Studies on High-Speed Melt Spinning of Noncircular Cross-Section Fibers. I. Structural Analysis of As-Spun Fibers

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ABSTRACT: Flat fibers and hollow fibers were prepared through the high-speed melt spinning of poly(ethylene terephthalate) (PET), and the structures of these fibers were compared with those of circular fibers. The cross-sectional shape of each fiber changed to a dull shape in comparison with that of the respective spinning nozzle. The change in the cross-sectional shape was slightly suppressed with an increase in the take-up velocity. There was a significant development of structural variation in the cross section of flat fibers in that the molecular orientation and crystallization were enhanced at the edge. Despite the difference in the cross-sectional shape, the structural development of flat, hollow, and circular fibers with increasing take-up velocity showed almost similar behavior. Considering that the tensile stress at the solidification point of the spin line is known to govern the structure development of high-speed spun PET fibers, it was speculated that the effects of the enhancement of cooling and air friction on the tensile stress at the solidification point cancel each other. © 2001 John Wiley & Sons, Inc. J Appl Polym Sci 80: 1575–1581, 2001

Key words: poly(ethylene terephthalate); melt spinning; noncircular fiber; birefringence; crystallization

INTRODUCTION

Development of melt-spun fibers with noncircular cross sections started in the 1960s. The first attempt was to mimic the gloss of expensive silk fibers by changing the cross section to a triangular shape. Since then various types of noncircular fibers have been developed to add functionality and aesthetics to synthetic fibers.[1,2] Flat fiber is the most simple type of noncircular fiber. It shows higher drapability because of reduced bending stiffness. It also has larger surface area than circular fibers of the same thickness (i.e., denier or linear density). Hollow fiber is another example of simple noncircular fiber that are widely used as membranes for artificial kidneys, water-cleaning systems, and so forth. The lighter weight and bulkiness of hollow fibers are also utilized for fabrication of cushions and garments.

In ordinary melt spinning processes, fibers with circular cross section are produced by the extrusion of a polymer melt from a circular nozzle. Ohwaki et al. reported the formation of complex wavy crimping after the drawing and annealing of rectangular cross section fibers that were melt spun using a slit nozzle with an aspect ratio of 80.[3] It was explained that the wavy crimp originated from the variation of heat shrinkage in the fiber cross section; however, the mechanism of the formation of structural variation in the fiber forming processes was not investigated.

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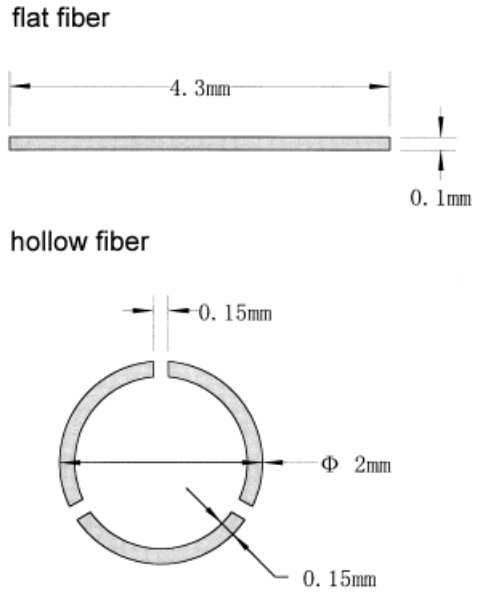

Figure 1 The cross-sectional shapes of the spinning nozzle.

On the other hand, high-speed melt spinning is an innovative fiber forming process in which fibers with highly developed structure can be obtained.[4–6] In the process, molecular orientation and orientation-induced crystallization occurs because high elongational stress can be applied to the molten polymer in the spin line. Accordingly, subsequent drawing and annealing processes for the control of the fiber structure can be eliminated and fibers directly applicable for the end use can be obtained. This leads to the establishment of a fiber forming process with high productivity.

In this study, flat fibers and hollow fibers were prepared by the high-speed melt spinning of poly(ethylene terephthalte) (PET) and the effect of the change in cross-sectional shape on the structure and properties of as-spun fibers was investigated. If the cross-sectional shape of the nozzle is varied, the spinning behavior may change significantly because heat transfer and air-friction effects for the noncircular cross section fibers are not the same as those for circular fibers. This effect may lead to the alteration of the structure and properties of as-spun fibers.

EXPERIMENTAL

Melt Spinning

The polymer material used for the high-speed melt spinning was PET chips kindly supplied by Teijin Ltd. The inherent viscosity of the polymer was 0.62 dL/g, and it contained 0.4 wt % titanium oxide. The extrusion system consisted of an extruder and a gear pump. Two types of spinnerets were prepared. One was for the flat fibers and the other was for the hollow fibers. The cross-sectional shapes of the nozzle hole in the spinneret are shown in Figure 1. A slit nozzle with a width and gap of 4.3 and 0.1 mm, respectively, was used for the preparation of flat fibers. The spinneret for the hollow fibers had three arc slits. The polymer melts were extruded from these slits and connected to each other immediately below the spinneret to form a tubular shape. For comparison, high-speed melt spinning of circular fibers was also performed using a 0.5-mm diameter circular nozzle. The spinning conditions are summarized in Table I.

Observation of Cross-Sectional Shape

As-spun fibers were impregnated in epoxy resin and cut perpendicular to the fiber axis. The cut surface was polished and subjected to observation under a reflection-type optical microscope.

Simultaneous Analyses of Shape and Structure

An interference microscope (Carl Zeiss Jena) was used for the analyses of the shape and refractive index of the as-spun fibers. Typical interference fringe patterns of flat and hollow fibers are shown in Figure 2. From the M-shaped fringe pattern for the flat fiber we found that there was a distinct distribution of the optical pathlength (= refractive index × thickness) along the transverse axis of the fiber cross section.

For the simultaneous analyses of the shape and structure of flat fibers, two interference fringe patterns were obtained for the same sam-

Table I Spinning Conditions

Polymer	Poly(ethylene terephthalate) $[\eta]$ = 0.62 dL/g, 0.4 wt % TiO_2	
Spinning temperature	290°C	
Throughput rate	5 g/min/hole	
Take-up velocity	Flat fiber	1–5 km/min
	Hollow fiber	1–7 km/min
	Circular fiber	1–6 km/min

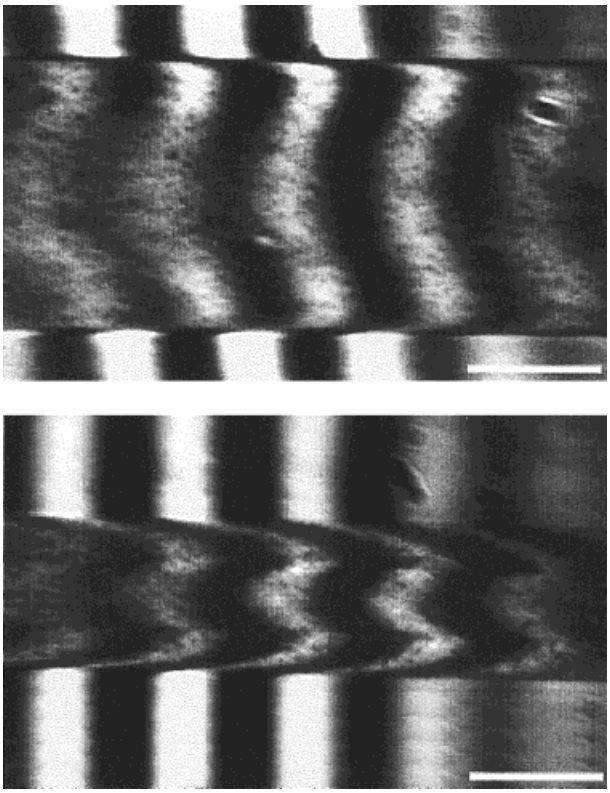

Figure 2 Typical interference fringe patterns for (a) a flat fiber (3 km/min take-up velocity) and (b) a hollow fiber (2 km/min take-up velocity) under an interference microscope. Scale bar = 50 μm.

ple using two immersion liquids of different refractive indices N_1 and N_2. The distributions of the refractive index and thickness along the transverse axis of the fiber cross section $n(x)$ and $y(x)$ were analyzed using the following equation[7]:

$$\frac{d_i(x)}{D_{Fi}}\lambda = [n(x) - N_i]y(x) \tag{1}$$

where x is the coordinate along the transverse axis, D_F is the spacing of the fringes, $d(x)$ is the fringe shift, and λ is the wavelength of the incident light. The suffix i (=1, 2) denotes the values for two different immersion liquids. The $n(x)$ and $y(x)$ were obtained by solving eq. (1) for $i = 1$ and 2 simultaneously.

The refractive index n of the hollow fibers can be calculated using the following equation if the outside and hollow part of the fiber is filled with the immersion liquid of refractive index N:

$$(n - N)(D_o - D_i) = \frac{d}{D_F}\lambda \tag{2}$$

where D_o and D_i are the respective outer and inner diameters of the hollow fiber.

Measurements for the flat and hollow fibers were performed using a polarizing filter and the refractive indices parallel ($n_\parallel$) and perpendicular ($n_\perp$) to the fiber axis were analyzed. The birefringence Δn was calculated as the difference of these two refractive indices, $\Delta n = n_\parallel - n_\perp$.

Assuming that any local part of the flat and hollow fiber has an axial symmetric structure with respect to the fiber axis, the Lorentz density LD, which has a linear relation with density and therefore the crystallinity, was also obtained according to the Lorentz–Lorenz equation given below.[8]

$$\mathrm{LD} = \frac{n^2 - 1}{n^2 + 2} = \frac{4\pi N_A P}{3M}\rho \tag{3}$$

where N_A is the Avogadro number, P is the molar polarizability, M is the molecular weight of the monomer unit, and ρ is the density. The mean refractive index $\bar{n}$ was calculated using the following equation:

$$\bar{n}^2 = \frac{n_\parallel^2 + 2n_\perp^2}{3} \tag{4}$$

RESULTS AND DISCUSSION

Cross-Sectional Shape

The cross-sectional shapes of flat and hollow fibers prepared at a take-up velocity of 1 km/min are shown in Figure 3. The flat fibers basically have a rectangular cross section with slightly rounded edges. The hollow fibers showed a cross section of concentric circles. The aspect ratio of the flat fiber (= width/thickness) was about 10, which was significantly smaller than that of the slit nozzle (43). The hollow fiber had a hollow ratio [$=(D_i/D_o)^2$] of about 0.2, which was also significantly lower than that of the nozzle (0.73). In other words, the cross-sectional configuration of the fibers changed to a rather dull shape in the spin line.

Cross-Sectional Shape and Structure Distribution

The cross-sectional shape of the flat fibers analyzed using the interference microscope is shown in Figure 4. Except for the slight roundness at the edge and slight concaveness at the center, the

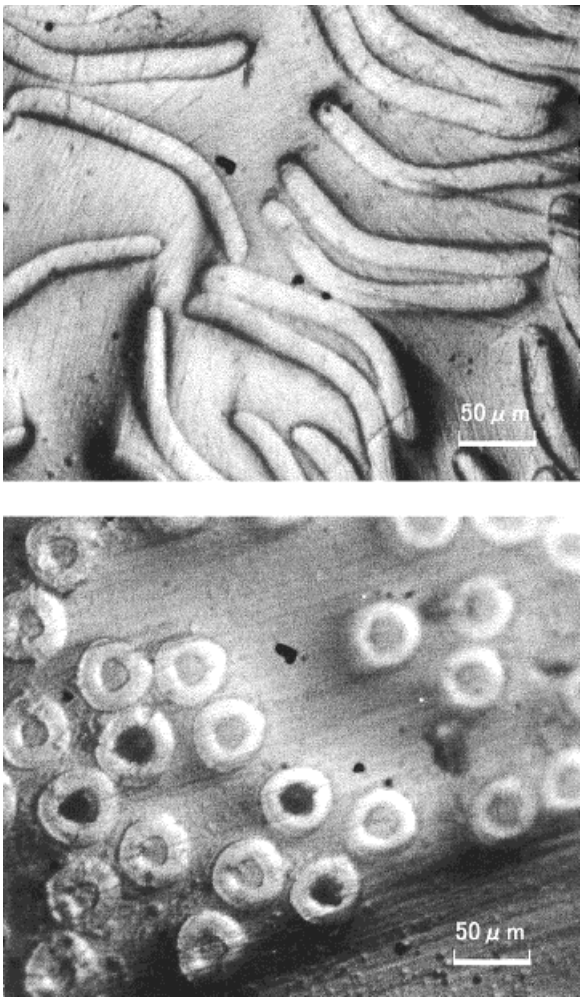

Figure 3 Typical optical micrographs of cross sections of flat and hollow fibers (1 km/min take-up velocity).

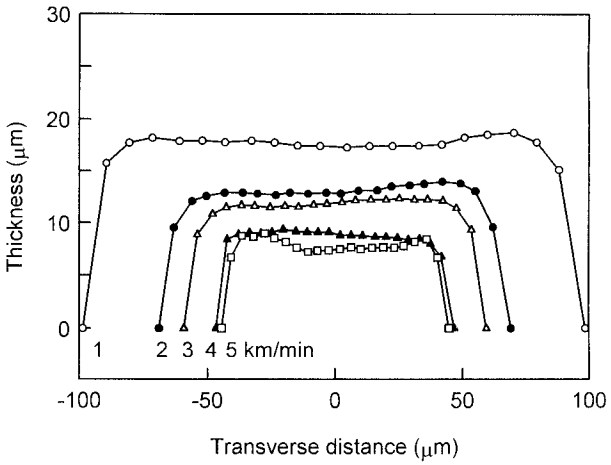

Figure 4 The cross-sectional shape of the flat fibers analyzed using an interference microscope.

overall cross-sectional shape was rectangular. The cross-sectional area decreased with increasing take-up velocity because the melt spinning was performed with a constant throughput rate. On the other hand, the cross-sectional shape for all the samples appeared to be "similar," irrespective of the take-up velocity and cross-sectional area.

The cross-sectional area of the flat and hollow fibers was calculated based on the results of the interference microscope measurements, and it is plotted against the take-up velocity in Figure 5. The solid line in the figure indicates the theoretical estimate calculated using the equation of continuity.[9] There was fair agreement between the experimental results and theoretical predictions.

The dependences of the aspect ratio of flat fibers and the hollow ratio of hollow fibers on the take-up velocity are shown in Figure 6. Both parameters showed a slight increase with an increase in the take-up velocity; however, the increment was small in comparison with the significant changes of the cross-sectional shape from that of each nozzle.

The distributions of birefringence and Lorentz density along the transverse axis of the flat fibers are shown in Figures 7 and 8, respectively. The birefringence increased with an increase in the take-up velocity. At the same time, there was a significant development of birefringence distribution in which birefringence at the edge was higher than that at the center. The birefringence difference between the edge and center increased with

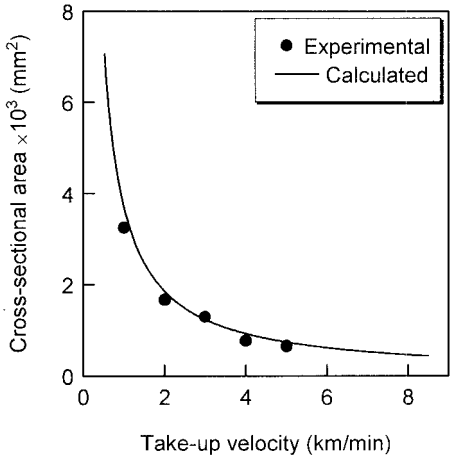

Figure 5 The experimental and calculated cross-sectional area of flat fibers plotted against the take-up velocity.

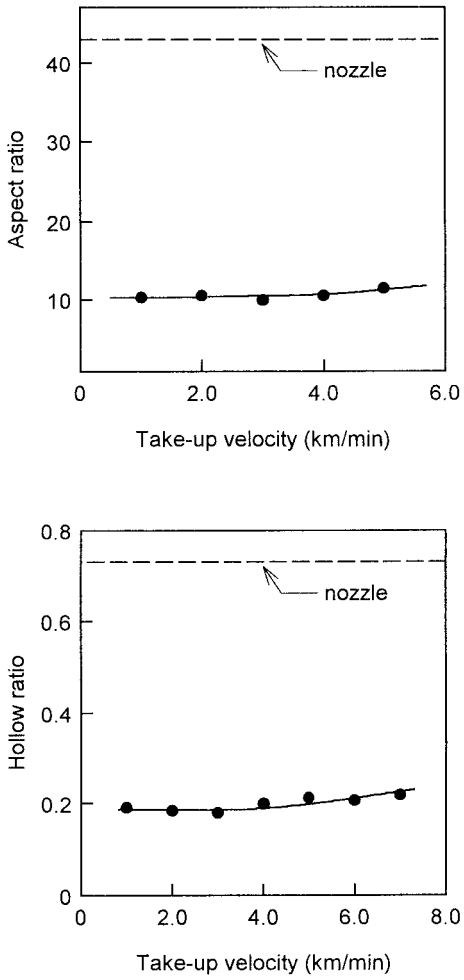

Figure 6 The aspect ratio and hollow ratio plotted against the take-up velocity.

the take-up velocity up to 4 km/min. The 5 km/min fibers showed a milder birefringence distribution than the 4 km/min fibers.

The Lorentz density was almost constant up to 3 km/min, indicating that the as-spun fibers obtained in this take-up velocity region had an amorphous structure. At 4 km/min there was a slight increase of the Lorentz density at the edge, indicating that the orientation-induced crystallization started to occur only in the edge part. The Lorentz density of the 5 km/min fiber increased in the whole cross section; however, the edge part still showed higher Lorentz density than the center part. This result also implied that in the 5 km/min fibers the crystallinity at the edge was higher than that at the center.

It was reported that the high-speed spun circular fibers of PET show significant structural variation in their cross section only when the take-up velocity exceeds 7 km/min.[7] The reason for the development of this structural variation was attributed to the temperature distribution in the cross section of the spin line. On the other hand, as mentioned in the introduction, Ohwaki et al. reported that the flat fibers produced through the conventional process (i.e., the drawing and annealing of fibers prepared by the low-speed spinning process) also show significant structural distribution.[3]

Therefore, it can be considered that in the spin line of flat fibers there is the development of a substantial temperature distribution in the cross section because of enhanced cooling of the edge part. The temperature distribution may lead to the distribution of elongational stress and thereby the distributions of molecular orientation and orientation-induced crystallization in the cross section of flat fibers. It is interesting to note that the birefringence distribution was most significant in the 4 km/min fibers in that only the edge part was crystallized and the center part remained in an amorphous state.

The interference fringe pattern of as-spun hollow fibers did not show any indication of the development of structural distribution.

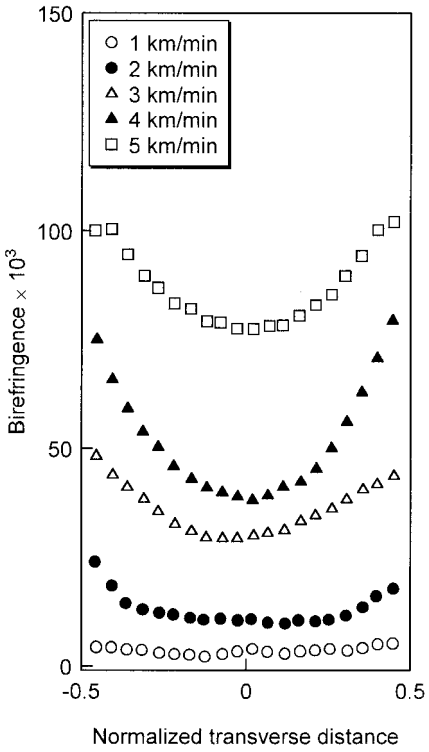

Figure 7 The birefringence distribution along the transverse axis of flat fibers prepared at various take-up velocities.

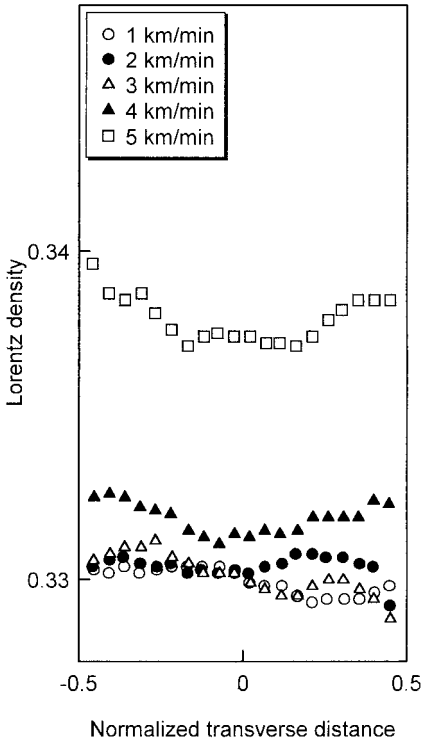

Figure 8 The Lorentz density distribution along the transverse axis of flat fibers prepared at various take-up velocities.

Comparison of Structure Development

The development of the birefringence and Lorentz density of flat, hollow, and circular fibers with an increase in the take-up velocity are compared in Figures 9 and 10. In the flat fibers the birefringence and Lorentz density were averaged along the transverse axis. Surprisingly, the birefringence of the three types of fibers increased with take-up velocity in almost an identical manner. The data for the Lorentz density scattered more because it was calculated from the mean refractive index and had lower accuracy. In general, the Lorentz density also showed a similar behavior for all three types of fibers; however, the orientation-induced crystallization of the flat fibers appeared to start at a slightly lower take-up velocity. The concentration of the tensile stress at the edge part of the fiber cross section that was caused by the development of the temperature distribution may be attributable to this behavior.

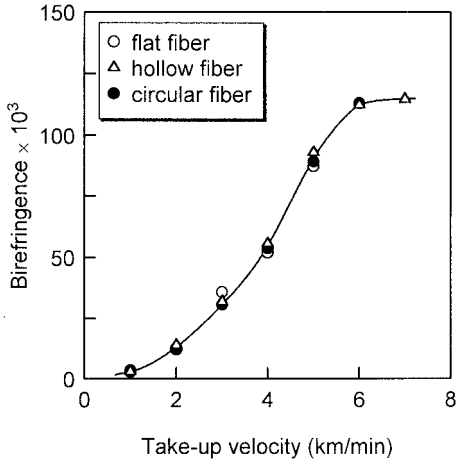

Figure 9 The dependence of the birefringence development on the take-up velocity for three types of fibers.

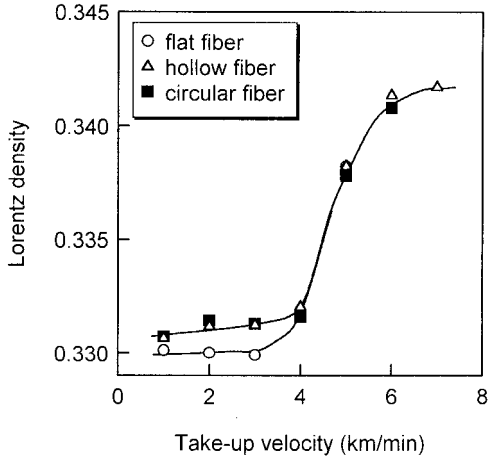

Figure 10 The dependence of the Lorentz density development on the take-up velocity for three types of fibers.

The fiber structure development in the melt spinning process of PET strongly depends on the temperature and stress histories applied to the material in the spin line. More specifically, the molecular orientation of as-spun fibers is mainly controlled by the stress applied at the solidification point.[10] It also should be noted that the stress at the solidification position is dominated by the contributions of inertia and air-friction forces if the take-up velocity is sufficiently high.[11]

The change in the cross-sectional shape of the spin line should lead to significant changes in the cooling behavior and air-friction force acting on the fiber surface. In the flat fibers the increased surface area should result in enhanced cooling. The air-friction force also should be larger because of the larger surface area. These considerations are totally inconsistent with the similarity in the structure development of the three types of as-spun fibers. There can be two possible reasons

for this behavior. One is that the effect of inertia force, which is independent of the cross-sectional shape, is dominant for the stress applied to the spin line at the solidification point. Another possible reason is that the effects of enhanced cooling and air friction cancel each other and therefore the air-friction force at the solidification point is not changed much by the change of the cross-sectional shape. If the enlargement of the surface area in the noncircular fiber is considered, the latter explanation may be more conceivable. In other words, a larger surface area can increase the air-friction force; however, it also enhances the cooling and shortens the distance from the spinneret to the solidification point, which leads to the reduction of the air-friction force.

In comparison with the flat fibers, the spin-line dynamics of hollow fibers is not easy to speculate because the effect of air captured inside the fiber is not clear. However, the experimental results for the three types of fibers seems to suggest that, in general, the change in the cross-sectional shape does not have much of an affect on the structure development in the high-speed melt spinning process.

CONCLUSION

The cross-sectional shape of flat and hollow fibers changed to a dull shape in comparison with that of the spinning nozzle. The change in the cross-sectional shape was suppressed slightly with an increase in the take-up velocity; however, this change was much less significant in comparison with the change from the spinning nozzle. Simultaneous measurement of the cross-sectional shape and refractive index of as-spun fibers using an interference microscope was found to be possible. In the cross section of flat fibers the molecular orientation and orientation-induced crystallization were enhanced at the edge part. The change in the cross-sectional shape did not have much affect on the structure development of as-spun fibers. We speculated that the larger surface area increased the air-friction force, enhanced the cooling effect, and shortened the distance from the spinneret to the solidification point, which led to the reduction of the air-friction force. Accordingly, the change in cross-sectional shape had little affect on the structure development in the high-speed melt spinning process in general.

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