between the background (heater) and the object (running filament). The IR microscope was moved up and down along the spinline until no difference signal could be observed when the filament is present and when the background is observed alone (without the filament). The filament temperature at this point was regarded the same as the heater temperature.

Birefringence Profile

An Olympus polarizing microscope equipped with a filar eyepiece and a Berek compensator was mounted on a special support in order to easily move the microscope up and down along the spinline. The retardation was measured using a guide placed well below the measuring point in order to render negligible interference caused by the guide.

Take-Up Force Measurement

The force was determined by inserting a highly sensitive tensiometer measuring head into the spinline at a point 190 cm below the spinneret. In order to get precise results the measuring head was connected to a microprocessor for data acquisition and averaging. A typical sample involved the collection of 200 readings of the force within 1 min. Before measuring, the whole system was calibrated by methods suggested by the manufacturer.

RESULTS AND DISCUSSION

Diameter Profiles

Figure 1 shows the statistical distribution of diameter values measured at 150 m from the spinneret for filaments spun at four different take-up veloci-

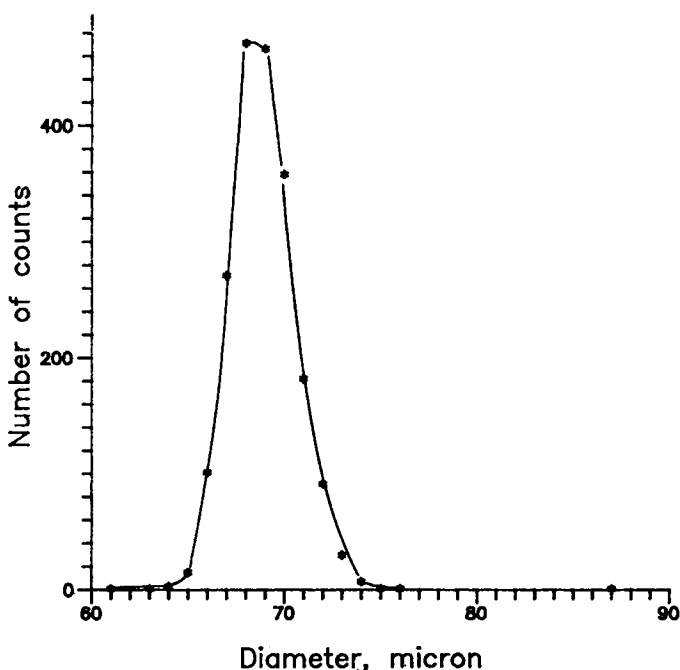


Fig. 1(a). Statistical distribution of diameter values measured at 150 cm from the spinneret. Sample IV = 0.75, mass throughput $Q = 4$ g/min. Take-up velocity $V = 1500$ m/min.

ties. These results illustrate the fact that at each position and at each take-up velocity the diameter values varied approximately in accord with a normal distribution. The largest number of counts corresponds to the most probable value of diameter. At a given position the diameter distribution is narrowed with an increase of take-up velocity. This appears to occur because the higher take-up force associated with higher spinning speed tends to stabilize the spinline and reduce vibration and lateral motion of the filament.

Figures 2 and 3 present diameter profiles along the spinline for low molecular weight and high molecular weight polymer, respectively. The plotted diameters are the most probable values. Because of the way the equipment was set up for these experiments, it was not possible to measure the diameter on-line at distances greater than 150 cm from the spinneret. The values plotted at 200 cm from the spinneret are final diameters measured on as-spun filaments.

The data indicate that at take-up velocities of 1500 and 2500 m/min, the diameters draw down smoothly, but have not yet reached their final values at a distance of 150 cm from the spinneret. However, at higher take-up velocities of 3500 and 4500 m/min, the drawdown occurred more quickly to essentially the final value before reaching 150 cm from the spinneret. In addition, a sharp decrease of diameter, i.e., a necking phenomenon, was observed at the higher take-up velocities of 3500 and 4500 m/min. After necking, the diameter of the filament remained almost constant, decreasing only slightly, with increasing distance from the spinneret. The position where necking occurs is closer to the

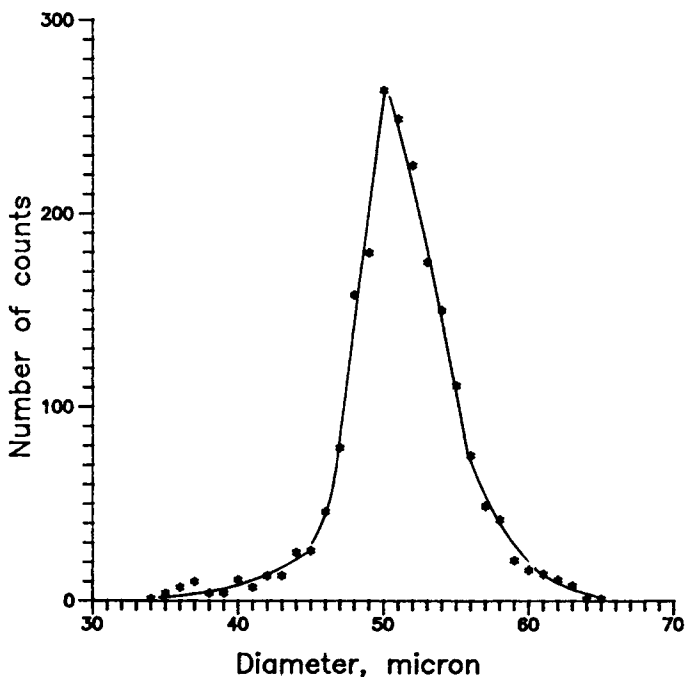


Fig. 1(b). Statistical distribution of diameter values. Data are the same as Figure 1(a) except take-up velocity $V = 2500$ m/min.

spinneret for 4500 m/min than for 3500 m/min. The magnitude of the drawdown ratio occurring in the neck increases with increase of take-up velocity.

It should also be noted that the position of the rapid diameter thinning is not completely stable, but it moves up and down the threadline over a distance of a few centimeters.

The effect of molecular weight on filament diameter profile can be found by comparison of Figure 2 with Figure 3. Diameter thinning took place earlier for the higher molecular weight polymer than for the lower molecular weight one at the same take-up velocities and same mass throughput. In the case of the high molecular weight polymer there is an indication that necking may occur at 2500 m/min at a position just below that which is accessible with the diameter monitor; i.e., at about 150–160 cm from the spinneret. Unfortunately, this could not be confirmed. The differences observed between the two molecular weights probably result because of higher rheological stress imposed on the running filament for the higher molecular weight polymer than for the lower molecular weight polymer.

X-ray diffraction patterns for as-spun filaments of the lower molecular weight polymer are shown in the inset in Figure 2. Since T_g for PBT is well above room temperature, crystallization of PBT during spinning can be confirmed by X-ray patterns of PBT as-spun filaments. These patterns show that PBT crystallized on the threadline during spinning, even at the lowest spinning speed. However, the level of crystallinity and perfection of the

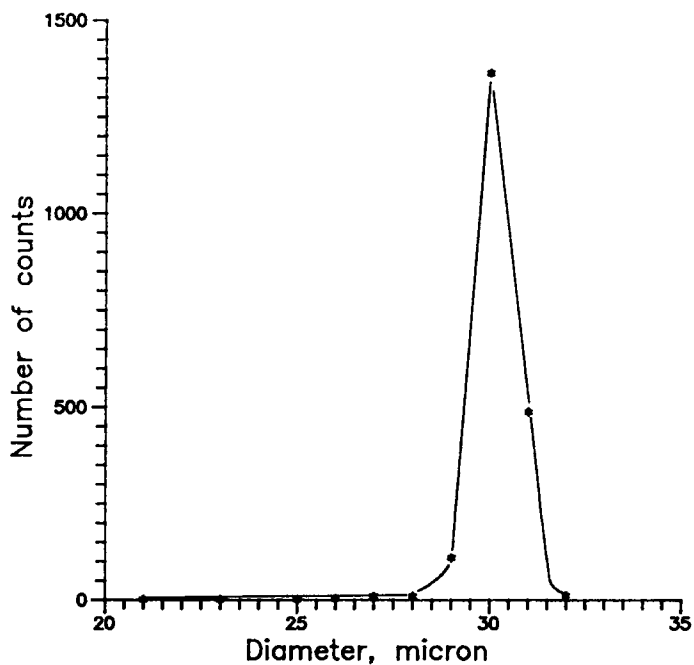


Fig. 1(c). Statistical distribution of diameter values. Data are the same as Figure 1(a) except take-up velocity $V = 3500$ m/min.

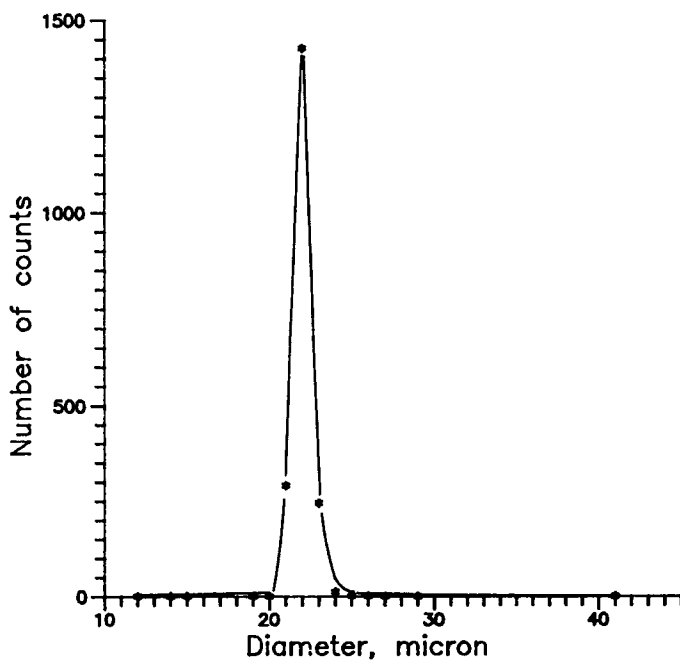


Fig. 1(d). Statistical distribution of diameter values. Data are the same as Figure 1(a) except take-up velocity $V = 4500$ m/min.

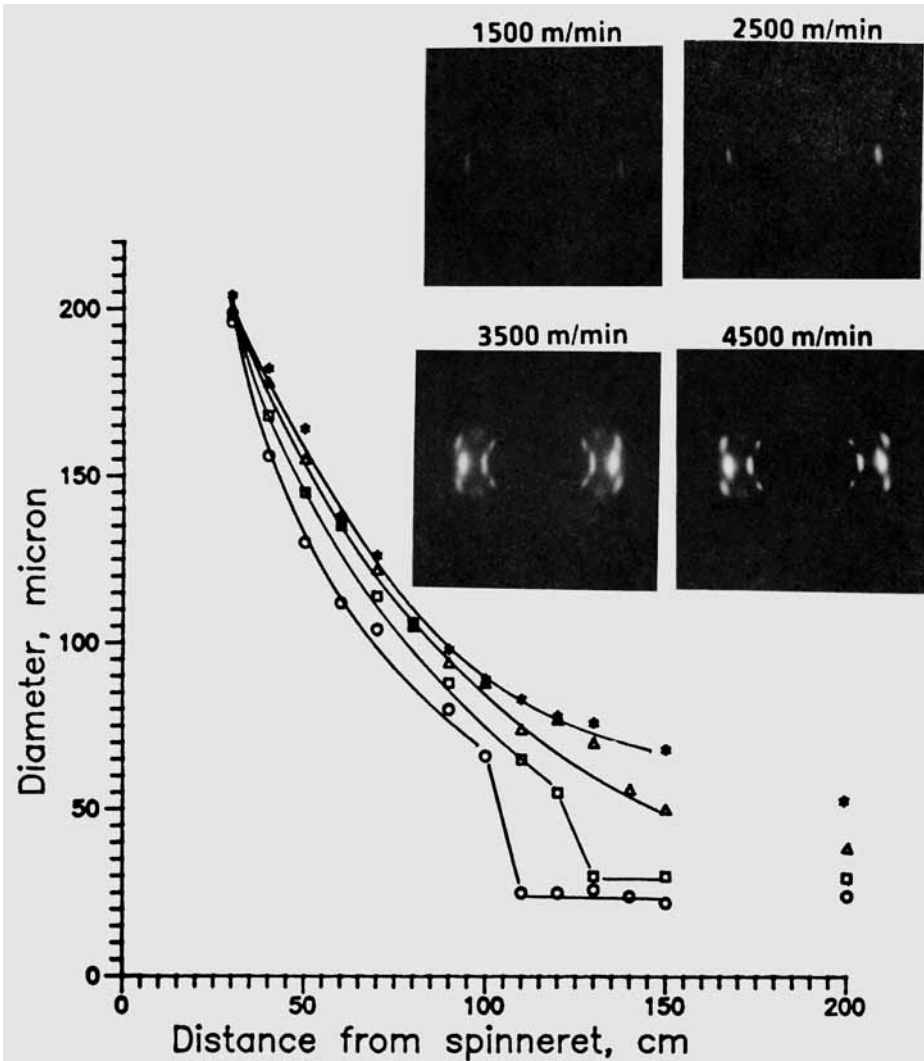


Fig. 2. Diameter profile measured along the spinline for low molecular weight PBT with a mass throughput of 4 g/min: (*) 1500 m/min; (Δ) 2500 m/min; (\square) 3500 m/min; (\circ) 4500 m/min. Inset shows X-ray diffraction patterns of as-spun filament.

crystals vary with take-up velocity. Filaments spun at 3500 and 4500 m/min contain highly oriented, well-developed crystals. Filaments spun at take-up velocities of 1500 and 2500 m/min contain less well-developed crystals and significantly lower levels of crystallinity. These effects for the as-spun filaments are described in more detail in our previous paper.¹

Comparison of the diameter profiles with the X-ray patterns in Figure 2 suggests that necking is intimately connected to the stress-induced crystallization phenomenon. The filament strain hardening that accompanies crystallization prevents further draw down after the neck.

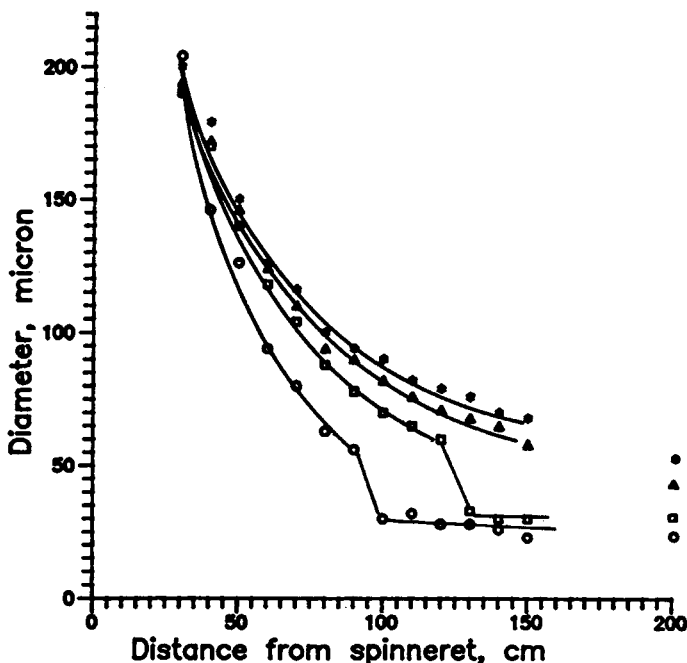


Fig. 3. Diameter profile measured along the spinline for high molecular weight PBT with a mass throughput of 4 g/min: (*) 1500 m/min; (Δ) 2500 m/min; (\square) 3500 m/min; (\circ) 4500 m/min.

Temperature Profiles

The temperature distribution along the spinline is important because the elongational viscosity, molecular orientation (relaxation), and crystallization of the polymer melt are all strongly influenced by the temperature.

Figures 4 and 5 show the temperature profiles for different molecular weight polymers. It is seen that an increase of take-up velocity causes a slight increase in the rate at which temperature decreases along the spinline. The temperature decreases smoothly with an increase of distance from the spinneret in the range of spinline lengths studied, except for the highest take-up velocity of 4500 m/min.

For high molecular weight polymer spun at 4500 m/min, a noticeable increase of the slope of the temperature profile occurs at a temperature of 150–140°C and in the range of distance 90–100 cm from the spinneret. For the low molecular weight polymer, a similar change in the slope of the temperature profile occurs at about 140–130°C and around 100–110 cm from the spinneret. These positions correspond to zones of diameter thinning (necking), as observed in Figures 2 and 3. An increased rate of cooling of the filament occurring immediately after the zones of diameter thinning probably can be traced to the fact that diameter thinning increases the rate of heat transfer due to an increase in the surface area per unit volume and a rapid increase of the velocity of the running filaments through the surrounding air.

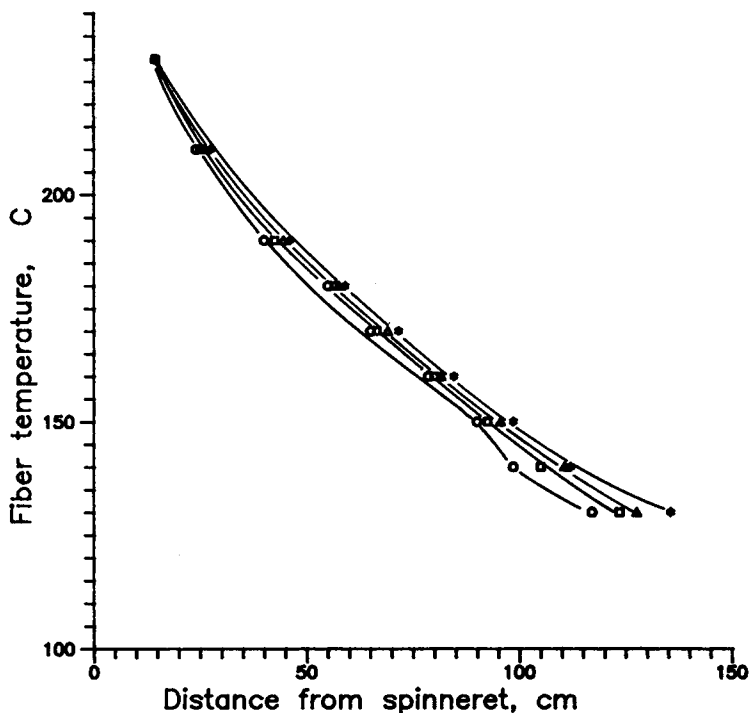


Fig. 4. Temperature profile along the spinline for high molecular weight PBT with a mass throughput of 4 g/min: (*) 1500 m/min; (Δ) 2500 m/min; (\square) 3500 m/min; (\circ) 4500 m/min.

If crystallization is occurring in the necking zone, we might expect to be able to detect a small temperature plateau at this position due to the sudden release of the latent heat of crystallization. Lu and Spruiell⁸ have shown that such a plateau is observable in the temperature profiles measured during high speed spinning of polypropylene. However, no sudden necking phenomenon was observed in the case of polypropylene. In the present case for PBT the occurrence of the sudden necking and the accompanying increased heat transfer apparently overrides the influence of the latent heat of crystallization on the temperature profile. The fact that the position of the neck is not completely stable also makes it unlikely that a temperature plateau could be observed for PBT even if it occurs. The movement of the point of crystallization (necking) would tend to smooth out small perturbations in the experimentally measured temperature profile.

Although the filament spun at 3500 m/min exhibits diameter thinning, no evidence of a more rapid temperature decrease is seen because the necking zone is beyond the measurement range of temperature. In any case the lower drawdown ratio occurring in the neck would tend to reduce the effect of the neck in producing a sudden increased cooling rate. Lower crystallinity development in the spinning fiber would also make observation of a temperature plateau more difficult. The same argument would hold for the lower spinning speeds.

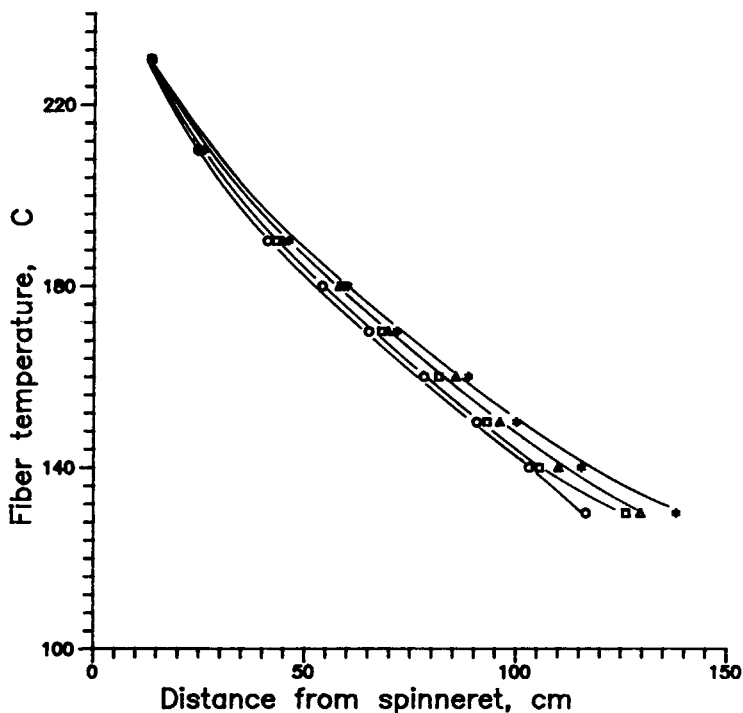


Fig. 5. Temperature profile along the spinline for low molecular weight PBT with a mass throughput of 4 g/min: (*) 1500 m/min; (Δ) 2500 m/min; (\square) 3500 m/min; (\circ) 4500 m/min.

Birefringence Profiles

Birefringence is a measure of molecular orientation developed in the filament. For the final filament it is related to tenacity, modulus, and storage stability of the filaments. On-line birefringence measurements give an indication of the location in the spinline at which the filament structure is developed.

Birefringence profiles of filaments prepared from polymer of either molecular weight are presented in Figures 6 and 7. Birefringence is generally higher for higher take-up velocities than for the lower ones at the same positions along the spinline. As can be seen from these figures, birefringence increases abruptly as the filament necks down for the higher spinning velocities. At lower take-up velocities, it increases gradually within the spinline distance range where measurements could be made.

For the higher molecular weight polymer, a rapid rise in birefringence was observed about 90 cm from the spinneret when take-up velocity is 4500 m/min, and at 120 cm from the spinneret when take-up velocity is 3500 m/min. Since the birefringence is the optical retardation divided by the diameter of the filament, any increase of retardation itself or any diameter decrease will directly affect the birefringence values. The position of the sharp increase of birefringence corresponds to the diameter thinning. An increase of retardation also occurred in the neck; however, it was not as obvious as

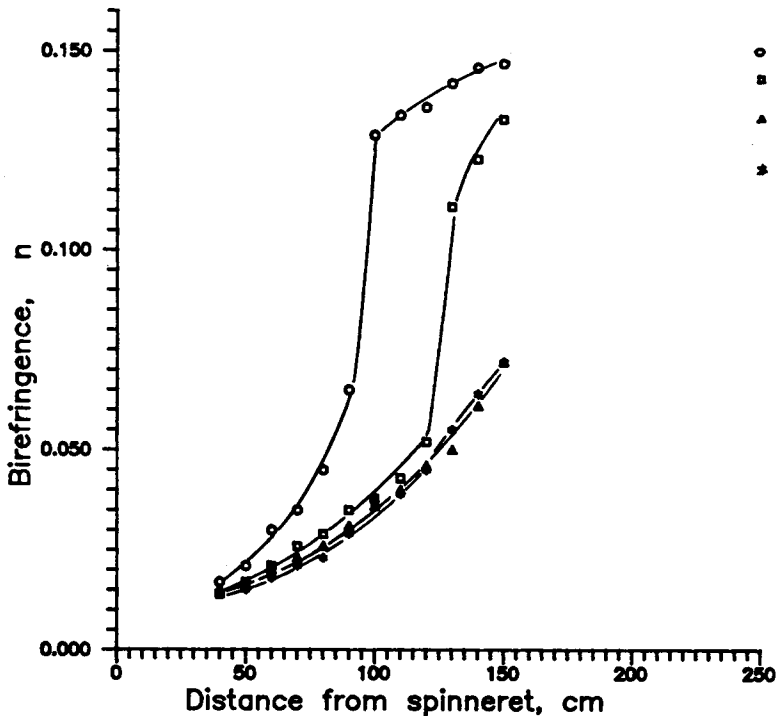


Fig. 6. Birefringence profile along the spinline for high molecular weight PBT with a mass throughput of 4 g/min: (*) 1500 m/min; (Δ) 2500 m/min; (\square) 3500 m/min; (\circ) 4500 m/min.

diameter thinning. Thus birefringence increases abruptly primarily as a result of diameter thinning.

The effect of molecular weight on birefringence along the spinline is quite interesting. At distance less than 100 cm from the spinneret (prior to the necking zone), the birefringence is higher for the higher molecular weight polymer than for the lower molecular weight one, at least for the higher take-up velocity. After the necking zone, the situation reverses; i.e., the birefringence is higher for lower molecular weight polymer than for the higher molecular weight one. The latter results are coincident with the results for the final birefringence of as-spun filament.¹ Similar results were also found in the case of PET.⁶

A reasonable interpretation of the results shown in Figures 6 and 7 is that the birefringence prior to necking represents the molecular orientation developed in the amorphous melt. This should be proportional to the rheological stress developed at any point along the spinline in accord with the rheoptical law¹⁰⁻¹²:

$$\Delta n = C(\sigma_1 - \sigma_2) = C\sigma \quad (1)$$

Here C is the stress-optical coefficient, and σ is the spinline stress. Higher molecular weight should produce higher melt viscosity and higher spinline stress. This, in turn, should produce a higher birefringence in the spinline for the higher molecular weight polymer.

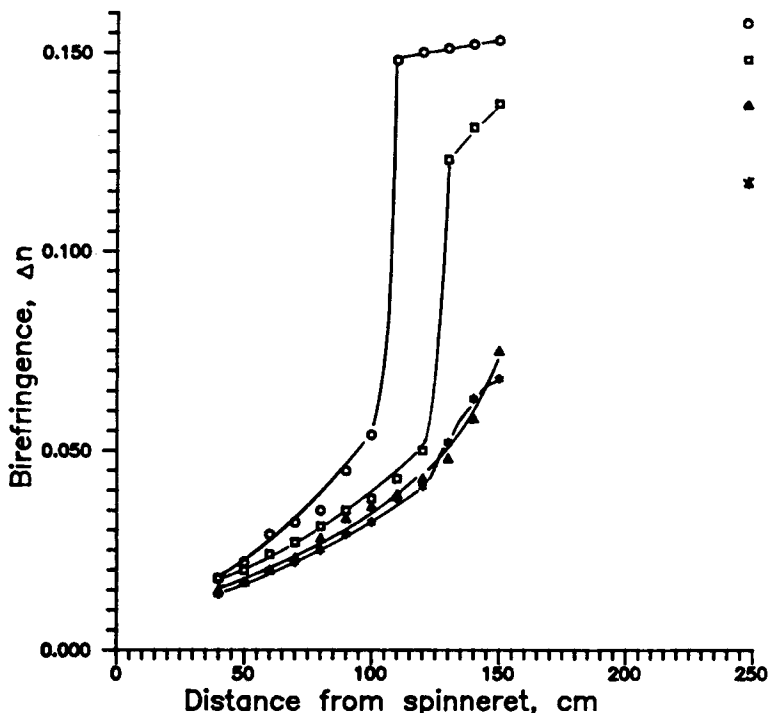


Fig. 7. Birefringence profile along the spinline for low molecular weight PBT with a mass throughput of 4 g/min: (*) 1500 m/min; (Δ) 2500 m/min; (\square) 3500 m/min; (\circ) 4500 m/min.

The abrupt increase of birefringence in the neck may be associated with a sudden increase of molecular orientation caused by nucleation and growth of highly oriented crystals. Although the spinline stress may be a gradually increasing function, the birefringence need not be due to the formation of oriented crystals that will cause a marked deviation from the rheoptical law. Finally, the lower birefringence in the region beyond the neck for the higher molecular weight sample appears to be related to a lower crystallinity developed in the higher molecular weight filaments at any given take-up velocity. This difference was documented in our earlier paper.¹

Take-Up Stress

The take-up stress is the force per unit area of the filament measured at a position just above the drawdown device. Thus the take-up stress is F_{ext}/A , where F_{ext} is external force applied by the drawdown device. The value of the take-up stress as a function of take-up velocity and polymer molecular weight is shown in Figure 8. The take-up stress increases rapidly with the take-up velocity and it also is higher for the higher molecular weight polymer at a specified take-up velocity. This is expected, as already mentioned, because an increase of molecular weight should lead to a higher elongational viscosity and require a greater stress to draw the filament down. These results are consistent with the interpretation given above for the development of birefringence in the spinline.

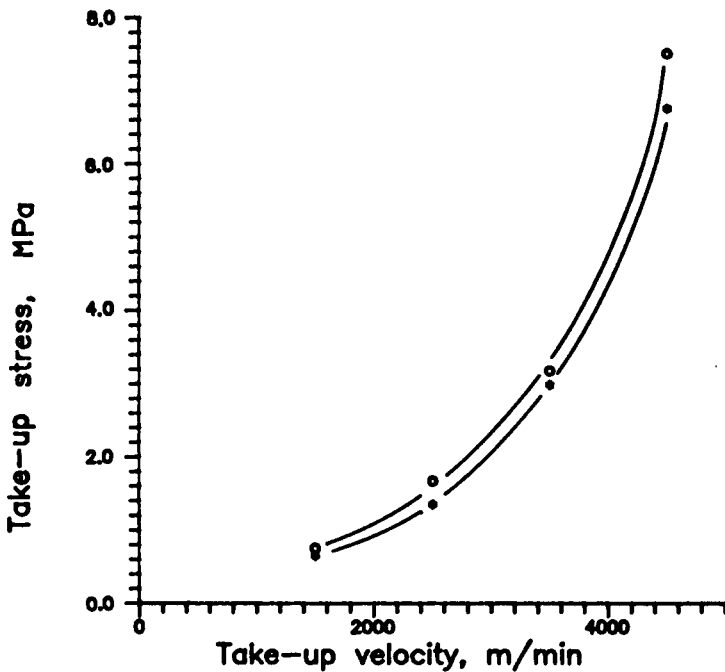


Fig. 8. Take-up stress as a function of take-up velocities for different molecular weight PBT: (*) IV = 0.75, $Q = 4$ g/min; (O) IV = 1.0, $Q = 4$ g/min.

Several investigators^{2,3,4,13,14} have argued that the rheological stress in the spinline at the point of crystallization is a fundamental quantity that controls the development of structure. In order to determine the rheological stress in the spinline, we must correct the take-up stress for contributions that occur beyond the point of crystallization.

The take-up stress depends on the molecular weight of the polymer and also on the spinning conditions such as spinning temperature, cooling rate, take-up velocity, and spinline length. A force balance for the forces acting in the spinline shows that

$$F_{\text{ext}} = F_{\text{rheo}} + F_{\text{inert}} + F_{\text{drag}} - F_{\text{grav}} \quad (2)$$

where F_{inert} is the inertial force due to acceleration of the mass in the spinline, F_{drag} is force due to air drag acting on the rapidly moving fiber, F_{grav} is the force due to gravity, and F_{rheo} is the rheological force tending to deform the polymer. For high speed spinning the contribution due to air drag is quite high particularly if spinline length is large. The contribution due to inertia can also be large and tends to build-up in the part of the spinline where the structure is being developed.

Shimizu et al.¹⁵ have suggested that the inertial contribution is the primary contributor that controls the structure development. In considering the data shown in Figure 8, it must be remembered that it includes a contribution due

to air drag that has little to do with structure development and which is rapidly increasing with take-up velocity.

Further Discussion of Results

The occurrence of a neck and structure change in the spinline are complex phenomena. Perez and Lecluse¹⁶ first reported that a necking zone was observed at a distance of 100 cm from the spinneret in PET high speed spinning. They assumed that the major part of the orientation and crystallization initiated in the zone. Matsui¹³ also reported that necking occurred in the spinline during high speed spinning of PET. He found that when filament birefringence reached 0.02–0.03, a rapid orientation-induced crystallization took place, and heat was generated by crystallization in this region. Both Perez and Lecluse¹⁶ and Bai and Spruiell⁷ have made X-ray diffraction patterns on each side of the necking zone for samples that have been cut from the running spinline. These patterns show that PET is amorphous prior to the neck and semicrystalline beyond it. Bai also observed that birefringence rises abruptly at the neck in samples cut from the spinline.

So far, none of the papers dealing with PP high speed spinning have reported necking in the spinline, although crystallization in the threadline is well established.^{3,4,8} To date, there seem to be no reports of necking in the absence of spinline crystallization. These facts suggest that crystallization is a necessary but not sufficient condition to produce necking in the spinline.

Shimizu et al.¹⁷ have reported that necking in PET is associated with the formation of a mesophase just prior to crystallization. Mathematical modeling of the spinline carried out by Bai and Spruiell⁷ for PET indicates that some form of strain softening is needed in order to introduce necking into the usual mathematical modeling approaches. It was suggested that a slight temperature rise at a point in the threadline caused by the rapid release of the heat of crystallization resulting from orientation-induced crystallization could provide the strain softening mechanism needed to induce the neck. This mechanism also explained the lack of an appreciable neck in branched PET samples, and in samples that develop low levels of crystallinity. The present results for PBT are also consistent with this explanation. When crystallization occurs during spinning under conditions that do not result in local heating of the spinline and consequent strain softening, no necking is expected. This presumably explains the lack of previous observation of necking in polypropylene, but does not rule out the possibility that it will be observed under other spinning conditions in the future.

In the case of PBT melt spinning, α -form crystals were found in the as-spun filaments made at take-up velocities of 1500–4500 m/min (see Fig. 2), but a small quantity of β -form was also detected in the samples made at the highest take-up velocity.¹

The primary difference between the α -form and β -form is that the crystal has a more extended conformation of the polymer chain in the β -crystal form. Previous studies^{18–20} have shown that α -form may be transformed to β -form by strains exceeding 4% elongation. In the range of strains between 4 and 14% the amount of β -form increases with strain, but the process is reversible upon

release of the stress. Higher strains result in the retention of some β -form unless the sample is annealed at sufficiently high temperature.²⁰ Deeg²¹ reported that the β -form can also be found in melt-spun filaments prepared from blends of PBT and PET. These observations, together with those of the present investigation, suggest that the β -form may form in the spinline by stress-induced crystallization aided by the high local strains occurring in the necking zone. Release of the stresses may permit transformation of the bulk of the β -form to the α -form, resulting in only a small amount of the β -form retained in the samples.

SUMMARY AND CONCLUSIONS

1. Spinline diameter measurements showed a phenomenon of diameter thinning (necking) at take-up velocities of 3500 and 4500 m/min for either of two PBT polymers. The location of the necking zone was affected by polymer molecular weight and spinning conditions. It moved closer to the spinneret for higher spinning speeds and for the higher molecular weight polymer.

2. Temperature profiles indicated that temperature decreased gradually along the spinline; however, the rate of temperature decrease at a take-up velocity of 4500 m/min was slightly increased in the range just beyond the necking zone. It was suggested that this occurred because necking increased the surface area per unit volume and the velocity of running filaments. The temperature of the spinline at the position corresponding to the necking zone was higher for the higher molecular weight polymer than for the lower molecular weight one.

3. Birefringence profiles exhibited an abrupt increase in birefringence in the necking zone for take-up velocities of 3500 and 4500 m/min due to both an increase of retardation and a sharp decrease of diameter. This was interpreted to result from oriented crystal formation as a consequence of stress-induced crystallization.

4. It was suggested that stress-induced crystallization is a cause of necking. The release of heat of crystallization provides sufficient "strain-softening" locally to induce the rapid drawdown associated with formation. Stress-induced crystallization is seen as a necessary but not sufficient condition to produce necking in the spinline, as some polymers crystallize in the spinline without the formation of necks in the diameter profiles.

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