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The application of factorial experimental design to the processing of polypropylene fibres

R. D. YANG, R. R. MATHER*, A. F. FOTHERINGHAM
Heriot-Watt University, Scottish Borders Campus, Galashiels, TD1 3HF, UK
E-mail: r.r.mather@hw.ac.uk

This paper outlines the application of factorial experimental design to the processing of polypropylene (PP) fibres. Two examples are given. The first covers the effect of melt-extrusion conditions on the overall orientation of the PP macromolecular chains in as-spun fibres. The second covers the effect of drawing conditions on crystallographic order and chain orientation in the first-stage drawing of as-spun PP fibres.

1. Introduction

The control of the processing of textile fibres is key to obtaining the correct fibre properties to meet a range of desired technical specifications. It is, therefore, highly desirable, or even essential, that a coherent overview of the processing can be presented through the identification of those processing control parameters which predominantly determine the properties of the fibres produced.

In any investigation into the processing of synthetic textile fibres, the structure and properties of the fibre have necessarily to be monitored under a wide variety of process control parameters: notably those specifying the raw polymer, extrusion parameters and fibre drawing parameters. All of these parameters will influence the properties of the resultant fibre. In addition, different parameters may interact with one another: the effects of a particular parameter may be different when other parameters are set at different values. Clearly then in synthetic fibre processing, there is a need for sound experimental design and careful analysis of the experimental results. Only in this way can the effect of a set of control parameters on a measured fibre characteristic be evaluated.

In the past, it has been common practice in the textile industry to adopt a univariate, "one factor at a time" approach to optimise the properties of the fibres produced. In this traditional approach, only one parameter at a time is varied, while all the others are maintained constant. The same procedure is then repeated for other control parameters. Whilst this approach possesses the advantages of simplicity, in that experimental design and interpretation of results do not necessarily require any special knowledge of statistics, it nonetheless has severe disadvantages. These disadvantages include the very large experimental scale often needed and difficulties in identifying and assessing any interaction effects between control parameters. In complex processing technologies, such as synthetic fibre production, these disadvantages constitute a major obstacle to the development of fibre processing technology. Moreover, when significant interaction effects are present, conclusions reached through the univariate approach are generally misleading. The univariate approach is, therefore, often unsound.

A more rigorous approach is to adopt factorial experimental design, first proposed by Fisher around seventy years ago [1] and subsequently developed by Taguchi [2, 3]. In this approach, many control parameters can be varied together within the experimental design [4–7]. The resulting effects, including interaction effects, can then be evaluated through systematic statistical analysis, and more reliable conclusions can be drawn.

In this paper, an integrated approach linking factorial experimental design with systematic statistical analysis is illustrated for polypropylene (PP) fibre processing. Examples for melt extrusion and fibre drawing are taken from our current work. In the past thirty years or so, PP fibres have been developed for a wide range of applications, and the manufacture and sales of PP fibres continue to increase rapidly [8]. Each one of these applications requires a particular set of specifications in terms of fibre properties. By adopting the approach of factorial experimental design, the individual processing control parameters which have significant effects on the structure (as assessed, for example, from crystallographic order and overall orientation of the PP macromolecular chains) and mechanical properties (e.g. tenacity, elongation to break and specific secant modulus) of as-spun and drawn fibres can be identified. Significant interactions between control parameters can also be identified.

Factorial experimental design in the study of PP fibre processing has been adopted by Andreassen *et al.* [9–12]. They report that they used a 'reduced factorial design', although the meaning of this term is not completely clear. The authors state that some of their results were analysed using statistical methods, but no

*Author to whom all correspondence should be addressed.

information on these methods is given. Fotheringham *et al.* [13–15] have also studied the processing of PP fibre, using factorial experimental design. An especially noteworthy result was the significant interaction effect on fibre tenacity between the temperature of one drawing roller and the speed of another drawing roller.

2. Factorial experimental design

In a full factorial experimental design, all possible combinations of the various control parameters are included. Thus, if k control parameters are investigated at q settings each, a series of q^k experimental trials is required. A practical drawback, therefore, is often the need for a large number of experiments. To obviate this difficulty, we have used a fractional factorial design, which requires fewer trials. This approach is particularly useful in initial screening experiments, when the most important control parameters can be identified prior to a more thorough detailed evaluation of them in subsequent experiments.

In a fractional factorial design, fewer trials, compared with the full factorial design, can be used by reducing the design resolution, i.e. confounding some of the combinations together. It is, however, general practice that three-factor and higher interactions are assumed insignificant in comparison with main effects from individual control parameters and two-factor interactions [7]. Thus, if these higher interactions are confounded with main effects or two-factor interactions, any conclusion reached, e.g. statistical significance of effects, from the confounding is assumed safely from the main effects or two-factor interactions. In fact, the effects of k factors at two levels can be analysed using one-half (2^{k-1}), one-quarter (2^{k-2}), and so on, of a 2^k design. Such designs of small scale are particularly common in factor screening experiments, performed in the early stages of an investigation to identify significant individual control parameters. The disadvantages of a fractional factorial design can be lack of resolution of some control parameters when the design resolution is too low.

An L16 design matrix [7] was used for the sixteen screening trials in our melt-extrusion experiment, involving seven control parameters and two levels for each parameter. For this system (seven factors and two levels), the L16 design matrix allows significant individual control parameters to be identified, but some two-factor interactions have to be combined. Initial and duplicate sets of the sixteen trials were carried out, and conducted in a random sequence for each set. In addition, a set of confirmatory experimental trials was conducted, to confirm (& maybe reject) individual parameters and to identify any significant interactions between pairs of control parameters. The same L16 design matrix was used for the confirmatory experiment, but with a different system which involved five control parameters and two levels for each parameter, allowing significant interactions from any pair of the control parameters involved to be identified.

For the drawing experiments, a different L16 array was used involving two process control parameters. The small number of factors used for the L16 array allows more levels to be investigated. Thus, instead of two levels as used in most of the previous experiments, four levels were used here.

A variety of standard methods of statistical analysis were employed, including effects plots and analysis of variance (ANOVA) [4–7]. The effects plots reveal graphically the relative magnitude and direction of the effects of individual process control parameters [7], as well as relative magnitude of the interactions between pairs of processing parameters, from the factorial design we have used.

3. Experimental methods

Raw PP granules were extruded on a pilot-plant single screw Labspin extruder, manufactured by Extrusion Systems Limited, as described elsewhere [16]. There were three heating zones in the extruder barrel, one zone in the metering pump and two zones in the die head, which contained a filter package and a spinneret of 55 circular holes. The spinning temperature was altered by adjusting the heaters in the metering pump and die head. The extruded filament was cooled by an air flow in a quenching chamber. An aqueous-based spin finish, supplied by Benjamin Vickers and Sons Ltd., was applied after the filament had been cooled.

Two grades of PP granule were used: PPH9069 from Petrofina and VC18 from Borealis. Their melt flow indices were determined as 22.4 g/10 min and 17.7 g/10 min, respectively. The determination of optical birefringence data and X-ray crystallographic data have been described elsewhere [17].

The seven control parameters for the melt-extrusion experiments were: melt flow index of the PP granules (MFI), hole size (area) of spinneret (HS), metering pump speed (MPS), spinning temperature (ST), quenching air speed (QAS), application speed of spin finish (SFS) and winding speed (WS). In order to expand design space, the two levels of each parameter were separated as far apart as possible from one another, but such that 'as-spun' fibre could be reliably produced. In the confirmatory trials, QAS and SFS were excluded, because analysis of variance showed these two parameters to be the least significant.

As-spun fibres were drawn on a draw-frame of scale commensurate with that of the extrusion equipment. The fibres were wound around an initial roller, roller 1, passed through a hot plate, plate 1, and then wound around a further roller, roller 2. The temperature of roller 1 and plate 1 (TRP1) and the speed of roller 2 (SR2) which controls the degree of drawing, given a fixed speed of roller 1 (50 m min^{-1}), were used as control parameters for the first-stage drawing. The temperature of roller 2 was set at room temperature. The sixteen trials were conducted in a random sequence.

4. Melt-extrusion

As an illustration, Fig. 1 shows the effects plot of the original experimental trials for birefringence (Δn). Seven of the columns are taken up by the individual

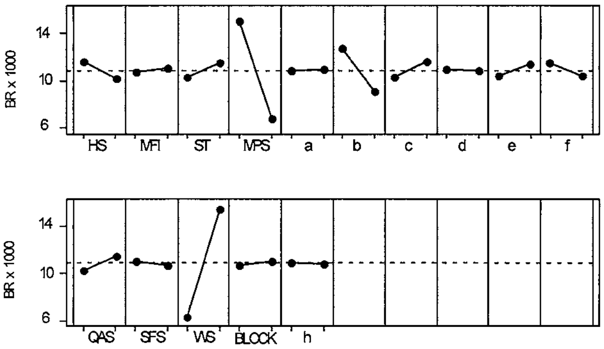

Figure 1 Effects plot of the original experimental trials for birefringence.

control parameters. One is used for a block effect. The remaining seven columns, labelled **a** . . . . **h**, are taken up by interactions, but due to the fractional nature of the factorial design, several interactions are confounded in each of these columns. The plots demonstrate that the individual parameters, MPS and WS, have highly significant effects. Fig. 1 also reveals an interaction effect which appears significant, shown as **b**; but the exact source of this interaction effect in **b** cannot be determined due to confounding. In the duplicate trials, the effect in **b** was less obvious. ANOVA has also shown the significance of MPS, WS and **b**.

To ascertain whether any of the interactions comprising **b** is indeed also significant, the confirmatory trials were carried out, which excluded the processing parameters, QAS and SFS. In the L16 array constructed, all the interactions between pairs of processing parameters can now be separated, and significant ones can be identified. Fig. 2 shows the effects plot for the confirmatory trials. As expected, MPS and WS are seen to be clearly significant. It is apparent too that the birefringence values are also significantly influenced by MPS*WS, i.e. the interaction between the two main control parameters. ANOVA has confirmed the significance of this interaction which was one of the confounding interactions assigned in column **b** in the original and duplicate sets.

Further analysis of the interaction effect was carried out using a response surface plot as shown in Fig. 3, a three-dimensional representation of the response Δn (z dimension) as a function of the various combinations of the interacting main factors MPS and WS (y and x dimensions respectively). The twist of the surface seen

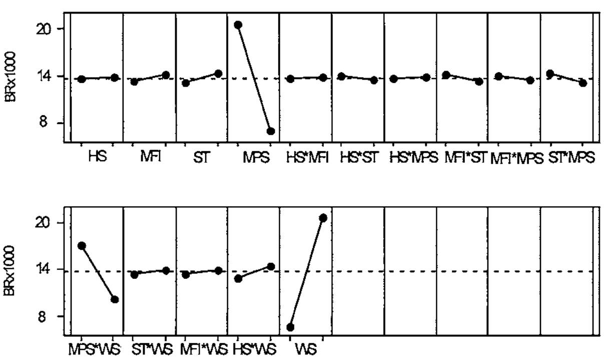

Figure 2 Effects plot of the confirmatory trails for birefringence.

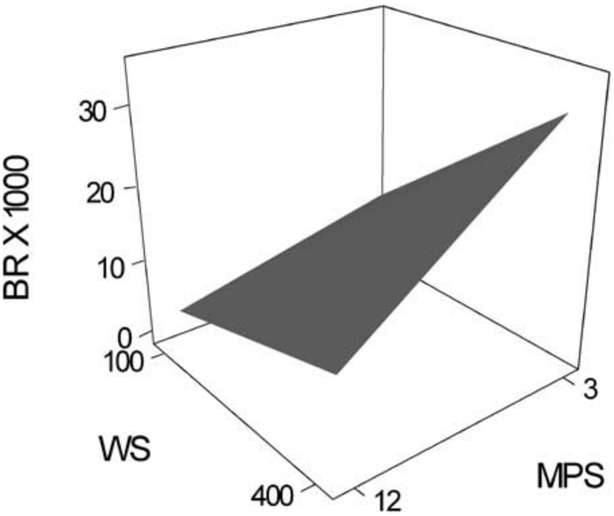

Figure 3 Surface plot of the response birefringence BR as a function of interaction between winding speed WS and metering pump speed MPS.

in the figure confirms the significance of the interaction MPS*WS on Δn. Note that the highest point of the surface is associated with the high level of WS and low level of MPS; conversely, the lowest point is associated with the low level of WS and high level of MPS. These features show that birefringence and hence overall orientation increases when WS is set at a high level and MPS is set at low level, and vice visa.

Based on the conclusions drawn from the screening and confirmatory experiments, a statistical model for optimising birefringence in as-spun fibres has been developed as shown in Table I. The model covers the identified significant main and interaction factors, and specifies the combination of factor levels for enhancing birefringence and thus overall orientation of as-spun PP fibres: the level of the winding speed WS should be set as high as possible and the level of the metering pump speed MPS as low as possible. Conversely, the direction of change in the levels should be reversed if overall orientation is to be reduced.

Both MPS and WS govern the draw-down ratio:

$$\mathrm{DDR} = (\mathrm{k} \cdot \mathrm{WS} \cdot \mathrm{HS})/\mathrm{MPS}$$

The constant, k, allows for the capacity and efficiency of the metering pump. DDR indicates the extent to which the extruded fibre has been lengthened during its passage from the spinneret to the winder. An increase in WS and a decrease in MPS result in an increase in DDR. The results shown in Figs 1 and 2 demonstrate too that increased WS and decreased MPS are also correlated with a rise in birefringence. In addition, the significant

TABLE I Statistical model for optimisation of birefringence of as-spun PP fibres*

Main factors	Interaction	F value	P value	Model
WS		354.72	0.000	WS (H)
MPS		329.81	0.000	MPS (L)
	MPS*WS	81.70	0.000	WS (H), MPS (L)

*The F values and P values are quoted from the confirmatory trials. $F_c\,(1, 9) = 5.12$ at $\alpha = 0.05$ under the conditions.

interaction between MPS and WS indicates that the relative value of these two parameters is also important. The overall orientation of the PP macromolecules is primarily determined by the extent of elongation of the newly formed fibre during draw-down [18]. This conclusion is consistent with the view that the structural order along the fibre axis is brought about primarily by stretching of the fibres. Increased stretching promotes extension and alignment along the fibre axis of the polymeric chains in the non-crystalline regions. Furthermore, we may envisage that the orientation of crystallites is promoted along the fibre axis.

5. Fibre drawing

We have also begun to extend factorial design to fibre drawing. PP fibres are often, in practice, drawn using a multi-stage process. However, the successful production of fibres with the required mechanical properties by a multi-stage process will be strongly dependent on the degree of crystallographic order and polymer chain orientation in the filaments attained during the first stage of the drawing process. We have investigated how the conditions at this drawing stage affect the structure of the first-stage drawn PP filaments.

Fig. 4 shows the effects plots of the first-stage drawing trials for crystallographic order (a) and birefringence (b). The four levels (on the x axis) of TRP1 are 40, 60, 80 and 100 °C, and those of SR2 are 100, 150, 200 and 250 m min^{-1}. It can be seen from Fig. 4a that the degree of crystallographic order increases with temperature in an essentially linear manner between 40 °C and 80 °C, but at a much slower rate at higher temperatures between 80 °C and 100 °C. The drawing speed does not have a significant effect on crystallographic order, as also confirmed by ANOVA. In addition, the data for the crystallographic order of the sixteen samples show that samples drawn at a low temperature 40 °C are in a paracrystalline state, those drawn at a medium temperature 60 °C are in a transitional state and those drawn at high temperature 80 °C and 100 °C are in a crystalline state. Therefore, the degree of crystallographic order of the drawn filaments is determined by the drawing temperature and is not significantly influenced by the degree of drawing. In contrast, the degree of overall orientation is, as expected, dependent upon the degree of drawing and is not affected significantly by drawing temperature in the range studied, as can be seen clearly from Fig. 4b.

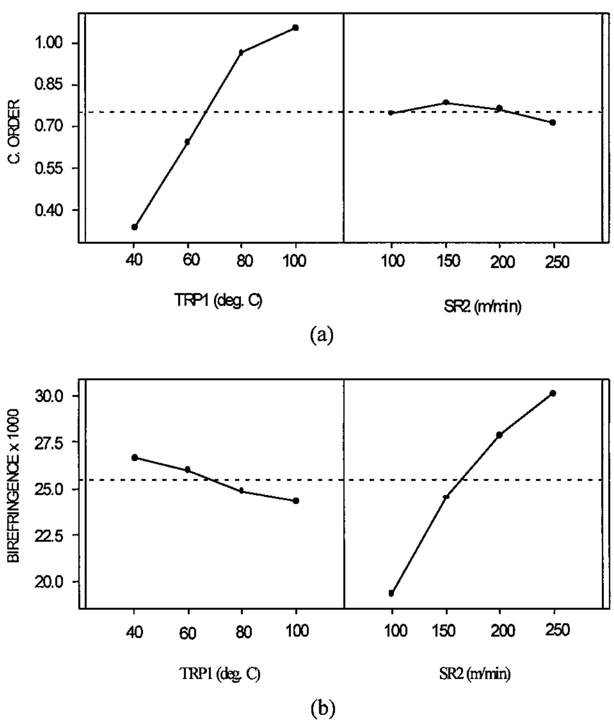

Figure 4 Effects plots for crystallographic order (a) and birefringence (b) of first-stage drawing.

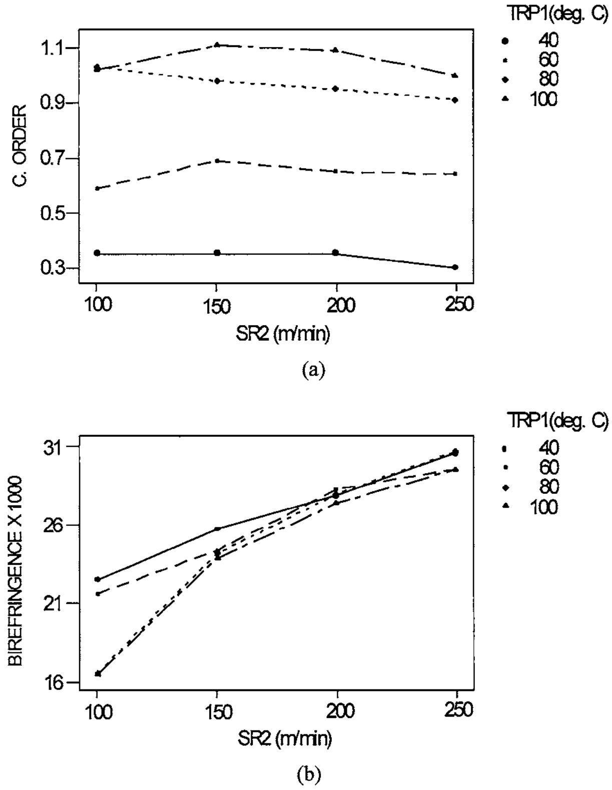

Figure 5 Interaction plots for crystallographic order (a) and Birefringence (b) for one-stage drawing.

Fig. 5 shows the interaction effect plots for crystallographic order and birefringence. The four lines in Fig. 5a for crystallographic order are almost parallel to each other in the region between SR2 levels 2 and 4, indicating that there was essentially no interaction between the two factors TRP1 and SR2 in this speed range. Between SR2 levels 1 and 2, on the other hand, some of the lines intersected each other at appreciable angles, indicating possible interaction in this range. Similar phenomena are observed in Fig. 5b for birefringence. That is, interaction between TRP1 and SR2 was suggested when SR2 is at low levels, but not at high levels. Definite confirmation of the interaction requires further experiments.

6. Conclusions

Factorial experimental design is a valuable approach for assisting the identification of those control parameters in melt-extrusion and drawing equipment which significantly affect the structures of as-spun PP fibres and drawn fibres, respectively. We are also adopting the approach to highlight those control parameters significantly influencing PP fibre mechanical properties.

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