博士論文の内容の要旨

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論文題目	Mechanical properties of polypropylene fiber – Effects of primary structure parameters and additive on the tensile and knot-pull strengths – (ポリプロピレン繊維の力学物性 – 一次構造と添加剤が引張強度 と結節強度に与える影響 –)

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Isotactic polypropylene (iPP) has excellent characteristics, and it is used in molded products and films, as well as in fibers. The mechanical properties, such as the tensile strength, are important in fibers. Higher-tensile-strength iPP fibers have been obtained with a narrow molecular-weight distribution iPP. Metallocene catalyst can synthesize narrow-molecular-weight-distribution iPP, but it tends to decrease the stereoregularity. Recently, a high stereoregularity iPP has been synthesized by using metallocene catalysts. This iPP is expected to increase the fiber tensile strength. High-strength fibers can be fabricated by using a high-molecular-weight polymer. The molecular weight is reduced by thermal degradation during melt spinning, but the narrow-molecular-weight-distribution iPP can suppress the decrease in molecular weight. Therefore, high-molecular-weight metallocene catalyst-synthesized iPP can be expected to increase the tensile strength. The knot-pull strength, which is the tensile breaking stress of a knotted fiber, is also an important fiber property. Despite the recent increase in iPP fiber tensile strength, the knot-pull strength has barely increased. The sluggish increase in knot-pull strength may be attributed to complex knotted fiber breakage. Tensile, radial compressional, twisting, and bending forces have been proposed to cause knot breakage, but the breakage mechanism has not been elucidated sufficiently. The effects of primary structure and additive on the tensile and knot-pull strengths were analyzed in this study by investigating the attainable maximum tensile strength of the high stereoregularity metallocene catalyst-synthesized iPP and the knot deformation behavior during a knot-pull test.

In chapter 2, the effects of the molecular weight distribution and stereoregularity of iPP on the structure and properties of its fibers were also analyzed using 20 g/10 min melt index iPPs. A fiber having a high tensile strength of 1.36 N/tex (1.2 GPa) and initial modulus of 27.4 N/tex (25 GPa) was obtained from metallocene catalyst-synthesized iPP of high stereoregularity. These features were better than those of both low stereoregularity metallocene catalyst-synthesized iPP fiber and equivalent stereoregularity Ziegler–Natta catalyst-synthesized iPP fibers. The metallocene catalyst-synthesized iPP fibers also had a lower creep strain at 125 °C. The higher tensile strength of high stereoregularity iPP can be explained by the greater maximum draw ratio, crystallinity, and crystallite size of higher stereoregularity which related to the higher drawing temperature that is achievable because of the higher melting temperature. In addition, a lower degree of macroscale void development or suppressed fibrillation of metallocene catalyst-synthesized iPP fibers was also observed. However, there were almost no differences in the melting temperature, dynamic viscoelastic behavior, crystallite size, and crystallinity observed between the spun and drawn fibers produced by the iPP synthesized by different catalysts.

In chapter 3, the effect of molecular weight in addition to the molecular weight distribution was investigated using 3 and 4 g/10 min melt index metallocene and Ziegler-Natta catalyst-synthesized iPPs. Metallocene catalyst-synthesized iPP fibers with higher tensile strength, higher initial modulus, and lower creep strain at 125 °C than Ziegler–Natta catalyst-synthesized iPP fibers were obtained. Moreover, the obtained maximum tensile strength of 1.39 N/tex (1.3 GPa) was higher than the maximum tensile strength obtained by 20 g/10 min melt index iPP. The higher strength of metallocene catalyst-synthesized iPP fibers was also explained by the smaller crack diameters estimated by ultra-small angle X-ray scattering.

In chapter 4, the effects of the draw ratio and a melt-kneaded additive on the knot-pull strength of iPP monofilament, that is, a thick single fiber, were also discussed by comparing the tensile and loop strengths

of the fiber. The knot deformation behavior during a knot-pull test was also investigated. In contrast to the increase in tensile strength at a high draw ratio, the knot-pull strength was hardly affected by the additive, and the loop strength was reduced by the additive. During a knot-pull test on a high-draw-ratio fiber, the knot thickness continued to decrease to breakage, even after the knot length stopped decreasing. This behavior indicates that the highly drawn fiber breaks at the buckled part in the knot owing to the lateral compressional forces.

The study conclusions are provided in Chapter 5.