Orientation effects during the flow of short-fibre reinforced thermoplastics

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A study has been carried out to investigate molecular orientation in injection moulded bars of short glass fibre reinforced polypropylene and polyethylene. For the range of fibre concentrations encountered commercially, the fibres do not appear to have any direct effect on the matrix orientation. As the fibre concentration increases, however, the matrix orientation becomes dominated by the orientation of the fibres. These effects are interpreted in terms of current ideas of the rheology of polymer melts during injection moulding and the crystallization of polymers at fibre surfaces.

INTRODUCTION

During the last few years there has been a rapid increase in the engineering applications of filled thermoplastics. The presence of a filler can often lead to significant improvements in the load bearing capabilities of the thermoplastic, often at a lower cost than that of the unfilled material. The range of fillers available is extensive and range from cheap mineral fillers, such as talc, to the high stiffness fibres, such as carbon. In the case where the filler used is a fibre, it is well established that the moulding process used to form the component can produce complex patterns of fibre orientation. There have been a number of studies carried out to relate processing conditions and fibre orientation and also to relate fibre orientation to the anisotropy of the physical properties, especially stiffness. A considerable amount of this effort has been devoted to short glass fibre reinforced thermoplastics *sfrtp*[1–5]. So far, the approach has been to interpret mechanical anisotropy data of *sfrtp* mouldings in terms of the orientation of the fibres. This is a natural progression from the established work concerned with the mechanics of traditional long fibre composites. However, current theories of fibre composite materials usually assume that both the fibre and matrix phases are isotropic. The application of such theories to conventional composites, most of which are based on thermosetting resins, is justified since matrix orientation is expected to be very small. However, it is not obvious, *a priori*, that such is the case where a thermoplastic is being used as the matrix phase. Indeed, a number of disparate studies suggest that this assumption is not justified, especially when reasonably large volume fractions of fibres are employed.

When a fibre filled crystallizable polymer is injection moulded and subsequently allowed to cool, it is reasonable to expect that a residual molecular orientation will exist, which could arise from a number of sources.

Firstly, it is well established that during injection moulding of unfilled thermoplastics a complex variation of orientation and crystal morphology occurs throughout the moulding. This is due to a combination of high shear and elongational flow rates occurring in the cavity under non-isothermal conditions. Detailed morphological studies have been reported for a number of polymers including polypropylene[6–8], polyacetal[9,10] and poly-4-methylpentene-1[11]. In particular, for polypropylene[6] a number of morphologically distinct layers could be identified through the moulding thickness by means of polarized light and X-ray diffraction. There appears to be a thin surface layer approximately 15μ thick that is largely unaffected by processing conditions. Because of its thinness, precision is needed to observe this layer in polypropylene and other polymers[12]. There is a thicker layer which is highly birefringent and whose thickness is markedly dependent on processing conditions; e.g. as the injection time increases, the layer thickness increases. This layer is probably the material that solidifies during mould filling. Following this there is a birefringent region often referred to as the 'shear zone', which is associated with the region of high shear in the hot core material. Finally, in the centre of the moulding the material is unoriented due to the fact that the molecules there have considerable time to relax before the material solidifies. Variation in processing conditions lead to changes in the relative thicknesses of these various layers, which are reflected in the anisotropy of the physical properties of the moulding, e.g. impact strength and Young's modulus. In the case of fibre filled polymers, similar variations in morphology can be anticipated especially when the fibre volume fraction is low, such that the fibres are separated by distances which are large compared to their diameter.

A second contribution to molecular orientation in fibre filled thermoplastics arises from the direct influence of the fibres on the morphology of the surrounding matrix. This occurs even in the absence of relative movement between the fibre and the matrix. Studies of the spherulitic growth of nylon 6 around various types of fibre have been reported by Bessell *et al.*[13,14]. Observations were made of the crystallization processes around both single fibres and in composites containing 15% volume fraction of uniaxially aligned fibres. In both cases the matrix was prepared by the *in situ* anionic polymerization of caprolactam. It was found that columnar spherulitic growth occurred around the fibres, the extent of this depending on the type of fibre and its surface treatment. Of considerable significance in the context of the present paper, is the

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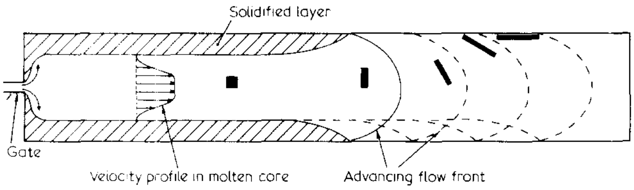

Figure 1 Schematic diagram of the mould filling process showing the deformation of an initially square fluid element at successive positions of the advancing flow front

observation that in the case of the composites containing 15% volume fraction of fibres, the columnar crystalline regions around adjacent fibres tend to impinge on each other thereby modifying the matrix material overall. Associated with this is a high degree of chain alignment parallel to the fibre axis, sufficient, in fact, that on stressing the composite to fracture along the fibre axis, fibre pull-out occurs but with a sheath of matrix material adhering to the fibres. Clearly, these effects should feature prominently in any fibre filled thermoplastic and should become progressively more significant as the volume fraction of fibres increases. If an indirect measure of matrix orientation is to be used experimentally (e.g. birefringence) notice has to be taken of the possible contribution to the birefringence arising from the inhomogeneous stress field around the fibres, especially the stress developed as a result of the different thermal expansion coefficients of the fibre and matrix. If the effect is important, it will develop after the composite has cooled to below its melting point since below this temperature, gross molecular relaxation will not occur during the time scale of the cooling process.

Finally, during the flow of a filled thermoplastic relative movement between the filler and matrix occurs[15,16]. This will result in a modification to the local flow field and hence molecular conformation of the matrix which may not relax entirely before cooling occurs.

The effects discussed can all contribute to the final state of molecular orientation in *sfrtp*. Indeed there is strong justification for examining each of these contributions independently and more work is required in this area. In this paper, however, we seek to examine whether molecular orientation is significant in *sfrtp* using birefringence techniques and whether there is any interaction between the fibres and matrix in establishing the final pattern of molecular orientation during injection moulding. The experimental work described in this paper was mainly carried out in the Department of Materials at Cranfield Institute of Technology. Subsidiary experimental work and further analysis of the results was conducted later in the Department of Non-Metallic Materials, Brunel University.

Rheology of polymer melts during mould filling

Injection moulding imposes a complex cycle of temperature, pressure and flow rate changes on the thermoplastic under consideration. The problem is so involved that a detailed quantitative analysis of the entire process is impossible at this time although parts of the process have been analysed theoretically. However, some general features associated with the flow of a polymer melt into a relatively cold mould cavity seem to be well established and serve to provide an indication of the flow processes occurring. One of the most informative treatments of mould filling has been reported by Tadmor[17]. It is interesting to examine the application of some of the ideas discussed in this and other papers to our particular mould geometry.

With reference to *Figure* 1, it can be seen that on entering the cavity, the fluid will initially be subjected to compressional flow leading to a divergence of the stream lines away from the gate. Fluid elements will be oriented orthogonally to the axis of the mould. Simultaneously, a solidified layer of polymer forms on the mould surface. A flow front establishes itself, advancing forward by the flow of the molten polymer through a channel (often referred to as the 'core' region) defined by the boundaries of the solidified polymer (often referred to as the 'skin' layer). The original compressional flow field in the melt at the gate is gradually replaced by a velocity profile shown schematically in *Figure* 1 and corresponding to the non-isothermal flow of a non-Newtonian fluid. The detailed shape of this velocity profile depends on the rheological characteristics of the fluid, the flow rate and the temperature of the incoming melt as well as the temperature of the mould cavity. To a first approximation, at least, this part of the flow process can be regarded as capillary flow. For the special case of radial flow into a disc-shaped mould, Berger and Gogos[18] have used numerical methods to calculate the velocity profiles for various locations of the flow front. Irrespective of the detailed shape of these profiles, their work indicated that a solidified layer soon develops during injection of the melt and that the position of maximum shear rate is not at the boundary between the skin and core but at some distance away, towards the centre line of the moulding. This can be seen schematically in *Figure* 1.

Further towards the flow front, the velocity field becomes considerably more involved. Assistance in understanding the flow process here, has come from allied studies of the movement of the fluid particles at the interface between two fluids, with the flow of one fluid replacing another, as in capillary flow[19,20]. In a Lagrangian frame of reference (i.e. one in which the observer moves with the same velocity as the advancing flow front) the motion of the fluid is similar to that of a 'fountain' with fluid elements decelerating as they approach the flow front from the core and acquiring a radial component of velocity as they move towards the wall. In a laboratory frame of reference, the actual deformation and change in orientation of an initially square fluid element (for two-dimensional flow only) during this process is shown in *Figure* 1. At the flow front, the velocity field is not simply elongational flow parallel to the curved surface of the flow front since relative movement between fluid elements and the front must occur. However, if the contribution of the shear flow at the front is ignored, then as Tadmor[17] has shown, the elongational flow field will result in a high degree of orientation parallel to the axis of the mould when the fluid elements arrive at the wall. The rate of elongation of the fluid elements is dependent on the speed of injection. The precise degree of orientation at the wall is determined by the time taken for a fluid element to travel from the axis of the mould to the wall during which time molecular relaxation is occurring. Hence a high speed of injection results in a large net molecular orientation at the wall.

Therefore, any orientation in the skin and core results from different flow histories. In the case of an unfilled polymer, the competing mechanisms of molecular elo-

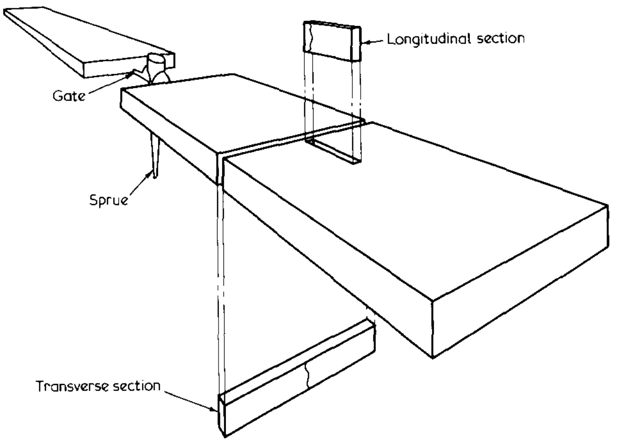

Figure 2 Two cavity strip moulding showing locations of the transverse and longitudinal sections used for birefringence and c.m.r. studies

ngation and relaxation will determine the degree of orientation remaining in the moulding. If short fibres are present and if, for the purposes of this discussion, they can be regarded as separate fluid elements, the fibre orientation distribution in the moulding will serve as a good indication of the general nature of the flow fields existing during the injection moulding process.

EXPERIMENTAL

Moulding equipment

Two moulds were used in this work. One was a two cavity bar mould, shown in *Figure* 2 and having dimensions of 190 mm × 30 mm × 1.5 mm with a semicircular gate of 1 mm radius. As in a previous paper[21], this was chosen to help develop an understanding of the flow behaviour in a simple geometry. One cavity was drilled to take a flush mounted pressure transducer which fed a signal back to an adaptive controller. The injection moulding machine used in conjunction with this mould was a Bipel 130/25 model with a 130 tonf. (~1.3 MN) clamping force and 6.6 oz (~187 g) maximum shot weight. A special feature of this machine was the closed loop control of the injection process using a Bosch SPR 200 adaptive control system which allowed close control of the speed of injection and the pressure in the mould cavity through a servo-hydraulic valve. This ensured good repeatability of mouldings from shot to shot. Where limited quantities of material were available then a smaller but similar bar mould was used. This had four cavities of dimensions 42 mm × 6 mm × 3 mm and was mounted on a Volant Bros. hand operated 1 oz (~28 g) machine.

Moulding conditions

The two bar strip mouldings were produced under two sets of conditions: (1) Fast constant speed injection (injection time = 0.2 s). (2) Slow constant speed injection (injection time = 11 s). Other machine conditions were:

Barrel temperature	240°C on all zones
Screw speed	100 rpm
Back pressure	100 psi (689 KN m^{-2})
Mould temperature	30–35°C

The four bar mouldings were only produced at a relatively fast injection speed (< 5 s) on the hand operated machine. Operating temperatures were:

Barrel temperature	240°C
Mould temperature	35–45°C

Materials

The injection moulding of the two bar strip mould was carried out using I.C.I. Propathene HW 60GR/20/001, a polypropylene containing 20% weight of glass fibres of modal length 600 μm. The four bar strip moulding was produced with filled and unfilled Hoechst Hostalen GM 7255 Pulver, a very high molecular weight polyethylene. This was chosen so that high molecular orientation could be examined in a separate experiment[22]. The filled mouldings contained 20 and 50% weight of short glass fibres. The flow properties of the unfilled high density polyethylene together with 30, 60, 70 and 80% weight of short glass fibres were examined using a capillary rheometer.

Production of the short glass fibre filled high density polyethylene. The 3 mm glass fibres used by I.C.I. in their Propathene grades were used to fill the high density polyethylene. Polymer and glass fibres were first dry blended and then extruded through a Locati Pavesi Plastimac extruder. This extrudate was granulated using a Leesona machine. The glass fibres were found to have a modal length of 700 μm after processing and as with the Propathene grades the distribution was skewed by the presence of long fibres.

Structural examination

Both fibre orientation and molecular orientation in the mouldings were investigated by cutting thin sections, approximately 80 μm thick, using a low speed diamond saw.

Fibre orientation. The examination of fibre orientation in the mouldings was performed using the technique of contact micro-radiography (c.m.r.). The application of this technique to the study of fibre-reinforced thermoplastics has been described by Darlington and McGinley[23]. A thin section is laid onto a fine grain photographic plate and exposed to a beam of X.rays, which casts 'shadows' of the fibres onto the plate.

Molecular orientation. The molecular orientation in the mouldings was studied by measuring the birefringence at successive points across the thin sections. These measurements were made using a Berek-type compensator fitted to a Vickers polarizing microscope. A quartz wedge was used to place the sample in the subtraction position relative to the compensator. For polypropylene and polyethylene this meant that the chain axis of the molecules was perpendicular to the axis of rotation of the compensator so that its rotation in either direction from the zero position gave rise to interference colours of small phase difference up to the point of compensation. The precision with which the points of compensation were established depended upon the accuracy with which the position of subtraction was set.

Although light scattering was considerable in the fibre reinforced samples, this did not prevent the accurate measurement of birefringence. Even at high fibre loadings the results obtained were comparable to those expected for the unfilled polymer.

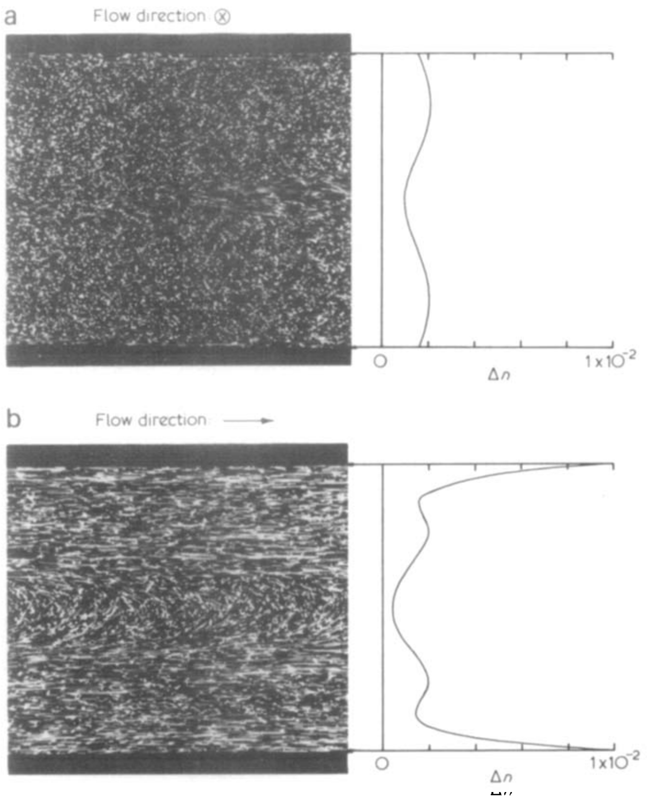

Figure 3 Birefringence variation through the moulding thickness and associated c.m.r. for fast injection; (a) transverse section (b) longitudinal section

Rheological examination

High density polyethylene compositions containing different proportions of short glass fibres were characterized rheologically using a Davenport capillary rheometer. Two 4.0 mm diameter dies of lengths 40 and 100 mm were used to obtain a range of apparent shear rates from 0.2–200 s^{-1}.

RESULTS AND DISCUSSION

Large parallel-sided bar

The description of the fibre orientation distribution through this moulding and its dependence on injection speed has been reported previously[21]. For completeness, however, the c.m.r.'s of the sections chosen for the birefringence studies are included since these current studies confirm some earlier speculations apart from any new informations they provide concerning the molecular orientation. The sections studied are defined in *Figure* 2.

The transverse sections were taken from a point midway between the gate and the end of the moulding. The longitudinal sections were taken from, an adjacent position along the centre line of the moulding. The c.m.r.'s of the transverse and longitudinal sections corresponding to fast and slow injection were reported previously[21] and are shown in *Figures* 3(*a*, *b*) and 4(*a*, *b*). The salient feature is that for fast injection, the fibres are aligned at 90° to the overall flow direction in the core of the moulding, whereas in the case of slow injection, the fibres are aligned parallel to the axis of the strip in the core of the moulding. A possible interpretation of these effects has also been discussed[21]. Two other points relating to these c.m.r.'s merit some comment. Firstly, in the case of slow injection a definite layer exists, some distance from the moulding surface, that is deficient in fibres. The origin of this effect has not been discussed hitherto but the birefringence work here provides a valuable guide to interpretation that would not be possible on the basis of c.m.r. alone. Secondly, if the change in fibre orientation through the moulding thickness is used to define the proportion of the skin and core material it would appear that the skin thickness for fast injection is considerably greater than that for slow injection. This is unrealistic. Again the anomaly is resolved using the birefringence data.

Fast injection. Figures 3 (*a*, *b*) show that for fast injection the molecular orientation is large close to the surfaces of the moulding and predominantly along the bar axis. Peaks in the Δn variation occur further away from the moulding surface and this is very characteristic of the variation of Δn through the thickness of mouldings of unfilled polymers. The high value of Δn at the surface is confined to a thickness of $\sim 30\mu$ and because of the thinness of the layer, it often escapes detection during an initial examination of a moulding. This point was raised by Janeschitz-Kriegl[12] and it is encouraging that the thickness of the layer observed in our experiments is of the same order as that previously reported by him for unfilled polystyrene. This layer is attributed to the elongational flow occurring at the advancing flow front, as described in the section dealing with rheology of polymer melts during mould filling. The fact that the speed of injection has a marked effect on the Δn variation near the moulding surface suggests that this explanation is correct, at least qualitatively. The peaks in Δn away from the surface are attributed to molecular orientation induced by shear flow close to the boundary between the skin and core material.

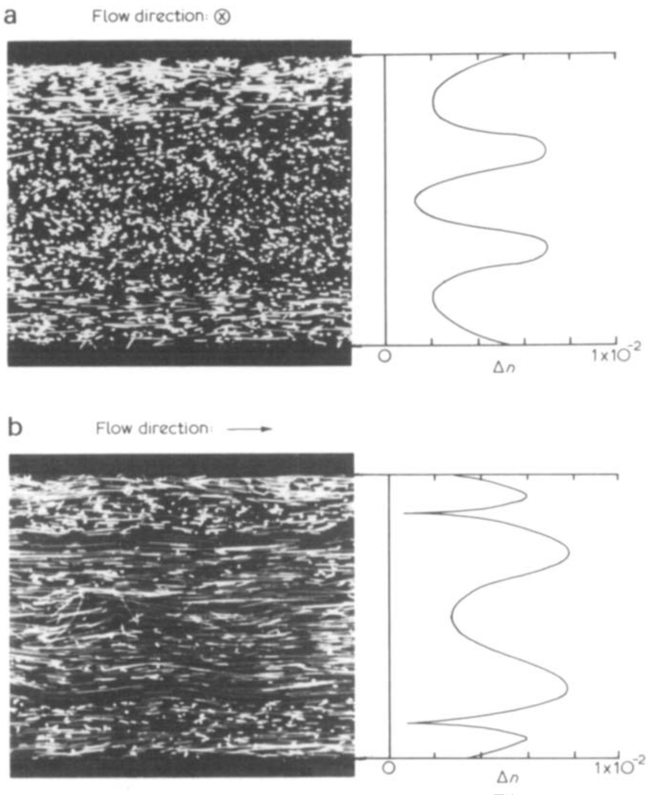

Figure 4 Birefringence variation through the moulding thickness and associated c.m.r. for slow injection; (a) transverse section (b) longitudinal section

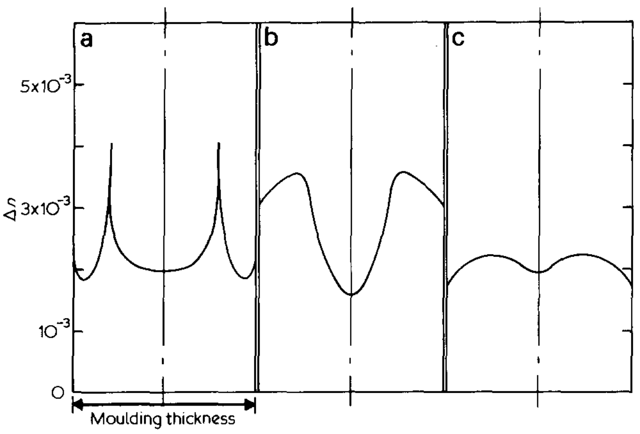

Figure 5 Birefringence variation through the thickness of the small bar moulding: (a) unfilled polyethylene, (b) polyethylene containing 20% wt of glass fibres, (c) polyethylene containing 50% wt of glass fibres

It is interesting that these peaks occur at a position where there is no real obvious change in fibre orientation, as judged from the c.m.r. A small but definite molecular orientation exists in the core of the moulding. The shear rate on the axis of the moulding will be zero and hence the molecules will not be oriented. Even slightly away from the axis where the shear rate is non-zero, little orientation is expected since this material takes the longest time to solidify after moulding and the molecules therefore have considerable time to relax. In mouldings of unfilled semi-crystalline polymers, large spherulites are observed in the centre of the moulding and the overall molecular orientation is zero. One explanation for the core orientation in our case could be that the major contribution to this comes not from flow induced molecular alignment but from columnar spherulitic growth that is known to occur around fibres in the absence of an externally applied flow field. Support for this supposition comes from the birefringence variation resulting from slow injection.

Slow injection. Figures 4(*a*, *b*) show the c.m.r. and corresponding birefringence variation through the moulding thickness for slow injection. The birefringence variation for the longitudinal section is more complicated compared to the case of fast injection insofar as there is an additional peak in Δn close to the moulding surface with the absolute value of Δn on the moulding surface being considerably lower compared to that for fast injection. Even the transverse section shows a much more dramatic variation in Δn which implies that the molecules although highly oriented are not uniformly aligned parallel to the principal flow direction as they were for fast injection. The peaks in Δn closest to the axis of the moulding are attributed to the molecular orientation induced by shear flow at the boundary between the skin and core. These peaks coincide closely with the region of the moulding that is deficient in fibres. This would suggest, as originally suspected, that this layer is generated during injection rather than during the subsequent holding pressure period. It is well known, that the motion of fibres during shear flow is perturbed in the vicinity of a wall and the restriction of the fibre orbit produces an excluded volume which manifests itself as a layer deficient in fibres. It is also interesting that a non-zero value of Δn is found on the moulding axis, as in the case of fast injection, but that the value for the slow injection case is larger. This again suggests that the fibres themselves are influencing the surrounding matrix, especially as the fibres are highly aligned along the flow direction in the core.

The additional peak in Δn close to the moulding surface demands interpretation and we would like to propose the following mechanism. In the case of fast injection, molecules lying on or close to the moulding axis are subjected to a zero shear field before their arrival at the flow front. They are then subjected to a predominantly elongational flow field which becomes increasingly more important as the rate of injection is increased. As the rate of injection is decreased the alignment of the molecules due to the elongational flow field decreases (as shown by the smaller value of Δn on the surface of the moulding for slow injection). Associated with this is a change in the velocity profile of the melt in the core of the moulding from essentially plug flow to pseudo-parabolic. This implies that molecules close to the axis will now be subjected to shear flow before arriving at the flow front. It would appear from our work that the particular conditions of slow injection that we use favour the shear flow rather than the elongational flow at the advancing front in effecting molecular alignment some distance from the moulding surface. Confirmation or otherwise of this argument must await a detailed examination of the velocity field close to an advancing melt front.

Small parallel-sided bar

The geometry of this small bar was very similar to that utilized in the work already described. It was chosen for the study of the effect of fibre volume fraction on the pattern of birefringence variation across the moulding since the polymer–fibre mixtures had to be prepared on a laboratory scale and only a limited amount of material was available for injection moulding.

In these experiments, since the estimated speed of injection was between 3 and 5 s, the fibre orientation in the core of the moulding was intermediate between that corresponding to fast and slow injection as described in the section on fast injection. This means that the fibres were not aligned precisely at 90° to the principal flow direction but had a component of orientation parallel to the flow direction. The fibre orientation in the skin layers was in all respects similar to that shown in *Figures* 3 and 4. The variation of birefringence Δn across the moulding thickness for longitudinal sections is shown in *Figure* 5 for three different values of fibre fraction. The variation of Δn for the unfilled polymer is typical of that observed for other polymers. The peaks in Δn associated with the shearing of the melt at the skin–core boundary are sharp but become less distinct and broader as the fibre fraction increases. The interpretation of this requires a knowledge of the rheological properties of the polyethylene having a variety of fibre volume fractions. Apparent viscosity versus shear rate data were obtained using a Davenport capillary rheometer following the procedures and recommendations for data analysis described by Crowson, Folkes and Bright[16]. These data are shown in *Figure* 6. It can be seen that for any given shear rate, the slope of the viscosity versus shear rate curve increases as the concentration of fibres increases. This corresponds to a progressive change in velocity profile of the melt from pseudo-parabolic for zero fibre concentration to almost complete plug flow at high concentrations. The implication of this for the Δn variation in the mouldings is that one would expect that as the fibre concentration increases,

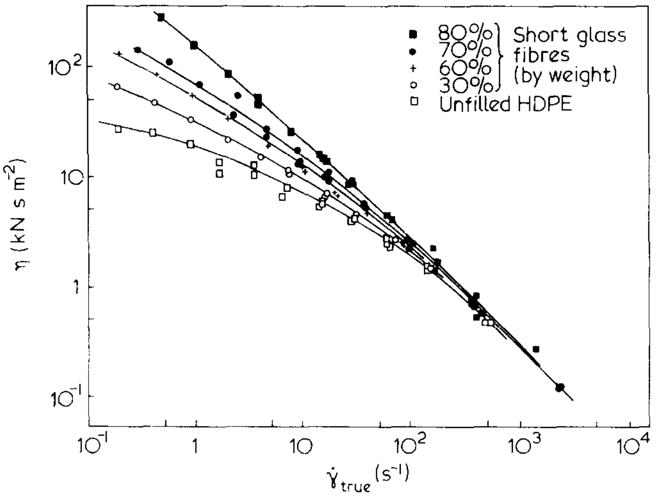

Figure 6 Variation of apparent shear viscosity with shear rate for polyethylene containing various concentrations of short glass fibres

the region between the skin and core over which an appreciable shear rate exists would become narrower and hence the associated peaks in Δn should also become much narrower. This is contrary to that observed experimentally (see *Figure* 5) and would seem to suggest that the molecular orientation which is responsible for the observed variation in Δn is not directly related to the flow processes that have occurred during injection moulding. In fact, the variation in Δn at large fibre fractions can be explained qualitatively by assuming that there is no orientation resulting from flow but that it is due entirely to the columnar spherulitic growth of the matrix around the fibres that would occur in the case of a stationary melt. In this case the pattern of variation of Δn in our longitudinal sections would be related directly to the degree of fibre orientation along the principal flow direction. Thus the maximum value of Δn obtainable would correspond to fully aligned fibres, while fibres aligned at 90° to the principal flow direction would result in optical isotropy for longitudinal sections. In the case of our injection moulded bars, a detailed examination of the variation in fibre orientation through the bar thickness shows that the surface layers contain fibres arranged randomly in a plane. Beneath this layer the fibres are partially oriented along the principal flow direction, while in the core they are partially oriented at 90° to the flow direction. This variation in fibre orientation distribution from skin through to the core would account for the observed maxima in Δn, shown in *Figure* 5(*c*), and also for their relative width compared to the case of the unfilled polymer. At least qualitatively, this model would appear to be in accord with our experimental data. For it to be valid we require that little, if any, molecular orientation be induced in the matrix during the short time of injection of the melt into the cavity. Other measurements indicate that close to the fibres, molecular reorganization during flow is inhibited leading to excessively long relaxation times for the molecular network. The effect of this on the matrix as a whole will become progressively more important as the concentration of fibres increases.

Interpretation of birefringence in fibre filled thermoplastics

Justification is needed for using birefringence as a method of assessing molecular orientation in short fibre filled thermoplastics. Unexpectedly pronounced variations in birefringence through the thickness of our mouldings could be detected even in the presence of high concentrations of fibres. Although extensive light scattering was observed, it does not appear to have been excessive enough to depolarize the incident plane polarized light even in the samples containing a high concentration of fibres. However, we are aware of the possibility that some depolarization may be occurring, in which case our birefringence measurements will underestimate the degree of molecular orientation in the matrix. Alternative techniques, such as wide angle X-ray diffraction, also have inherent limitations when applied to this type of material. Birefringence measurements are straightforward and give additional insight at least into the orientation processes occurring in the material during injection, which could not be inferred from a study of fibre orientation alone.

In a fibre filled thermoplastic molecular orientation can also result from the difference in thermal expansion coefficient between the fibre and matrix. In our case, during cooling from the processing temperature, the matrix will contract more than the glass fibre and this will generate a tensile stress parallel to the fibre axis which can result in molecular orientation. An estimate of the relative strain can be made and this gives a value of 2.25%. If molecular relaxation was totally absent, this would result in a value of 4.2×10^{-4} for the birefringence of polypropylene. Compared to the average values of birefringence that we measure in our mouldings, this value is 15–20 times smaller, so that although it is not insignificant, this contribution is small enough for us to ignore at this stage in the work.

CONCLUSION

Birefringence measurements in short glass fibre filled thermoplastics can be used to give additional information on the flow processes occurring during injection moulding. For the range of fibre concentrations normally encountered in commercially available grades of polypropylene (20–30% of fibres by weight), it appears that the presence of the fibres is having little direct effect on the molecular orientation in the matrix. However, as the concentration of fibres increases, the molecular orientation in the matrix becomes dominated by the orientation of the fibres. This is consistent with current knowledge of the effect of the fibre surface on the surrounding matrix morphology, deduced from experiments involving stationary melts. For all concentrations of fibres, we observe molecular orientation in the matrix of a magnitude comparable to that observed in unfilled polymers. The implications of this for the interpretation of the anisotropy of the physical properties in the mouldings is self evident and indicates, in particular, that a rigorous interpretation of the mechanical anisotropy will require composite theories which allow for the matrix phase being anisotropic.

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