# Structure and mechanical properties in injection moulded discs of glass fibre reinforced polypropylene\*

#### M. W. Darlington, B. K. Gladwell and G. R. Smith

Department of Materials, Cranfield Institute of Technology, Cranfield, Bedford MK43 OAL, UK (Received 15th June, 1977)

Edge-gated discs of glass fibre reinforced polypropylene have been produced over a range of injection moulding conditions. Examination of the structure of the discs has shown that injection moulding conditions which resulted in retained fibre length in the mouldings also produced serious fibre agglomeration and increased voiding. However, despite this agglomeration and voiding, average mechanical properties were still improved for the disc with retained fibre length. Fibre orientation distribution showed little variation over the range of moulding conditions.

## INTRODUCTION

As a first stage in a general study of the effects of processing conditions on mechanical properties, a series of edge-gated disc injection mouldings of glass fibre reinforced polypropylene (GFPP) has been produced using three different sets of moulding conditions: (A) very low screw back pressure, large gate; (B) high back pressure, large gate; (C) high back pressure, small gate size.

Earlier work<sup>1,2</sup> has suggested that the principal parameter affected by such variations in injection moulding conditions would be the fibre length. Hence the stiffness, strength and anisotropy of properties should vary between the three sets of GFPP discs used in this work. However, additional factors such as fibre dispersion, fibre orientation and void content may be affected by injection moulding conditions and perhaps override the benefits that might result from retention of fibre lengths in the moulded product.

In order to gain greater insight into the effects of the above mould process variables, a detailed structural study has been conducted on the GFPP discs. The fibre volume fraction  $(V_F)$ , fibre length distribution (FLD) and fibre orientation distribution (FOD) have been quantitatively determined for each set of discs. Qualitative assessment of fibre dispersion and voiding in the composites has also been undertaken.

Although not the prime concern of this paper, a theory found useful in other work<sup>3,4</sup> to predict the stiffness of short fibre reinforced composites has been utilized here to give further insight into the observed mechanical behaviour of the GFPP discs. With this modified rule of mixtures approach to composite stiffness:

$$E_{c} = \eta_{0} \eta_{L} E_{F} V_{F} + E_{m} (1 - V_{F})$$
(1)

where  $E_c$ ,  $E_F$  and  $E_m$  are the composite, fibre and matrix Young's moduli respectively,  $\eta_0$  is an orientation efficiency factor (due to Krenchel<sup>5</sup>) and  $\eta_L$  is Cox's fibre length efficiency factor<sup>6</sup>.

# MATERIAL AND SAMPLE PREPARATION

The material used in this study was glass fibre reinforced polypropylene (GFPP) produced by Imperial Chemical Industries Ltd, Plastics Division in granule form and containing nominally 25% of coupled short glass fibres (ICI 'Propathene' grade PXC 5563, later redesignated HW 70 GR). The corresponding grade of non-reinforced polypropylene was also examined.

The material was injection moulded in an instrumented 'Ankerwerke' screw injection moulding machine by the Explosives Research and Development Establishment, Waltham Abbey, UK, into edge-gated discs of thickness 6 mm and diameter 100 mm. The asymmetric gating arrangement and other details of the mould geometry used have been described elsewhere<sup>7</sup>. Details of the rectangular gate dimensions and back pressure settings used for each set of discs are given in *Table 1*. All other independent injection moulding parameters were held constant. The conditions chosen to mould disc A were those known to produce little fibre breakage during injection moulding<sup>1,2</sup>, while the moulding conditions chosen for discs B and C were chosen to produce more serious decreases in fibre length. Fibre contents for each type of disc moulding were measured by standard burnoff techniques and the results are included in *Table 1*. Dif-

 
 Table 1
 Injection moulding conditions and fibre concentrations of the GFPP edge-gated discs

Disc	Gate size	Back pressure (MN/m <sup>2</sup> )	Fibre content (wt %)
A	7 mm X 2 mm	0	25.8
В	7 mm X 2 mm	2.76	30.6
С	4 mm X 1 mm	2.76	30.5

<sup>\*</sup> Presented at the conference 'Processing Structure, Properties and Performance of Polymers', University of Nottingham, UK, July 1977



Figure 1 Location of creep and strength test specimens

Table 2 Experimental low strain isochronous modulus data for the GFPP edge-gated discs

	Tensile m		
Disc	 0°	90°	Anisotropy ratio
A	2.55 (±0.06)	3.92 (±0.1)	1.54
В	2.95 (±0.04)	4.18 (±0.07)	1.42
с	2.87	4.18	1.46
C (core)	2.12	4.53	2.14
Unfilled PP	1.53 (±0.01)	1.50 (±0.01)	1.02

A spread of results is indicated for data which have been averaged from more than one specimen

ferences between the fibre contents of disc A and discs B and C are thought to be the result of batch variations rather than of any effects due to injection moulding procedures.

## MECHANICAL PROPERTIES

Specimens for both creep and constant elongation rate tests were cut from the GFPP discs as shown in *Figure 1*. The 03  $(0^{\circ})$  and 02  $(90^{\circ})$  axes were constructed on the discs and then two specimens were taken from either the 02 or 03 direction for any one disc, symmetrically about one of the constructed axes.

The creep response of the GFPP (and corresponding unfilled PP) specimens was monitored by means of highly accurate creep apparatus<sup>8</sup>. Creep tests were conducted using procedures based on those given in ref 9. Table 2 presents isochronous creep modulus values at the low 100 sec strain of 0.2% for the specimens cut at 0° and 90° from the GFPP discs ('0° and 90° discs') and for non-reinforced polypropylene (unfilled PP). Additionally, similar stiffness data are presented for specimens machined from the central 3 mm core of disc C.

Long term creep response of the 0° and 90° GFPP disc specimens has also been monitored. Creep stresses have been chosen to produce 100 sec creep strains in the range  $(0.80 \pm 0.03)\%$  and these creep data are presented as a plot of compliance against log (time) in *Figure 2* for the three disc types.

In addition to these stiffness results, strength data for  $0^{\circ}$  and 90° specimens from each disc type have been obtained on an Instron type TT – C test machine. A constant elongation rate of  $3 \times 10^{-4}$  mm/sec was used and four specimens of each disc type and specimen orientation were tested. These results are presented in *Table 3*. The similarity of the stiffness and strength data of *Tables* 2 and 3 and *Figure* 2 for the three GFPP disc types is quite surprising when the wide range of processing conditions employed is recalled. However, it is of interest to note:

(i) that the GFPP disc processed to contain the longest fibre lengths (disc A), is in fact less stiff then either of discs B or C which were processed to contain shorter fibre lengths. However, weighting these results using a theoretical basis, a value of  $4.5 \text{ GN/m}^2$  (disc 90°) would be expected at a fibre concentration of 30.5% by wt for disc A, and this value is slightly above the values for discs B and C (as expected). On a similar basis, the tensile strengths of disc A specimens at equal fibre concentration would be above the values for discs B and C in *Table 3*.

(ii) that any differences in the compliance *versus* log (time) creep plots for the three disc types in *Figure 2* correspond to differences in the low strain stiffnesses of the discs (in *Table 2*). Furthermore, it is notable that there is a very large increase in disc anisotropy with creep time for the discs.

These points will be further discussed with the presentation of structural data for the 3 GFPP discs below.

## STRUCTURAL STUDIES

As the basis of this work has been to assess the effects of



*Figure 2* Creep compliance plotted against log time for the creep in air at 23°C of GFPP edge-gated disc specimens

Disc A			Disc B		Disc C	
0°	90°	0°	90°	0°	90°	

 $^{\dagger}\sigma = 17.00 \odot \sigma = 25.60 \quad \sigma = 19.50 \ \Box \ \sigma = 26.40 \quad \sigma = 20.66 \times \sigma = 28.00$  $\sigma = 17.84 \ \bigtriangleup \sigma = 27.52 \quad \sigma = 19.50 + \sigma = 27.57$ 

Creep stresses in MN/m<sup>2</sup>

Table 3 Tensile strength data for the GFPP edge-gated discs

Tensile strength (MN/m <sup>2</sup> )		
	90°	
33.2 (±0.5)	46 (± 4)	
36.6 (±0.4)	45 (± 1)	
34.2 (± 0.2)	46 (± 1)	
	Tensile strengt 0° 33.2 (±0.5) 36.6 (±0.4) 34.2 (± 0.2)	



Figure 3 Fibre orientation and length distributions, specimen sites. A and B, slices perpendicular to the plane of the disc; C, slices in the plane of the disc



*Figure 4* Fibre length distributions of the GFPP edge-gated discs (plotted as smoothed curves from normalized histograms). A, disc A; B, disc B; C, disc C

Disc	Mode fibre length (μm)	Mean (number-average) fibre length (μm)
A	350	720
В	350	360
С	350	400

altering processing conditions on the fibre length distributions in GFPP disc mouldings, quantitative assessment of the *FLD* in these mouldings has been undertaken. Detailed 'burn-off' experiments, pyrolysing the polypropylene matrix to leave clean matrix-free fibres, have been performed on sections cut from the surface, intermediate and core layers of each disc type (see *Figure 3*). The weight fraction and distribution of lengths of the burnt-off fibres were measured for each section. Within the experimental errors involved, no trend in the *FLD* (or weight fraction of fibres) was observed *through the moulding thickness* for each disc type. However, the accuracy of this *FLD* determination suffers with such wide *FLD*'s as occur in these discs (particularly disc A) and thus it is difficult to be conclusive about *FLD* variations through thickness.

Figure 4 summarizes FLD data for full through-thickness sections from each disc type. Disc A exhibits a very broad

distribution of fibres, with a significantly higher numberaverage fibre length than the narrower FLD's of discs B and C. This order of FLD's supports the order of stiffness data for the discs shown in *Table 2* when allowance for fibre concentration differences is taken into account. However, this FLD evidence must not be viewed in isolation. For example, varying the processing conditions may also lead to differences in fibre dispersion, fibre orientation and voiding.

A contact microradiographic (c.m.r.) technique has been employed to examine fibre dispersion and orientation in the discs. Details of this technique have been presented elsewhere<sup>10,11</sup>. The sites of slices examined are shown in *Figure* 3. Photographs of through thickness slices viewed in the 0° and 90° directions illustrate that good dispersion of fibres occurs in discs B and C, but that massive fibre agglomeration occurs in the central core of disc A (*Figures 5* and 6). In all three discs there is significant out of plane fibre alignment in the core region and this is difficult to characterize, particularly in the heavily agglomerated core of disc A. However, the surface layers are well oriented in the disc plane, and the predominant fibre orientation plane (despite the out of plane core fibres) does remain the plane of the disc (the 2–3 plane).

For discs B and C slices have been cut parallel to the surfaces of the discs (as shown in *Figure 3*) and fibre orientation distributions in the 2–3 plane have been measured from c.m.r. photographs for each slice. This procedure has not been attempted for the heavily agglomerated disc A. These distributions for slices parallel to the surface have been summed (as described elsewhere<sup>11</sup>), to produce histograms of the projected through-thickness *FOD* in the 2–3 plane for discs B and C in *Figure 7*.

Figure 7 indicates significant fibre alignment in the 02  $(90^{\circ})$  direction in agreement with the anisotropy of mechanical data of *Tables 2* and 3 and *Figure 2*. However, the orientation distribution of disc B is offset to the flow axis. With the mould geometry employed for this work<sup>7</sup>, the feed to the gate in the injection moulding is at an angle, thus an off-set *FOD* may occur in the mould. For disc C, the use of a small gate has overcome this lead-in effect and the resulting *FOD* is symmetric to the flow axis.

Although it is not possible to quantify the FOD of disc A, inspection of c.m.r. photographs suggests it is similar to the FOD of disc B (although with a much higher local variation due to fibre agglomeration). This FOD similarity might be expected as both discs have the same gate size.

While the total through-thickness FOD's in Figure 7 give a good indication of the overall disc anisotropy, these results mask the changes in fibre orientation that occur in layers through the disc thickness. Thus in Figure 8, Krenchel's<sup>5</sup> fibre orientation parameter,  $\eta_0$ , has been plotted against the depth below the disc surface, using the FOD's determined for each slice. Figure 8 shows that discs B and C are very similar in the fibre orientation of each layer; the only differences due to gate size being to angle the flow for disc B (as shown in Figure 7) and perhaps to produce subtle differences at the skin. The surface layers of discs B and C have fibres oriented randomly in the plane of the disc, whereas in the core, fibres tend to be aligned transverse to the major flow direction<sup>11</sup>. Basically there is little difference in either the FLD or FOD of discs B and C and therefore it is not surprising that the mechanical properties of these two disc types are very similar. The slight asymmetry in the FOD of disc B has had little effect on mechanical properties.

Observation of a cross-section through thickness for each of these discs shows the occurrence of a central white layer,



Figure 5 Contact microradiographs of through thickness sections in the  $0^{\circ}$  (a) or  $90^{\circ}$  (b) direction from GFPP disc A

Figure 6 Contact microradiographs of through thickness sections in the 0° (a) or 90° (b) direction from GFPP disc B. (c.m.r. results for disc C were very similar to those shown above for disc B)



*Figure 7* Projected through thickness fibre orientation distributions in the 2–3 plane for GFPP discs B (– – – –) and C (– – –). Fibre orientation efficiency factor,  $\eta_0$ :

Disc	0°	90°
B	0.24	0.52
C	0.23	0.55



Figure 8 Krenchel's tibre orientation efficiency factor,  $\eta_0$ , plotted against the depth through thickness for GFPP discs B and C. Disc B:  $\blacktriangle$ , 90°;  $\bullet$ , 0°, large gate, high back pressure; Disc C:  $\triangle$ , 90°;  $\bigcirc$ , 0°, small gate, high back pressure

the incidence of which has previously been related to the occurrence of voiding<sup>12</sup>. This white layer appears most pronounced in disc A. Measurements of the fibre content and density for both the surface and core layers of the discs show no discernable differences for disc C. However, for disc A, whilst the fibre content of the core and skin layers is the same  $(25.8 \pm 0.1)\%$ , the core is 3% less dense than the surface layers. These small density differences may be attributed to voiding in the disc core.

#### PREDICTION OF STIFFNESS

If the 3-dimensional FOD in the discs is approximated by the projected FOD in the 2-3 plane (the plane of the disc), then the use of equation (1) with the full FLD and this FOD data provides predictions of stiffness for discs B and C. These values are presented in Table 4. In view of the assumptions made in this prediction, there is surprisingly good agreement with the experimental stiffness values of Table 2. Allowance for out of plane fibre alignment in the core of the discs would significantly reduce the stiffness in the 90° direction (E<sub>90</sub>)

# Injection moulded glass fibre polypropylene: M. W. Darlington et al.

but have little affect on  $E_0$ , thereby further improving the agreement between experiment and theory for discs B and C.

A calculation of the stiffness of the core of disc C is also included in *Table 4*. Comparison with experimental data in *Table 2* shows that the predicted values of stiffness for the core are significantly higher than experiment. These high predicted stiffness values are obviously a consequence of neglecting the effect of out of plane fibre alignment which is most significant in the disc core.

While it is not possible to measure an FOD experimentally for disc A, a prediction of the stiffness of disc A can be made if the FOD of disc A is assumed to be similar to disc B (both discs with the large gate size). The effects of the long fibre lengths of disc A can then be determined. These predicted values for disc A have been calculated for fibre contents of 25 and 30% by wt and are also shown in *Table 4*. The disc A prediction at 25% fibre content provides a comparison with the experimental data for disc A in *Table 2* and shows the same order of predictive ability as was seen above for discs B and C.

The prediction at 30% fibre content permits a direct comparison with discs B and C and thus of the expected effects of the longer fibre lengths of disc A. These predictions show that the longer fibre lengths and broader *FLD* of disc A result in predicted  $E_0$  and  $E_{90}$  values 10 and 14% stiffer respectively than those for disc B.

If the experimental values for disc A (25% fibres) in Table 2 are weighted (using a theoretical basis) to allow for fibre content differences, then stiffness values for disc A (30% fibres) of 2.8 GN/m<sup>2</sup> ( $E_0$ ) and 4.5 GN/m<sup>2</sup> ( $E_{90}$ ) result. This  $E_{90}$  value for 'disc A (30% fibres)' is of the order of 7% stiffer than the corresponding experimental values for discs B and C, while the  $E_0$  values for the three discs are very similar. Thus, at the same fibre content, the three disc types do exhibit a stiffness response which varies as a function of fibre length. The high degree of fibre agglomeration and increased voiding in disc A appear to have only a minor effect on stiffness, possibly causing a slight reduction. However, it is worth noting that the incidence of significant fibre agglomeration in GFPP injection mouldings does lead to increased interspecimen variability.

## DISCUSSION AND CONCLUSIONS

Examination of experimental and predicted stiffness values for GFPP edge-gated discs produced over a range of injection moulding conditions show that attempts to preserve fibre length by careful choice of injection moulding conditions

 Table 4
 Low strain stiffness predictions for the GFPP discs using

 FOD and FLD data
 FLD data

Disc and fibre	Tensile modulus (GN/m <sup>2</sup> )		
concentration (w/w)	0°	90°	<ul> <li>Anisotropy ratio</li> </ul>
A* (25.8%)	3.09	5.03	1.63
A* (30.5%)	3.40	5.73	1.68
B (30.6%)	3.07	5.03	1.64
C (30.5%)	3.03	5.41	1.79
C (core) (30.5%)	2.15	6.68	3.12

\* Disc A calculations have used the *FOD* for disc B. Calculations are shown for two fibre concentrations to allow comparisons (i) with discs B and C at the same fibre content and (ii) with disc A experimental data

do result in improved product stiffness, despite the degree of fibre agglomeration and increased voidage that occurs with these mouldings.

The strength of these GFPP mouldings also improves with preserved fibre length. In *Table 3*, the tensile strength values for disc A are given for 25% fibre concentration. Clearly, if these results for disc A are weighted to 30% fibre concentration, higher strength values for the  $0^{\circ}$  and  $90^{\circ}$  directions would be obtained than for discs B and C.

FLD determinations for the three disc types reveal that, as expected, it is back pressure rather than gate size (at least for the gate size range employed in this study) which has the predominant effect on fibre length in the GFPP mouldings examined. FOD does not appear to vary significantly in these disc mouldings over the range of conditions examined, however fibre dispersion is seriously affected by back pressure. It is noteworthy that had disc A been prepared with 30% fibre concentration (rather than 25%), then even greater fibre agglomeration might well have been expected for disc A moulding conditions.

Additional measurements at Cranfield<sup>13</sup> have been made on the other GFPP ICI 'Propathene' grade, HW 60 GR, which has much shorter, predispersed glass fibres, rather than the long fibre granule form (HW 70 GR) discussed above. It is worth noting that fibre length distribution measurements of typical injection mouldings of this predispersed grade have shown quite similar *FLD*'s to discs B and C (which were produced from the long fibre granule material).

It is quite significant that increases in stiffness and strength do result with adjustments in processing conditions to maintain fibre length, despite the reduced fibre dispersion that also occurs. Studies on sheet moulding compounds<sup>14</sup> have shown that products moulded to have agglomerated rather than well dispersed fibres show useful improvements in impact properties. These improvements result from the greater work required for fibre pull out after debonding in the agglomerated systems. Similarly enhanced impact properties might thus be expected for short fibre reinforced thermoplastics moulded with long, but agglomerated fibres.

Fibre agglomeration in GFPP (and other short fibre reinforced thermoplastics), however, does result in an inferior interspecimen reproducibility. In fact this inferior reproducibility may alter the apparent advantage in properties which is observed when only averaged data are considered. Furthermore, mould appearance has been observed to be inferior for GFPP mouldings in which serious fibre agglomeration has occurred.

# ACKNOWLEDGEMENTS

Grateful acknowledgment is made to the Science Research Council and to the Ministry of Defence (Procurement Executive) for the provision of grants in support of this work. The authors would like to thank Dr D. Sims of the Explosives Research and Development Establishment, Waltham Abbey for the production of mouldings and for helpful discussions and also Mr M. A. Christie of the Materials Department at Cranfield for carrying out much of the computation.

### REFERENCES

- 1 Schlich, W. R., Hagan, R. S., Thomas, J. R., Thomas, D. P. and Musselman, K. A. Soc. Plast. Eng. J. 1968, 24, 43
- 2 Filbert, W. C. Soc. Plast. Eng. J. 1969, 25, 65
- Darlington, M. W., McGinley, P. L. and Smith, G. R. Plast. Rubber Mater. Appl. 1977, 2, 51
- 4 Darlington, M. W. and Smith, G. R. Proc. Inst. Civ. Eng. Conf. 'Fibre Reinforced Materials: Design and Engineering Applications' London, 1977, p 57
- 5 Krenchel, H. 'Fibre Reinforcement' Akademisk Forlag, Copenhagen, 1964
- 6 Cox, H. L. Br. J. Appl. Phys. 1952, 3, 72
- 7 Sims, D. Plast. Polym. 1974, 42, 254
- 8 Darlington, M. W. and Saunders, D. W. 'The Structure and Properties of Oriented polymers' (Ed. I. M. Ward), Applied Science, London, 1975, Ch 10
- 9 BS 4618, part 1 (1970)
- 10 Darlington, M. W. and McGinley, P. L. J. Mater. Sci. 1975, 10, 906
- 11 Darlington, M. W., McGinley, P. L. and Smith, G. R. J. Mater. Sci. 1976, 11, 877
- 12 Darlington, M. W. and Smith G. R. Polymer 1975, 16, 459
- 13 Bright, P. F., unpublished data
- 14 Burns, R. and Pennington, D. Plast. Rubber Inst. Conf. 'Research projects in reinforced plastics' London, 1976