Mechanical Properties of Polypropylene Fibers Produced from the Binary Polymer Blends of Different Molecular Weights

DIANA GREGOR-SVETEC

Department of Textiles, Faculty of Natural Sciences and Engineering, University of Ljubljana, Snezniška 5, SI-1000 Ljubljana, Slovenia

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ABSTRACT: The mechanical properties of multifilament yarns, spun from the blends of a plastic-grade polymer with a fiber-grade CR-polymer in the composition range of 10–50 wt % added, were investigated. The predicted modulus of a two-phase blend, calculated from several representative equations, was compared with the elastic modulus of drawn yarns, determined from the stress vs. strain curve and dynamic modulus obtained from the sound velocity measurements. The best fit was achived with the Kleiner's simplex equation. For both the static and dynamic elastic modulus, the largest negative deviation is seen at the 80/20 and 60/40 plastic/fiber-grade polymer blend composition, while the largest positive deviation is seen at the 90/10 plastic/fiber-grade polymer blend composition, suggesting good compatibility of both polymers, when only a small percent of the fiber-grade CR-polymer is added. Improved spinnability and drawability of blended samples led to the yarns with the tensile strength over 8 cN/dtex, elastic modulus over 11 GPa and dynamic modulus over 15.5 GPa. Structural investigations have shown that the improved mechanical behavior of blended samples, compared to the yarn spun from the pure plasic-grade polymer, is the consequence of a higher degree of crystallinity, and above all, of a much higher orientation of macromolecules. © 2000 John Wiley & Sons, Inc. J Appl Polym Sci 75: 1211–1220, 2000

Key words: polypropylene fibers; polymer blends; mechanical properties; elastic modulus

INTRODUCTION

It is well known that with increasing the molecular weight of a polymer the mechanical properties of fibers are enhanced. High molecular weight polymers are, on the other hand, inconvenient for melt spinning. To overcome this drawback a number of modifications on processing techniques have been carried out. Some attempts to improve spinning performance by means of a polymer blending have also been studied. Hinrichsen and Green[1] studied the rheological behavior of the blends of two different molecular weights of nylon 6. They have found that processing of a high molecular weight component can be improved by blending it with a low molecular weight polymer, at specific ratios. Similarly, Bhateja and Andrews[2] observed an improved processing behavior of a ultrahigh molecular weight linear polyethylene when blended with a normal molecular weight linear polyethylene. They also found that the mechanical properties of the blended samples were intermediate between that of the parent samples. By blending polypropylene (PP) with the small amounts of polyethylene (PE), the processability and impact properties of PP are im-

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proved. Depending on the composition of the PP/PE blends, the mechanical properties can be improved also. A linear relationship between the modulus and the composition was observed for the blends of PP with high-density PE by some authors, while others reported a large positive deviation.[3]

The literature dealing with the blends of different molecular weights of PP is sparse. Most of it is dealing with the blends of a low molecular weight polymer with a small percent of a high molecular weight polymer added. This present work deals with the multifilament yarns produced from the blends of a high molecular weight PP with a low molecular weight PP added.

EXPERIMENTAL

Materials

The two kinds of commercial Hoechst polypropylene chips selected were Hostalen PPN 1060F, a plastic-grade homopolymer, with MFR = 2 g/10 min, and Hostalen PPU 1780F2 a fiber-grade homopolymer, with MFR = 18 g/10 min. The melt flow rate (MFR) was determined as the amount of polymer extruded at the temperature of 230°C, with the forcing load of 2160 g in 10 min. Hostalen PPN 1060F with = 280,000 g/mol is a broad molecular weight distribution polymer, with the ratio of weight-average to number-average molecular weight ($\bar{M}_w$) of 5, while Hostalen PPU 1780F2 with = 210,000 g/mol is a narrow molecular weight distribution polymer, so-called controlled rheology or CR-polymer, with $\bar{M}_w/\bar{M}_n$ = 3.3. Five blend samples were prepared by blending the plastic-grade polymer with 10, 20, 30, 40, and 50% of the fiber-grade CR-polymer by weight. From these polymer blends and from the pure plastic-grade polymer multifilament yarns were produced by melt spinning.

Preparation of Multifilament Yarns

The melt spinning and in-line drawing of PP multifilament yarns was carried out on an Extrusion Systems Ltd. laboratory spin-draw device. The filaments were extruded through a spinneret with 10 holes of a diameter 0.35 mm each. The spinning temperature in the range of 235–280°C and the mass outflow were adjusted so that a continuous steady spinning could be carried out. The cooling of the resultant filaments was achieved with a crossflow air quenching at the temperature of 6°C. The as-spun filaments were three-stage, moderately drawn at the temperature of 50°C in a continuous spin-drawing process. Continuously moderately drawn multifilament yarns were additionally drawn on a Zimmer laboratory draw device. In this subsequent slower stage, the yarns were drawn through a hot plate at the temperature of 145°C to the limiting draw ratio.

Tensile Testing

The tensile properties of the PP multifilament yarns were measured with an Instron 6022 tensile testing machine. The samples of a initial gauge length of 25 cm were stretched at a crosshead speed of 1.6 mm/s for discontinuously highly drawn yarns and of 5.5 mm/s for continuously moderately drawn yarns. The conventional stress–strain data were obtained under the conditions of the controlled temperature and humidity (temperature = 21°C, relative humidity = 65%). The mechanical data presented in this article are the average of 50 parallels and the stress–strain curves of about 20 parallels.

Structural Characterization

The wide-angle X-ray scattering (WAXS) technique was used to explore the structure of PP multifilament yarns. CuK_α radiation was monochromatized with the aid of a 10 μm Ni filter. The WAXS film patterns were taken on a vacuum flat-film camera with pinhole collimation. The WAXS curves (intensity as a function of scattering angle) were made for the normal transmission geometry of samples by using a two-circle goniometer developed by Kratky. The degree of crystallinity was estimated by the Hermans and Weidinger method[4] over the angular range from the scattering angle 2θ = 10 to 30°. The apparent crystallite dimensions were determined by means of the Scherrer's equation,[5] from the half-widths of diffraction curves of crystalline peaks, which were approximated by the pseudo-Voigt functions. The crystalline orientation function was calculated with the help of (110) and (040) reflections, using the Wilchinsky relation[6] for a monoclinic crystal system.

The crystalline fraction in fibers was evaluated also from the density data. The density of PP multifilament yarns was determined with the flotation method as described by Juilfs,[7] using a mixture of isopropylalcohol and water.

The birefringence of the filaments was measured using a Zeiss polarizing light microscope and an Ehringhaus compensator.

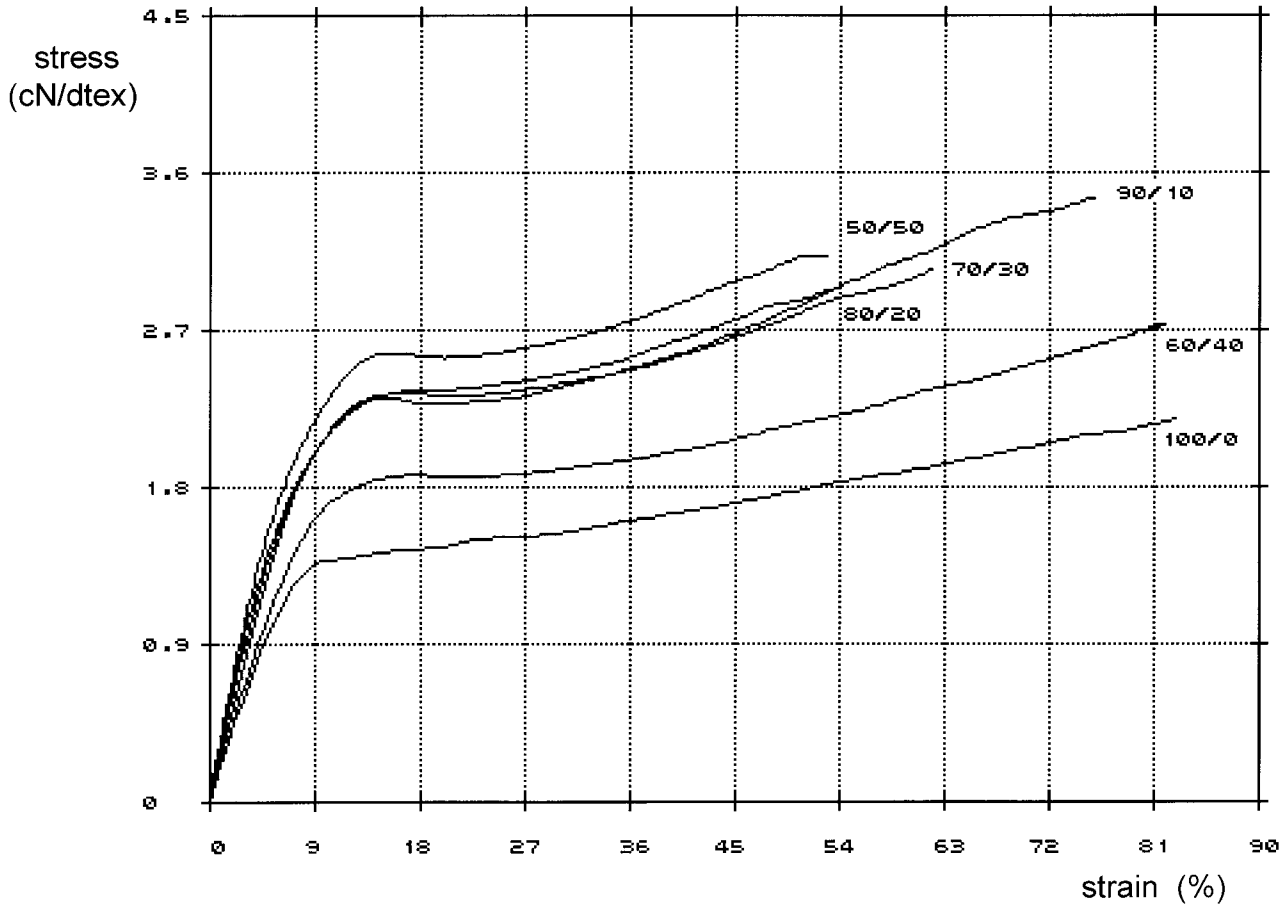


Figure 1 Stress vs. strain curves of continuously moderately drawn multifilament yarns.

The average molecular orientation and dynamic modulus were obtained by the sonic-velocity method. The velocity of sound waves in the filaments was measured on a Morgan's Dynamic Modulus Tester PPM-5R. The amorphous orientation function was calculated from the experimentaly determined dynamic modulus, and from the fraction of crystals and crystal orientation function determined by WAXS measurements in a manner applied to PP by Samuels.[8] The morphological studies involved scanning electron microscopy of the filament surface. A JEOL JSM-2 electron microscope was used for all morphological studies.

RESULTS AND DISCUSSION

Mechanical Properties

In Figure 1 the stress–strain curves of continuously moderately and in Figure 2 of discontinuously highly drawn multifilament yarns, spun from the pure plastic-grade polymer and from blends of this polymer with added fiber-grade CR-polymer in the composition range of 10–50 wt %, are shown. Mechanical properties of these yarns are presented in Table I.

Continuously moderately drawn multifilament yarns of low tensile strength and high toughness are easily extensible. The shape of the stress–strain curves (Fig. 1) shows after an initial linear portion a marked yield point, and up to the break, a region of low slope, where large extensions are produced by small increases in stress. Adding the fiber-grade CR-polymer to the plastic-grade polymer resulted in achieving higher stresses at the same percent of extension for yarns spun from the blends than for yarn spun from the pure plastic-grade polymer.

Discontinuously highly drawn multifilament yarns exhibit brittle mode of deformation behavior, with a short initial period of steep slope, no apparent yield point, and a regime of sharply

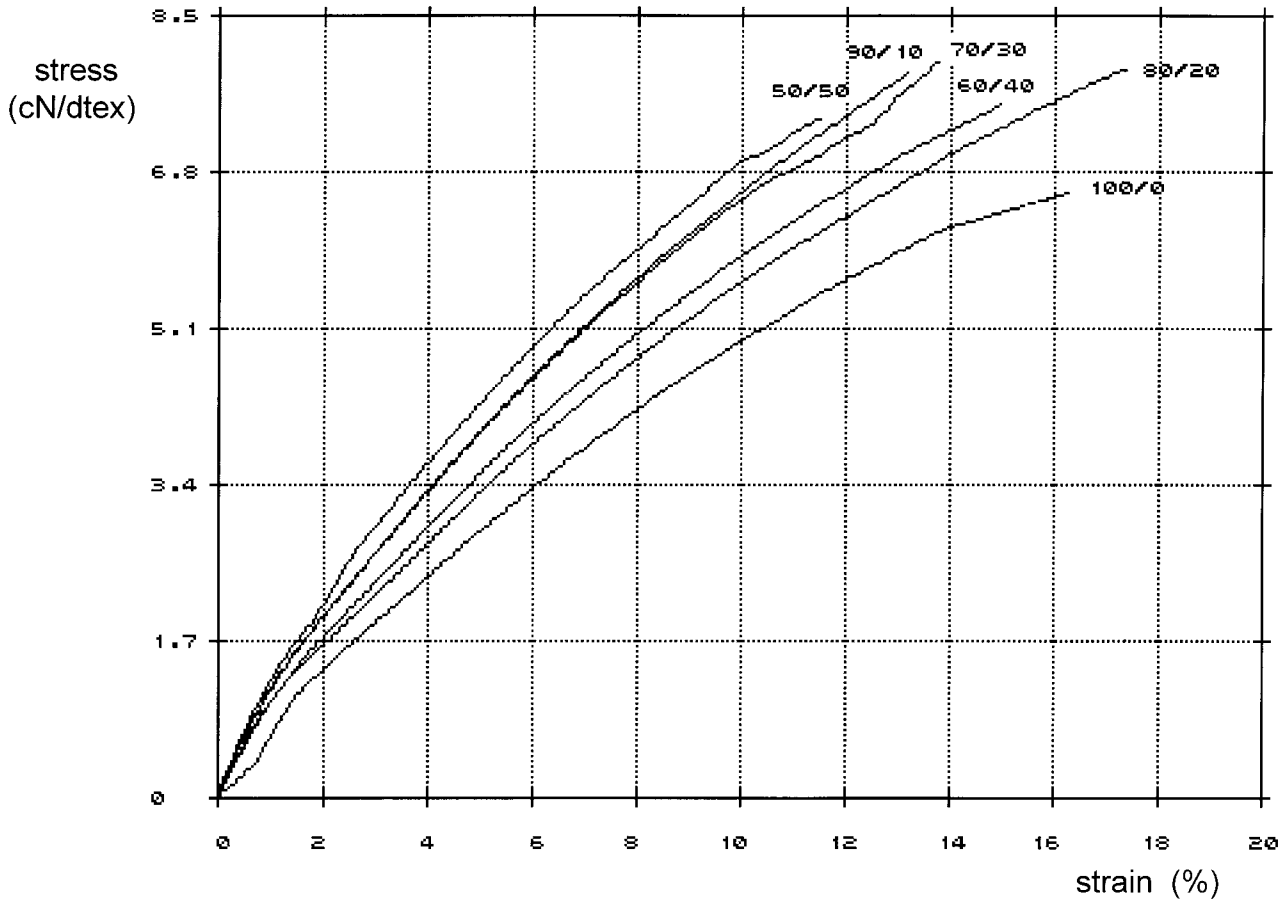


Figure 2 Stress vs. strain curves of discontinuously highly drawn multifilament yarns.

rising stress until fracturing occurred at the extensions between 12 and 18% (Fig. 2). Similar to continuously drawn multifilament yarns, the addition of the fiber-grade CR-polymer to the plastic-grade polymer resulted in achieving higher stresses at the same percent of extension for yarns spun from the blends. These yarns also show a steeper initial slope of the stress/strain curve, indicating larger initial resistance to the applied tensile force.

The addition of a small amount of the fiber-grade CR-polymer to the plastic-grade polymer resulted in improved spinnability and drawability of a given resin. Multifilament yarns spun from the blends could be drawn to the higher limiting draw ratio than yarns spun from the pure plastic-grade polymer. Discontinuously highly drawn multifilament yarn spun from the 90/10 plastic/fiber-grade polymer possesses the highest draw ratio among all samples and the best mechanical properties. Almost the same high limiting draw ratio and excellent mechanical properties achieved discontinuously highly drawn multifilament yarn spun from the 70/30 plastic/fiber-grade polymer. The tensile strength over 8 cN/dtex and elastic modulus over 11 GPa was obtained for both yarns.

The addition of 20 and 40% of the fiber-grade CR-polymer also resulted in improved drawability in comparison to the yarn spun from the pure plastic-grade polymer. These two polyblend samples also show the increase in tensile strength and elastic modulus, but compared to the yarns spun from the 90/10 and 70/30 plastic/fiber-grade polymer the improvement of the mechanical properties is not so significant.

The dynamic modulus of discontinuously highly drawn multifilament yarns shows the same trend of improvement by blending the plastic-grade polymer with the fiber-grade CR-polymer in this composition range, as is seen with the elastic modulus. Multifilament yarn spun from the 90/10 plastic/fiber-grade polymer reached the

Table I Tensile Strength, Elastic, and Dynamic Modulus of Continuously Moderately and Discontinuously Highly Drawn PP Multifilament Yarns

Blend Composition (Plastic/Fiber Grade Polymer)	σ_{br} (cN/dtex)		ε_{br} (%)		E_0 (GPa)		E_{dy} (GPa)	
	Mean Value	Standard Deviation	Mean Value	Standard Deviation	Mean Value	Standard Deviation	Mean Value	Standard Deviation
Continuously moderately drawn yarns								
100/0 p/f	2.24	0.17	87.23	9.71	1.77	0.11	6.51	0.38
90/10 p/f	3.65	0.21	75.21	8.91	2.40	0.12	9.53	0.34
80/20 p/f	3.03	0.20	53.69	8.41	2.90	0.16	9.82	0.28
70/30 p/f	3.55	0.26	64.30	9.92	2.59	0.11	10.60	0.60
60/40 p/f	2.76	0.18	82.13	9.79	1.93	0.09	8.69	0.21
50/50 p/f	3.24	0.26	53.90	10.10	3.12	0.18	9.71	0.25
Discontinuously highly drawn yarns								
100/0 p/f	6.91	0.41	16.40	1.31	9.65	0.78	12.20	1.00
90/10 p/f	8.01	0.51	14.41	1.19	11.82	0.91	15.50	1.03
80/20 p/f	7.67	0.37	17.99	1.06	10.25	0.40	13.80	0.66
70/30 p/f	8.13	0.47	14.95	0.98	11.03	0.86	16.60	0.70
60/40 p/f	7.43	0.45	15.40	1.32	9.64	0.71	14.80	0.79
50/50 p/f	7.38	0.55	12.38	1.09	12.51	0.84	16.90	0.72

σ_{br} tensile strength or specific stress at break, i.e., rupture load divided by the original weight per unit length.
ε_{br} extension at break, i.e., change in length divided by the initial length.
E_0 elastic modulus, i.e., tangent of the angle between the initial part of the stress–strain curve and the horizontal axis.
E_{dy} dynamic modulus, i.e., product of the velocity of sound waves and density.

dynamic modulus of 15.5 GPa and yarn spun from the 70/30 plastic/fiber-grade polymer reached even higher value, that is 16.6 GPa.

The elastic modulus is beside tensile strength one of the most important properties of the technical fibers. The static elastic modulus is determined as a ratio of the specific stress vs. strain that is as a tangent of the angle between the initial part of the stress–strain curve and the horizontal axes. The dynamic elastic modulus is determined as a product of the velocity of sound waves and the density.

Many theories have been developed to predict the modulus of a two-phase blends. As a first approach, the simple law of mixtures for the elastic modulus of the blends was consider.[9] The equation for the upper bound of the modulus is given by

$$E_b = E_1\phi_1 + E_2\phi_2 \tag{1}$$

The corresponding equation for the lower bound is given by

$$\frac{1}{E_b} = \frac{\phi_1}{E_1} + \frac{\phi_2}{E_2} \tag{2}$$

where E_1 and E_2 are the elastic moduli of pure components, i.e., the fiber-grade CR-polymer and the plastic-grade polymer, ϕ_1 and ϕ_2 are corresponding volume fractions of both components.

Kleiner et al.[10] proposed the following empirical equation of the second order

$$E_b = E_1\phi_1 + E_2\phi_2 + \beta\phi_1\phi_2 \tag{3}$$

The empirical parameter β is obtained from the following expression

$$\beta = 4E_{12} - 2E_1 - 2E_2,$$

where E_{12} represents the modulus of the 50/50 plastic/fiber-grade polymer.

If we consider the blends as a matrix with inclusions and not as a continuum of two phases, then the equations by Kerner,[11] Uemura and Takayanagi,[12] Halpin and Kardosz,[13] are relevant. In the case of particulate-filled or two-phase composite system, the resultant modulus of the composite is a function of the moduli of the individual pure components, the volume of the weight

fraction, the geometry, and packing of the disperse phase and the Poisson ratio of the matrix.

The Kerner equation is

$$E_b = E_1\left(\frac{\alpha\phi_2 E_2 + \beta(1-\phi_2)}{\alpha\phi_2 E_1 + \beta(1-\phi_2)}\right) \quad (4)$$

where $\alpha = [(7 - 5\nu)E_1 + (8 - 10\nu)E_2]^{-1}$ and $\beta = [15(1 - \nu)]^{-1}$ with ν being Poisson's ratio.

Halpin and Tsai have obtained the following equation [Eq. (6)]

$$E_b = E_1\left(\frac{1 + AB\phi_2}{1 - B\phi_2}\right) \quad (5)$$

where A is an empirical constant, which depends on the stress distribution in the composite, and takes into account the shape and the geometry of the filler phase.

The theoretical predictions on the modulus on the basis of the various theories cited above [eqs. (1)–(5)] are given in Figure 3 together with the experimentally obtained elastic modulus from the tension test and in Figure 4, together with the experimentally obtained dynamic modulus from the sonic-velocity method.

Among all cited equations in this article, the Kleiner's equation is the only one capable of showing a synergistic effect leading to the modulus values above the upper bound. From Figures 3 and 4 it is evident that the experimental data for discontinuously highly drawn yarns spun from the binary blends are best represented by Kleiner's equation. In this equation the term β expresses the magnitude of the deviation from linearity, that is, from the ideal "rule of mixtures behavior," and may be a relative measure of the blend compatibility. A positive β represents a nonlinear synergism, i.e., the criterion for compatibility, while a negative β expresses a nonlinear antagonism, the criterion for incompatibility. The calculated value of β is positive for both moduli, indicating a synergistic effect of blending the different polymer grades on the modulus. For both, the static and dynamic elastic modulus, the largest negative deviation is seen at the 80/20 and 60/40 plastic/fiber-grade polymer blend composition, while the largest positive deviation is seen at the 90/10 plastic/fiber-grade polymer blend composition, suggesting good compatibility of this polymer mixture.

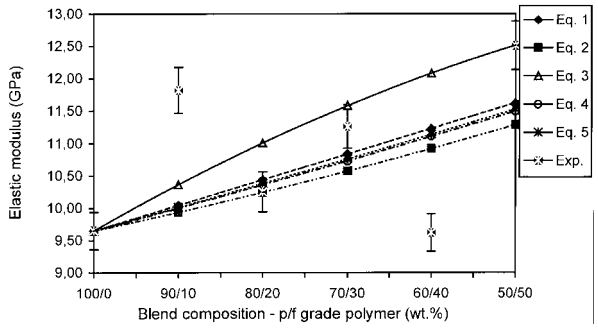


Figure 3 Elastic modulus of discontinuously highly drawn multifilament yarns as a function of the blend composition predicted by eqs. (1)–(5) and the experimentally determined elastic modulus shown with the 5% error amount.

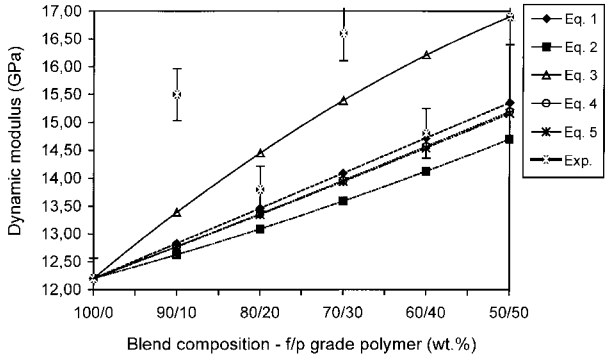


Figure 4 Dynamic modulus of discontinuously highly drawn multifilament yarns as a function of the blend composition predicted by eqs. (1)–(5) and the experimentally determined dynamic modulus shown with the 5% error amount.

Correlation of the Mechanical and Structural Properties

To find structural correlation to the mechanical properties, the crystallinity and molecular orientation were investigated. The crystallinity was determined from the density and average molecular orientation from the sonic-velocity measurements.

It is well known that the crystallinity, the size of crystallites, and their orientation have basic influence on the mechanical properties of drawn fibers. In Figures 5 to 7 the tensile strength (Fig. 5), the elastic (Fig. 6), and dynamic modulus (Fig. 7) are shown as a function of the crystallinity. From Figure 5 it is clearly seen that the crystallinity exhibits no correlation with the tensile strength, and has a decreasing tendency and low correlation for the elastic (Fig. 6) and dynamic modulus (Fig. 7).

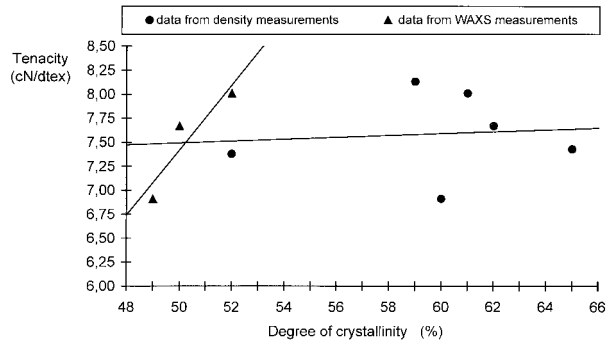


Figure 5 Tensile strength of discontinuously highly drawn multifilament yarns as a function of crystallinity.

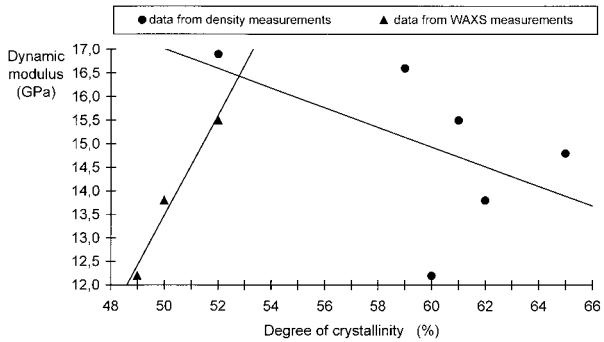


Figure 7 Dynamic modulus of discontinuously highly drawn multifilament yarns as a function of crystallinity.

To reveal this unexpected result multifilament yarns spun from the 100/0, 90/10, and 80/20 plastic/fiber-grade polymer were additionally investigated by the WAXS method. The degree of crystallinity, the crystalline orientation, and the apparent crystallite size were determined for these samples. Figures 5–7 show that for these yarns the tensile strength as well as elastic and dynamic modulus increase with the degree of crystallinity, as expected.

The WAXS film patterns of drawn multifilament yarns, taken so as to determine the crystallographic crystal system present, are shown in Figure 8 for continuously moderately and in Figure 9 for discontinuously highly drawn yarns.

Continuously moderately drawn multifilament yarns show wide equatorial spots of the (110), (040), and (130) planes united in one reflection on the WAXS film pattern (Fig. 8). Overlapping reflections of (110), (040), and (130) planes are united in one equatorial peak at the scattering angle $2\theta = 14.6°$, whereas a second broad peak exists at $2\theta = 21.2°$. The presence of so-called oriented "smectic" structure in these samples was confirmed by the WAXS curves also.

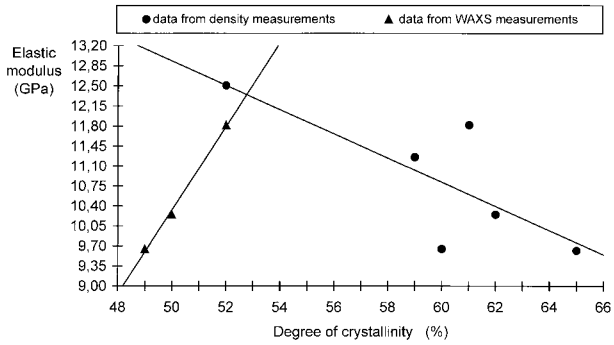


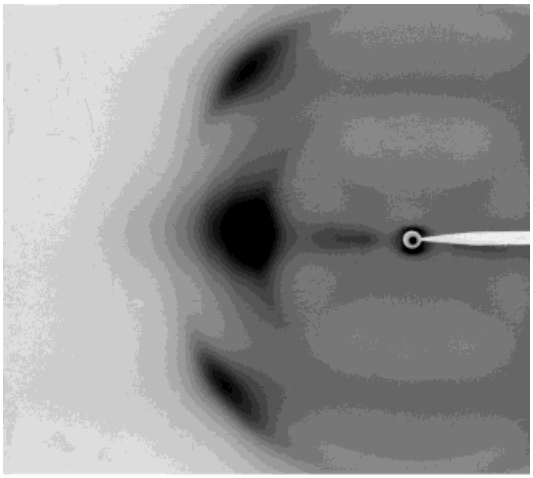
Figure 6 Elastic modulus of discontinuously highly drawn multifilament yarns as a function of crystallinity.

Discontinuously highly drawn multifilament yarns show discrete reflections of the α-iPP crystal structure on the WAXS film pattern (Fig. 9). This typical pattern of a well-oriented fibrous structure, with the above broad equatorial maximum split up into narrower (110), (040), and (130) reflections is clearly seen on the WAXS curves also. Only a c-axis orientation of the monoclinic crystallites is present in these yarns.

The apparent crystallite size of all continuously moderately drawn yarns is low, i.e., 3.4 nm. With the additional hot drawing, the size of crystallites has increased to a value of 9.5 nm. It was also found that all highly drawn yarns show no differences in the lateral crystallite dimensions.

The mechanical properties of drawn yarns depend beside on the crystallinity and number of taut tie molecules, also on the orientation of the macromolecules in the crystalline and amorphous domains. In Figures 10–12 the mechanical properties of discontinuously highly drawn multifilament yarns are plotted against the average orientation function. It is clearly seen that the tensile strength, the elastic, and dynamic moduli increase with the molecular orientation. The tensile strength as a function of the molecular orientation has a linear character, although the values deviate in some cases (Fig. 10). Linear dependence is also seen in the case of both moduli (Figs. 11 and 12), correlation factor being of the order of 0.75 for the static elastic modulus and 0.95 for the dynamic modulus.

According to Peterlin,[14] both the tensile strength and elastic modulus increase with the fraction of



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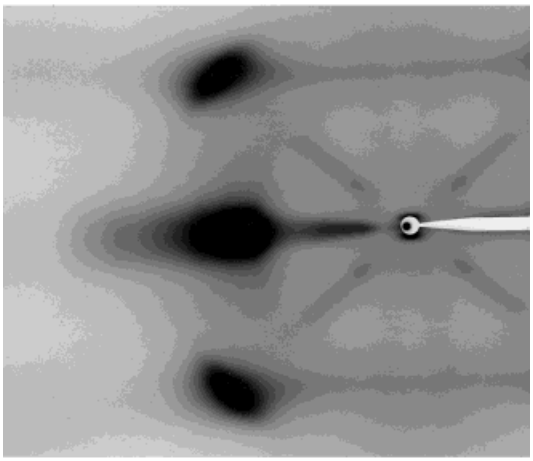

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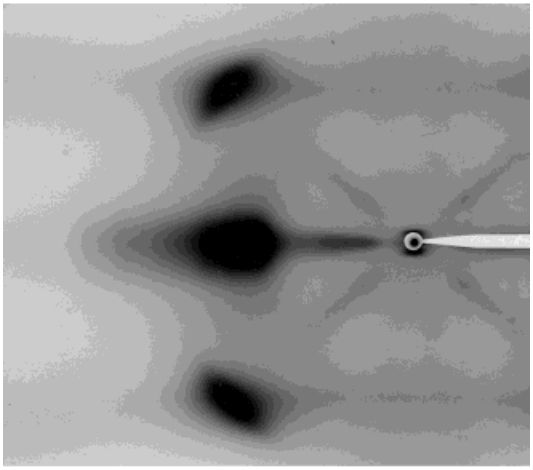
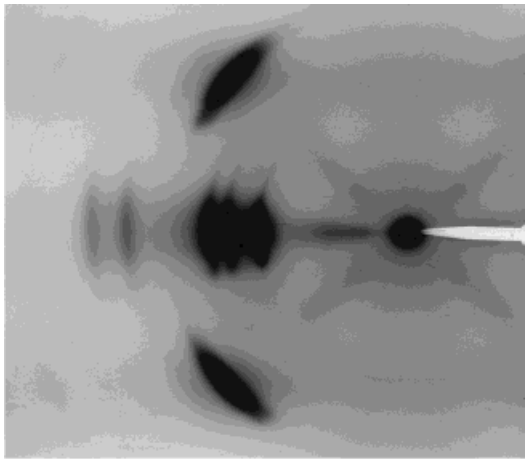


(c)

Figure 8 WAXS film pattern of continuously moderately drawn multifilament yarns spun from (a) pure plastic-grade polymer, (b) 90/10 plastic/fiber-grade polymer (c), 80/20 plastic/fiber-grade polymer.



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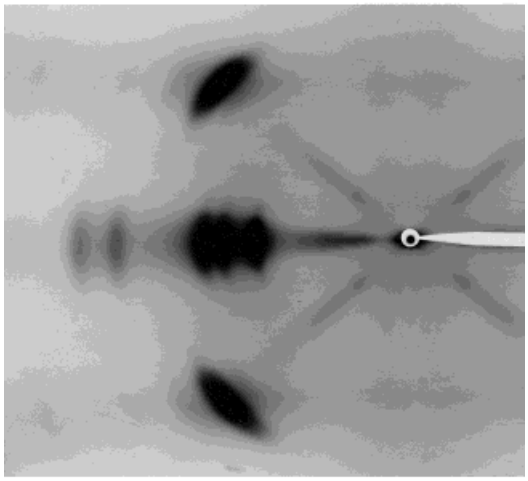

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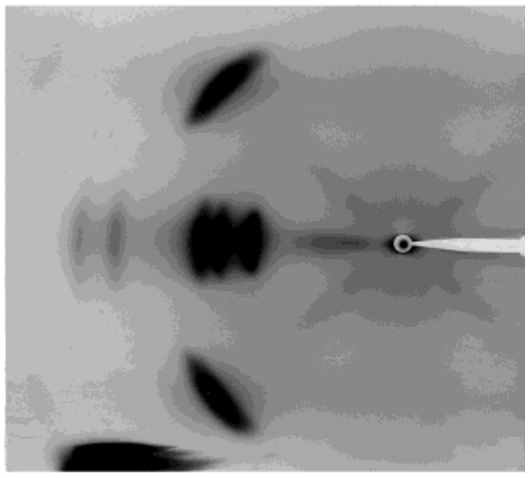

(c)

Figure 9 WAXS film pattern of PP discontinuously highly drawn multifilament yarns spun from (a) pure plastic-grade polymer, (b) 90/10 plastic/fiber-grade polymer, (c) 80/20 plastic/fiber-grade polymer.

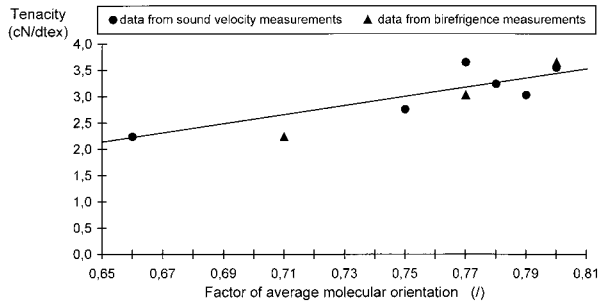

Figure 10 Tensile strength of discontinuously highly drawn multifilament yarns as a function of average molecular orientation function.

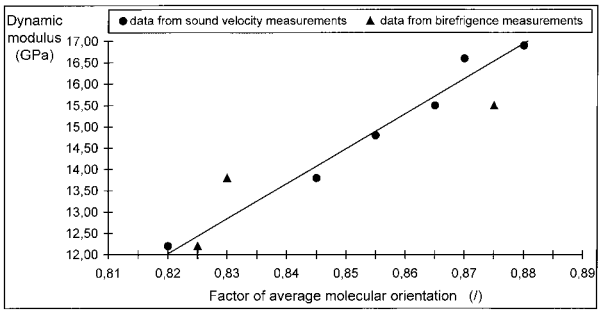

Figure 12 Dynamic modulus of discontinuously highly drawn multifilament yarns as a function of average molecular orientation function.

the taut tie molecules and alignment of macromolecules along the draw direction. White et al[15] attributed the increasing elastic moduli and tensile strength in drawn fibers mainly to the increasing molecular orientation in the amorphous domains. The mechanical properties of multifilament yarns investigated in our study show the best agreement with the amorphous orientation, the correlation factor being in all cases higher than 0.95.

The electron microscopy was used to examine in some details the morphological changes, which have occurred on drawing. Under the microscope all continuously moderately and most of the discontinuously highly drawn multifilament yarns exhibited smooth surface, without any features. At some parts of discontinuously highly drawn multifilament yarns spun from the 90/10 and 70/30 plastic/fiber-grade polymer craze-like markings perpendicular to the filament axes were detected. These two yarns also exhibited, besides transparent filaments, some limited parts of filaments, which were opaque. At the optical birefringence measurements these opaque places revealed the presence of transverse lines. These features are the consequence of defects present, possibly a regions of reduced density because of more voided structure. This explains discrepancy found between crystallinity determined from the density data and from the WAXS curves in correlation to the mechanical properties of highly drawn yarns.

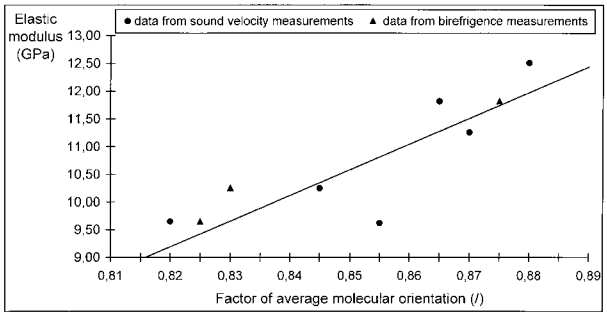

Figure 11 Elastic modulus of discontinuously highly drawn multifilament yarns as a function of average molecular orientation function.

CONCLUSIONS

Melt spinning of binary polypropylene blends of different molecular weights on a laboratory spin-draw device, with immediate continuous and subsequent discontinuous drawing at elevated temperature, enables the production of PP multifilament yarns suitable for industrial aplications. By blending the plastic-grade polymer with the fiber-grade CR-polymer, maximization of the tenacity and elastic modulus has been achieved. The elastic modulus determined from the initial slope of the stress–strain curve, and the dynamic modulus determined from the sonic- velocity measurements, gave the best fit to Kleiner's simplex equation. The large positive deviation reflected by highly drawn multifilament yarns, spun from the 90/10 plastic/fiber-grade polymer, from the values predicted by Kleiner's simplex equation, suggests good compatibility of this polymer mixture. Improved spinnability and drawability of blended samples led to the yarns with the tensile strength over 8 cN/dtex, elastic modulus over 11 GPa, and dynamic modulus over 15.5 GPa.

The structural investigations have shown that, with the severe drawing at an elevated temperature, a well-oriented fibrillar structure,

with only c-axis–oriented monoclinic crystalline modification present, develops from an oriented "smectic" structure. The morphological studies have shown that a low correlation of the mechanical properties with the crystallinity evaluated from the density is the consequence of a localized highly voided structure. The correlation of the mechanical and structural properties has shown that the improved mechanical behavior is mainly associated with the higher orientation of macromolecules in the amorphous domains.

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