Fibres from polypropylene and liquid crystal polymer blends: 2. Effect of extrusion and drawing conditions

Y. Qin, D. L. Brydon, R. R. Mather* and R. H. Wardman
Department of Technology, The Scottish College of Textiles, Galashiels, TD1 3HF, UK
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The drawing conditions were studied for a polyblend fibre of polypropylene (PP) and a thermotropic liquid crystal polymer (LCP). It was found that, in one-stage drawing, the LCP fibrils were split into short fragments with aspect ratios of around 10. Although fibre properties were improved with the increase in drawing temperature, the best properties of the polyblend fibre from one-stage drawing were poorer than those of the pure polypropylene fibre. A two-stage drawing process was studied in terms of the ratios and temperatures of the first- and second-stage drawings. It was found that the fibre properties were strongly affected by these parameters. At best, the fibre tenacity and initial modulus in the two-stage drawing were 14% and 39% higher than the highest values from the one-stage drawing. The LCP fibrils were found to split to a much lower extent in the two-stage drawing. The optimum drawing procedure was applied to a series of polyblend fibres extruded under various extrusion rates and with a number of draw-down ratios.

(Keywords: polypropylene; liquid crystal polymer; polyblend fibre; drawing; mechanical properties)

INTRODUCTION

One of the distinct differences between liquid crystal polymers (LCPs) and conventional polymers is that, when in solution or melt, the LCPs exist in oriented poly-domains that can be aligned in the direction of flow with little shear or elongational strain[1]. LCP fibres exhibit a highly oriented structure even in the as-spun state and require no stretching process. Conventional polymers usually have short relaxation times and the as-spun fibres possess a random structure. To obtain fibre strength and stiffness, conventional polymers require a stretching process to achieve molecular orientation[2]. For a polyblend fibre comprising a liquid crystal polymer and a conventional polymer, the as-spun structure is composed of highly oriented LCP domains, which are stiff and inextensible, and the conventional polymer matrix, which is flexible and requires a stretching process to obtain strength. Part 1 of this work[3] has shown that, when the polyblend fibre of polypropylene and Vectra A900 is stretched, the LCP fibrils split into short fragments, which significantly reduces the capacity of the LCP as the reinforcing component. It is therefore desirable to preserve a good length of the LCP fibrils. In part 2 we present the results of our studies on the drawing conditions. We investigated the effect of the drawing temperatures and ratios in one- and two-stage drawing, and developed an optimum drawing procedure, which we applied to some polyblend fibres spun under various extrusion conditions.

EXPERIMENTAL

An as-spun polyblend fibre with a polypropylene/Vectra A900 ratio of 100/10 (wt/wt) was prepared via the procedure described previously in part 1. This fibre was prepared with a temperature profile of 230/285/285/285/280/280°C, an extrusion rate of 20 m min^{-1} and a take-up speed of 60 m min^{-1}. The fibre had a linear density of 47 tex (47 g/1000 m).

One-stage drawing was carried out by stretching the as-spun fibre between two pairs of advancing rollers over a hot plate at temperatures of 110, 120, 130, 140 and 150°C. The feeding speed was 6 m min^{-1} and the fibre was stretched to the highest ratio before fracture. The draw ratio was calculated from the fibre thickness of the as-spun and drawn fibres.

In studying the two-stage drawing process, the as-spun fibre was fed at 6 m min^{-1} and was partially stretched in the first stage. This was followed by a second-stage drawing where the partially drawn fibre was fed through the drawing unit at a feeding speed of 20 m min^{-1} and with a maximum drawing speed. The temperature and the ratio of the first-stage drawing and the temperature of the second stage were varied as will be mentioned later. Eventually, the following optimum procedure was developed. The as-spun fibre was stretched to a ratio of 6 at 120°C and the partially drawn fibre was stretched again at 160°C to the maximum ratio before fracture.

The as-spun and drawn fibres were characterized as described in part 1[3].

RESULTS AND DISCUSSION

Figures 1 and *2* show the maximum draw ratios and the tensile properties of the polyblend fibre drawn at temperatures between 110 and 150°C. It is clear that higher draw ratios were possible at higher temperatures. The fibre tenacity and modulus increased

* To whom correspondence should be addressed

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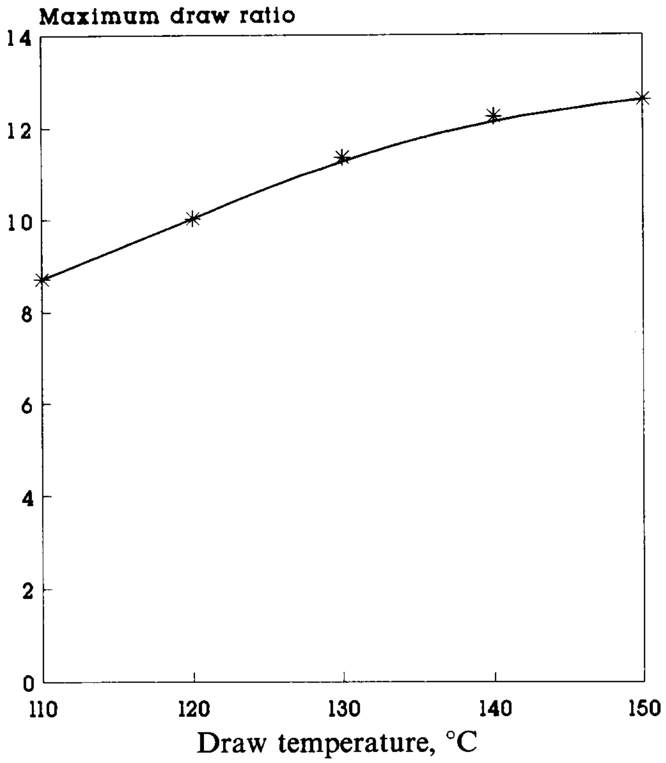

Figure 1 Maximum draw ratio vs. drawing temperature

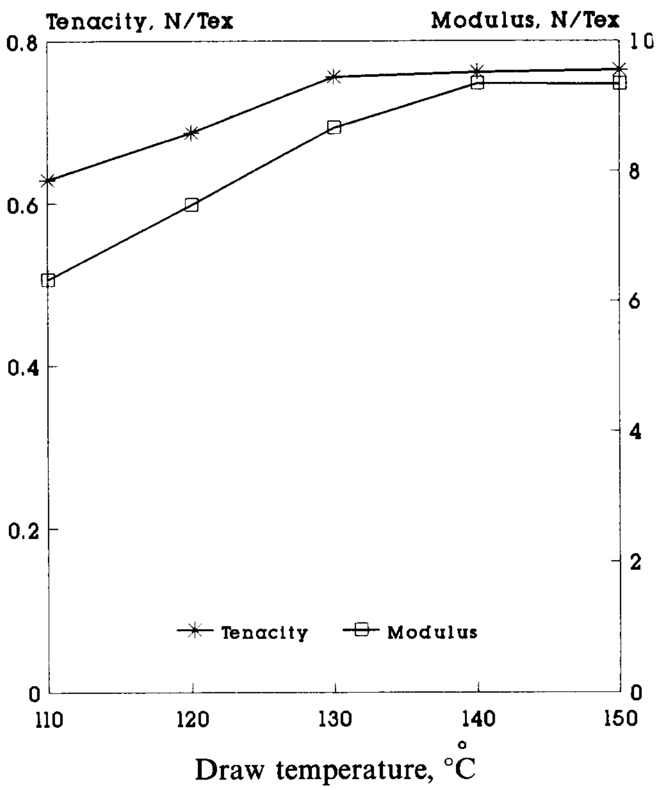

Figure 2 Fibre tenacity and initial modulus vs. drawing temperature

with increase in drawing temperature. The highest tenacity of 0.764 N/tex was obtained at 150°C; above this temperature, drawing was not possible as the fibre began to stick onto the hot plate. *Table 1* shows the fibre properties of pure polypropylene (PP) and PP/Vectra A900 blend extruded at the same conditions and drawn at 150°C both to the maximum extent. It can be seen that, although the polyblend fibre had a slightly higher initial modulus, the pure polypropylene fibre had a much higher tenacity than the polyblend fibre. The hot-stage photomicrographs, as shown in *Figure 3*, revealed that, whereas the as-spun polyblend fibre contained well oriented thin LCP fibrils, they were split into short pieces in the drawn fibre. The aspect ratios of these fragments were around 10, which is very short. A large aspect ratio of above 40 has been suggested as necessary for an effective reinforcement[4].

Part 1[3] of this work has shown that a two-stage drawing process resulted in a low degree of fracture of the LCP fibrils and as a result both the initial modulus and fibre tenacity can be improved for the polyblend fibres. The two-stage drawing process comprised mainly four experimental parameters, i.e. the temperature and ratio of the first-stage drawing and the temperature and ratio of the second-stage drawing. In order to obtain maximum fibre properties, the ratio of the second-stage

Table 1 Fibre properties of PP and PP/LCP blend drawn at 150°C

	PP	PP/Vectra A900 blend
Maximum draw ratio	12.1	12.6
Fibre thickness (tex)	3.70	3.71
Tenacity (N/tex)	0.960	0.764
Elongation (%)	23.6	16.3
Initial modulus (N/tex)	8.32	9.33

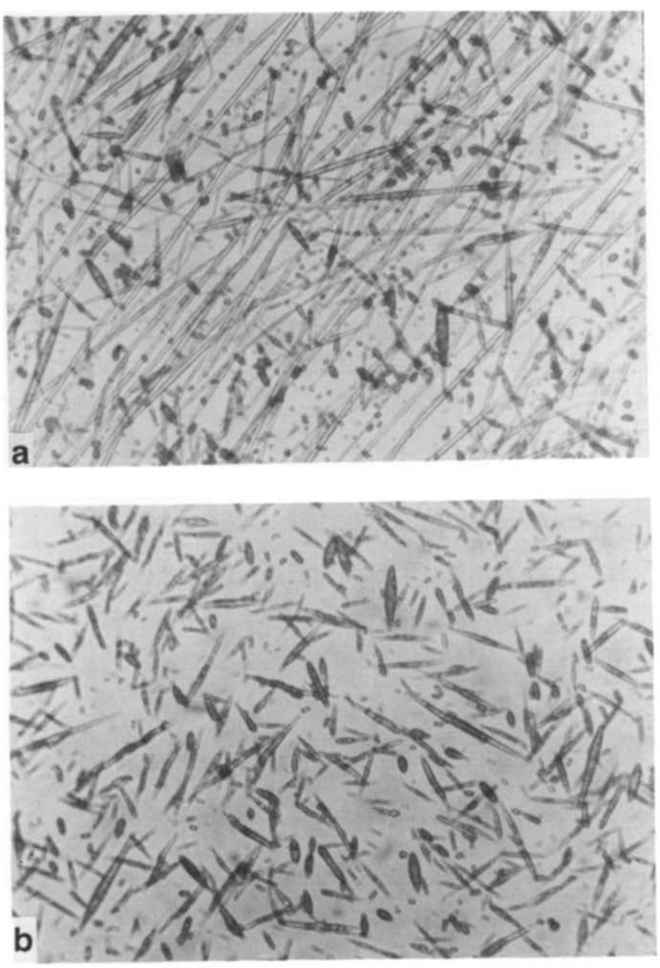

Figure 3 Hot-stage photomicrographs of the PP/LCP polyblend fibres: (a) as-spun; (b) one-stage fully drawn at 150°C

drawing is usually fixed at the maximum. Therefore, the temperatures of the two stages and the ratio of the first-stage drawing were studied in order to develop an appropriate drawing procedure.

Table 2 shows the fibre properties from a two-stage drawing with the first-stage ratio at approximately 5 and with varying temperatures. Initially it was found that, when the temperature was too high or too low, it was difficult to obtain evenly drawn samples. The optimum temperatures were between 120 and 140°C. As is shown in *Table 2*, the highest fibre tenacity was obtained at a first-stage temperature of 120°C. The fibre tenacity of 0.806 N/tex and the initial modulus of 10.0 N/tex are about 5% and 7% higher than the highest values from one-stage drawing.

Table 3 shows the drawing conditions and fibre properties of a series of samples drawn at a first-stage temperature of 120°C to ratios between 5 and 7 and a second-stage drawing at 150°C to the maximum extent. The fibre properties of the sample drawn at 120°C to the maximum extent are also presented in the table for comparison. It can be seen that, apart from the significant increases in the maximum draw ratios, the fibre tenacities were generally much higher in the two-stage drawing. The highest tenacity of 0.868 N/tex was obtained at a first-stage ratio of 6.1.

From *Tables 2* and *3* the optimum temperature and ratio for the first-stage drawing were decided as 120°C and 6 respectively. *Table 4* shows the effect of temperature on the maximum draw ratio and fibre properties of the second-stage drawing. It is clear that the second-stage draw ratios were much higher at higher temperatures. Both fibre tenacity and initial modulus showed improvement with increase in drawing temperature. In particular, the initial modulus showed a significant increase. The value of 13.0 N/tex at 160°C is 36% higher than that at 130°C.

Table 2 Effect of the first-stage drawing temperature

	Temperature, T_1 (°C)		
	120	130	140
r_1	5.3	5.2	5.2
T_2 (°C)	150	150	150
r_2	2.8	2.9	2.8
Overall draw ratio	14.8	15.0	14.6
Fibre thickness (tex)	3.17	3.11	3.19
Tenacity (N/tex)	0.806	0.795	0.733
Elongation (%)	15.8	13.1	14.0
Initial modulus (N/tex)	10.00	10.79	8.95

Figure 4 shows the hot-stage photomicrographs of the as-spun fibre drawn to a ratio of 6 and to the maximum extent (10.0) at 120°C. It is clear that the LCP fibrils were split to a much smaller extent when the fibre was partially drawn. Comparing *Figures 4b* and *3b*, it is interesting to see that the LCP fibrils were split to a low extent at a low temperature. This may suggest that, at a low temperature, the polypropylene matrix withstands a large percentage of the stress applied in the drawing process.

Figure 5 shows the hot-stage photomicrographs of the fibres drawn at second-stage temperatures of 130 and 160°C respectively. Apparently, the LCP fibrils were better preserved at the higher temperature. Considering the fact that the draw ratio at 160°C is higher than at 130°C, the low degree of splitting at 160°C can be attributed to the reduction in the interfacial adhesion between the polypropylene matrix and the LCP fibrils with the increase in drawing temperature, producing mobility for the LCP fibrils during stretching. This is a reasonable suggestion because, for immiscible polymer blends, poor interfacial adhesion and phase separation can be produced at increased temperature[5]. There can be two ways that a high second-stage drawing temperature is beneficial. First, as is common elsewhere[2], a high temperature can reduce the intermolecular entanglement and increase the molecular mobility, thereby giving a high draw ratio. Secondly, when the polyblend fibre is drawn and cooled down, the shrinkage force from thermal expansion can result in an improved adhesion between the polypropylene and the LCP phases. A similar 'shrinkage bonding' in polyblend fibres has been mentioned by Paul[5] where, in a polyblend of nylon and poly(ethylene terephthalate) (PET), the interfacial adhesion can be improved through the different thermal properties of nylon and PET. In the as-spun state, PET is mainly amorphous whilst nylon is semicrystalline. Therefore, when the fibre is composed of a core of nylon and a sheath of PET, the interfacial adhesion can be improved by a heat treatment; the crystallization of the

Table 4 Effect of the second-stage draw temperature, $T_1 = 120°C$, $r_1 = 6.0$

	T_2 (°C)			
	130	140	150	160
r_2	2.0	2.2	2.5	2.8
Fibre thickness (tex)	3.84	3.52	3.05	2.76
Tenacity (N/tex)	0.729	0.778	0.806	0.873
Elongation (%)	17.0	16.4	14.4	12.4
Initial modulus (N/tex)	9.54	9.78	11.27	13.02

Table 3 Effect of the first-stage draw ratio, $T_1 = 120°C$

	r_1					
	5.0	5.6	6.1	6.6	7.3	10.0
T_2 (°C)	150	150	150	150	150	
r_2	2.9	2.6	2.5	2.4	2.1	1
Overall draw ratio	14.5	14.6	15.2	15.8	15.3	10.0
Fibre thickness (tex)	3.24	3.22	3.10	2.96	3.03	4.67
Tenacity (N/tex)	0.834	0.825	0.868	0.849	0.840	0.686
Elongation (%)	14.8	13.6	15.1	13.6	13.6	22.7
Initial modulus (N/tex)	10.07	10.71	11.12	11.07	11.11	7.48

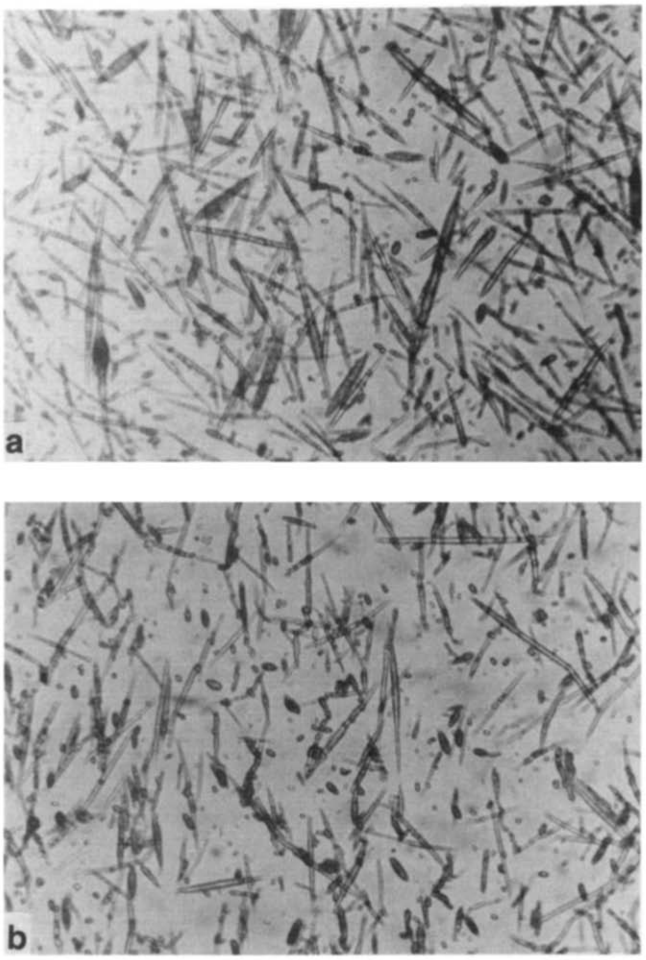

Figure 4 LCP phases from one-stage drawn fibres: (a) to a ratio of 6 at 120°C; (b) to a ratio of 10.0

PET produces a strong shrinkage force that holds the nylon core.

From the results shown in the previous tables, it is clear that the properties of the polyblend fibres were strongly affected by the drawing conditions. The highest tenacity and modulus from a two-stage drawing are 14% and 39% higher than the best from a one-stage drawing. The appropriate conditions seem to be $T_1 = 120°C$, $r_1 = 6$, $T_2 = 160°C$ and r_2 being the maximum.

The two-stage procedure was applied to draw a series of polyblend fibres. *Table 5* shows the drawing conditions and fibre properties of four samples of polyblend fibres that were produced using extrusion rates between 16.2 and 37.5 m min^{-1}. It can be seen that the sample from the highest extrusion rate had a much lower overall draw ratio. The highest tenacity was obtained at an extrusion rate of 25 m min^{-1}.

It has been mentioned in the literature[6,7] that the formation and structure of LCP fibrils are affected by shear and elongtional strain applied to the polymer melt. For the liquid crystal polymer, the fibre structure and mechanical properties are directly related to the shear rate, as the molecules in the mesophase are more readily aligned in the direction of flow at higher shear rates. In the present work, when the polymer melt is extruded through the spinneret capillary, the degree of shearing is directly proportional to the extrusion rate. In part 1, it has been reported that, at a higher extrusion rate, the polyblend fibre contained thin LCP fibrils. Although it was difficult to measure the properties of these fibrils, it can be reasonably assumed that at a high extrusion rate the LCP fibrils have a better oriented structure and higher strength and modulus. This high strength and modulus can produce a strong resistance towards the mobility of the polymer molecules during hot stretching. In fact, results in *Table 5* show that the overall draw ratios generally decreased with increase in the extrusion rate. Therefore, the extrusion rate can affect the fibre structure in two ways. First, an increase in the extrusion rate can deform the LCP phase and produce thin and well oriented fibrils. Secondly, as the strength and modulus of these fibrils are improved with the increase in extrusion rate, the fibrils can generate a strong resistance towards the

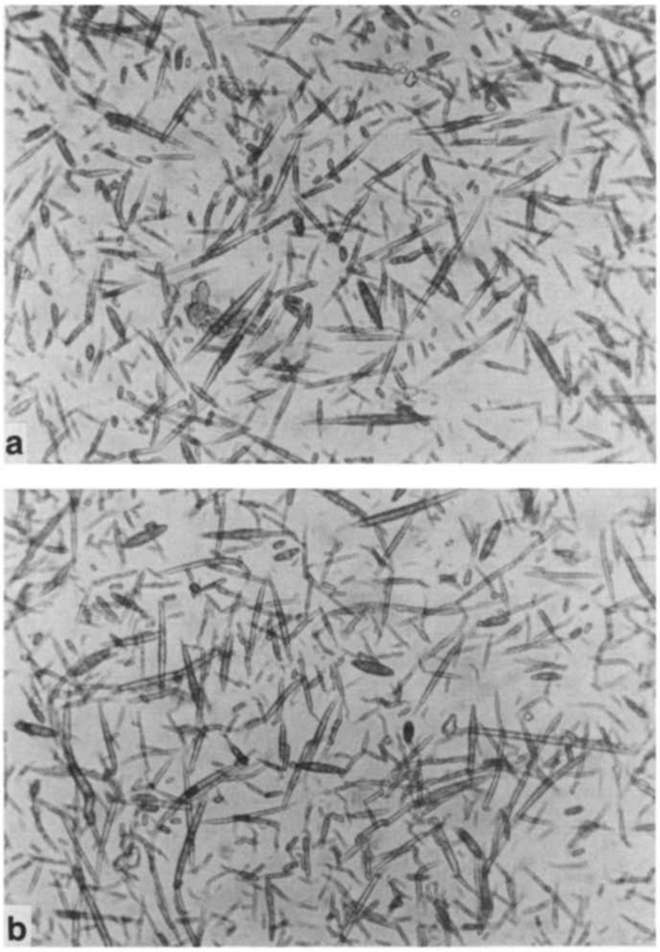

Figure 5 LCP phases of two-stage fully drawn fibres: (a) at a second-stage drawing temperature of 130°C; (b) at 160°C

Table 5 Effect of the extrusion rate on fibre properties at a draw-down ratio of 4

	Extrusion rate (m min^{-1})			
	37.5	25.0	20.0	16.2
T_1 (°C)	120	120	120	120
r_1	5.9	5.8	5.7	5.7
T_2 (°C)	160	160	160	160
r_2	2.0	2.7	3.0	3.0
Overall draw ratio	11.8	15.7	17.1	17.1
Fibre thickness (tex)	2.90	2.44	2.13	2.05
Tenacity (N/tex)	0.819	0.851	0.827	0.838
Elongation (%)	16.0	10.9	10.6	10.6
Initial modulus (N/tex)	9.65	12.51	12.84	12.68

Table 6 Effect of the draw-down ratio on fibre properties at an extrusion rate of $25\,m\,min^{-1}$

	Draw-down ratio			
	3	4	5	6
T_1 (°C)	120	120	120	120
r_1	5.8	6.3	6.0	6.0
T_2 (°C)	160	160	160	160
r_2	2.5	2.4	2.2	2.1
Overall draw ratio	14.5	15.1	13.2	12.6
Fibre thickness (tex)	3.25	2.35	2.24	1.96
Tenacity (N/tex)	0.802	0.886	0.791	0.796
Elongation (%)	13.6	12.4	12.4	12.4
Initial modulus (N/tex)	10.16	12.24	10.65	11.07

stretching of the as-spun fibres, thus reducing the maximum draw ratio and giving an adverse effect to the mechanical properties of the fibre.

Table 6 shows the drawing conditions and fibre properties of four samples prepared with an extrusion rate of $25\,m\,min^{-1}$ and draw-down ratios between 3 and 6. In general, the maximum draw ratio was low at a high draw-down ratio. This effect is similar to that observed earlier with a high extrusion rate. In part 1, it has been reported that, at a higher draw-down ratio, the LCP fibrils were better aligned along the fibre axis. Literature information[6,7] has also revealed that the elongational strain below the die head is an important factor in the formation of LCP fibrils. The mechanical properties for extruded strands and films were improved with a high draw-down ratio. It can be generally concluded that a high draw-down ratio produces well oriented LCP fibrils. In this work, the as-spun fibres are hot stretched to produce molecular orientation for the polypropylene matrix. The improved strength and modulus of the LCP fibrils can lead to a strong resistance to stretching and give a low draw ratio. As with extrusion rate, the draw-down ratio can affect the structure of the polyblend fibres in two ways. First, a high draw-down ratio can produce well oriented LCP fibrils that align along the fibre axis. Secondly, as the strength and modulus of these fibrils increase with the increase in the draw-down ratio, the LCP fibrils can generate a strong resistance towards hot stretching, thereby giving an adverse effect to the fibre properties.

As can be seen in *Table 6*, the fibre tenacity showed a maximum value of 0.886 N/tex at a draw-down ratio of 4.

CONCLUSIONS

1. In one-stage drawing, the properties of the polyblend fibre were improved with increase in the drawing temperature. Higher draw ratios were possible at higher drawing temperatures.
2. The fibre properties were generally better in two-stage drawing than in one-stage drawing. They were strongly affected by both the ratio and the temperature of the first- and second-stage drawing.
3. The improvement in the fibre properties in the two-stage drawing can be the result of high draw ratios and low extent of fracture of the LCP fibrils.
4. The properties of the polyblend fibres were also affected by the extrusion conditions. The highest tenacity of 0.886 N/tex was obtained at an extrusion rate of $25\,m\,min^{-1}$ and a draw-down ratio of 4, coupled with the optimum drawing temperatures and ratios of $T_1=120$°C, $r_1=6$, $T_2=160$°C and r_2 the maximum ratio.

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