JOURNAL OF MATERIALS SCIENCE 11 (1976) 877–886

Structure and anisotropy of stiffness in glass fibre-reinforced thermoplastics

M. W. DARLINGTON, P. L. McGINLEY, G. R. SMITH
Department of Materials, Cranfield Institute of Technology, Cranfield, Bedford, UK

Techniques which are readily available, and which may be considered suitable for the qualitative or quantitative assessment of fibre orientation distribution in short glass fibre-reinforced thermoplastics are reviewed. The results of using several of these techniques in structural studies on injection mouldings of glass fibre-reinforced grades of polypropylene and polyamide 66 are presented. Uniaxial tensile creep tests were carried out on specimens cut from the mouldings and the anisotropy of stiffness of each moulding is compared with that predicted from the structural studies. Certain of the structural techniques are considered to be unreliable or of restricted applicability and it is concluded that the technique of contact micro-radiography is the most versatile; being capable of yielding reliable qualitative or quantitative information on fibre orientation distribution. Detailed structural studies on edge-gated injection moulded discs, using the technique of contact micro-radiography, show that the fibre orientation distribution varies dramatically through the thickness of the mouldings, even in cases where uniaxial tensile creep tests suggest isotropy of stiffness in the plane of the moulding. Care must therefore be taken when seeking to relate flexural data to tensile data and strength data to stiffness data.

1. Introduction

Test specimens may be prepared from unfilled thermoplastics with a molecular orientation that is reasonably uniform throughout the specimen, is of controlled intensity and is readily identified and reproduced. Thus, for unfilled thermoplastics, the anisotropy of mechanical properties may be studied even on an inter-laboratory basis with a satisfactory degree of reproducibility.

For short fibre-reinforced thermoplastics, inter-laboratory comparisons of mechanical properties are commonly made, giving reference only to the shape of the mould from which the test specimens were taken [1]. However, the process variables and, as will be seen in this paper, the particular materials used to fill the mould, can influence the orientation of the fibres in the moulding. As the mechanical properties are usually dominated by the orientation of the fibres, with the matrix orientation playing only a secondary role, it becomes essential to be able to fully characterize the nature of the fibre orientation present. The mechanical properties can then be measured for a specimen of known structure and inter-laboratory comparison of results becomes possible.

In this paper, techniques which are readily available and which may be considered suitable for the assessment of glass fibre orientation distribution are reviewed, and a critical appraisal is made of the results of applying several of these techniques to a range of injection mouldings. One of these, contact micro-radiography, is shown to be capable of providing both qualitative and quantitative details of fibre orientation distribution. As an example, the technique is used to provide a quantitative explanation for the apparently anomalous reversals of mechanical anisotropy in a group of injection moulded discs. Particular attention has been paid to the structure of edge-gated discs as they have been proposed as the source of "lower bound" (or weak direction) specimens in the extension of British Standard B.S. 4618 (parts 1.1 and 1.1.1) to the determination of the anisotropic creep properties of reinforced thermoplastics [2]. It should be noted that many of the structural features described below have also been observed by Darlington and McGinley [3] in ASTM tensile bars which have been proposed as the source of "upper bound" (or stiff direction) specimens in the above-mentioned standard.

2. Materials and mouldings

The materials used in this study and the abbreviated nomenclature used in the text are as follows:

GFPP–polypropylene containing nominally 25% by weight of coupled short glass fibres. (ICI "Propathene" grade HW 70 GR).

GFPA 66–nylon 66 containing nominally 33% by weight of short glass fibres. (ICI "Maranyl" grade A190).

Details of the various injection moulded samples of these materials used in this study are given in Table I.

With the exception of disc C, all the injection mouldings were obtained from one source. Plaque A is a flash-gated square plaque. The remainder of the mouldings are edge-gated discs of various sizes as detailed in Table I.

3. Stiffness tests and data

A standard technique has been employed in machining tensile test specimens from injection mouldings in order to measure stiffness properties. Specimens from edge-gated discs have been selected by initially constructing the 02 and 03 axes on the disc (as shown in Fig. 1) and then two specimens have been taken from either the 02 or 03 direction for any one disc, symmetrically about one of the constructed axes. Tensile dumb-bells with a parallel-sided gauge length of 46 mm have then been routed from the specimen blanks on a pneumatic routing machine.

The tensile creep modulus of these specimens has been measured on highly accurate creep apparatus capable of measuring all three principal strains during creep in uniaxial tension [4,5]. Creep experiments have been conducted in accordance with the procedures set down in British Standard B.S. 4618, part 1 (1970). The test temperature was 23 ± 0.3° C.

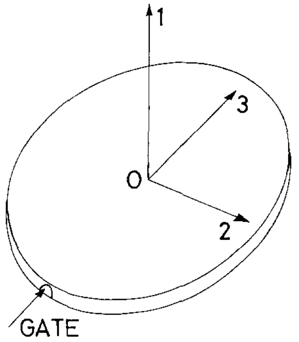

Figure 1 Definition of axes for edge-gated injection moulded discs. Direction 0 1 is perpendicular to the plane of the disc and 0 3 is parallel to the major flow direction. For the flash-gated plaque direction 0 3 was similarly taken to define the major flow direction.

Discussion below will be limited to results obtained using the 100 sec isochronous stress–strain procedure [6]. Isochronous tensile creep modulus data at the 100 sec axial strain of 0.002 are presented in Table I.

4. Structural analysis

4.1. Visual inspection

In translucent mouldings of short glass fibre-reinforced thermoplastics up to 2 or 3 mm thick a texture is often visible in transmitted light. This texture may be related to flow during the moulding process. An example is given in Fig. 2 of a region of plaque A. Despite the presence of the readily observable features depicted in Fig. 2, stiffness measurements on specimens cut parallel and perpendicular to the texture markings show only a 5% anisotropy with the weak direction lying *parallel* to the texture markings (see Table I).

TABLE I Dimensions and modulus data for the mouldings

Sample	Material	Diameter (mm)	Thickness (mm)	Tensile modulus		Anisotropy ratio
				Direction 0 3	Direction 0 2	
Plaque A	GFPP	(165)	3	4.26	4.05	1.05 : 1
Disc B	GFPP	150	3	4.58	4.41	1.04 : 1
Disc C	GFPP	100	6	2.48	4.00	1 : 1.61
Core (C)	GFPP	100	3	1.58	2.79	1 : 1.77
Disc D	GFPP	100	3	3.26	3.96	1 : 1.23
Disc E	GFPA – 66	100	3	7.76	6.54	1.19 : 1
Disc F	GFPA – 66	115	6	6.47	7.31	1 : 1.13

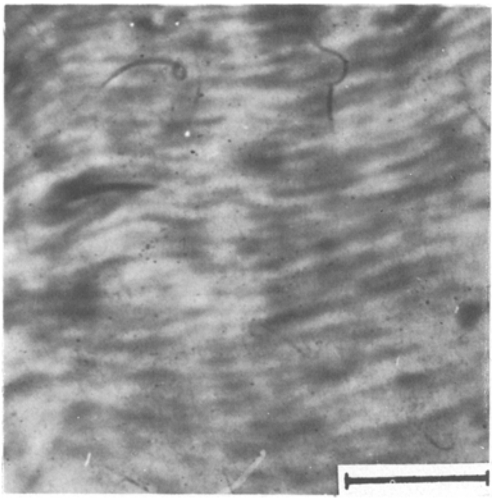

Figure 2 Appearance of a region of plaque A when viewed in transmitted light. The photograph is in the 2–3 plane with the 02 direction horizontal. Bar represents 1 cm.

Optical examination via transmitted light of slices cut parallel to the plaque surface at various depths through the thickness direction, reveals that the observable texture is confined to a central planar zone of approximately 1 mm thickness. If a through thickness section is viewed in a direction parallel to the plane of the disc, the central zone has a white appearance which Darlington and Smith [7] have shown to be associated with the occurrence of voids in the moulding. Thus the readily observed texture in the moulding depicted in Fig. 2 does not give any direct evidence of the overall fibre orientation. Such visual examination techniques can, therefore, be misleading if used to predict the anisotropy of mechanical response.

Rather more sophisticated optical techniques are discussed elsewhere [8–12]. However, none of these offer more than a semi-quantitative assessment of fibre orientation distribution and those involving removal of the matrix [8, 11, 12] are prone to experimental errors.

4.2. Macro-radiography

The possible application of macro-radiography to the determination of fibre orientation in short glass fibre-reinforced thermoplastics mouldings has been considered in this laboratory and elsewhere [13]. Radiographs of regions near the centres of discs B and C are shown in Fig. 3a and b. Both discs show a similar flow pattern radiating from the gate in the mould, suggesting that in the central areas of these discs the fibre orientation distribution (and thus the anisotropy of stiffness) should be similar.

Isochronous tensile creep moduli of specimens cut from the central region of discs B and C are given in Table I. In disc B the 03 direction is 4% stiffer than the 02 direction, whereas in disc C, the 02 direction is 38% stiffer than the 03 direction. The order of anisotropy of the two mouldings is reversed, despite the similar appearance of the radiographs of Fig. 3. Clearly, information available from macro-radiography is of little value for the estimation of stiffness or anisotropy of stiffness if it cannot detect the differences between the two discs.

In order to determine the origin of the texture observable in the radiographs, the 6 mm thick specimen of disc C used for Fig. 3b subsequently had 1.5 mm machined from both faces. A radiograph of the remaining central zone is presented in Fig. 3c. It is apparent that the texture of the core radiograph is identical to that of the full disc thickness. Radiographs of regions close to the mould surfaces were found to be featureless. Thus the structure that produces the radiographic texture of the whole disc is contained in the central core zone.

The moulding conditions used in the production of disc C were selected with the intention of retaining fibre length [14]. Other GFPP discs were produced using the same mould, but with moulding conditions known to result in increased fibre breakage (and fibre dispersion). A macro-radiograph of one of the latter discs (coded C*) is shown in Fig. 3d and is seen to be featureless.

It will be seen from contact micro-radiographic data, presented in Section 4.4, that disc C contained many fibre clumps, most of which were contained in the central 4 mm thick core zone of the 6 mm thick disc. Contact micro-radiographic studies on disc C* have shown the absence of fibre clumps throughout the disc.

This confirms that the texture apparent in the macroradiographs of Fig. 3 is associated with fibre clumps and this technique, therefore, gives no information on the well dispersed fibres and hence on the overall fibre orientation distribution.

4.3. Metallographic polishing

The orientation of the fibres that intersect a plane in the specimen has often been investigated by the metallographic polishing technique [15] since the methods and equipment are widely

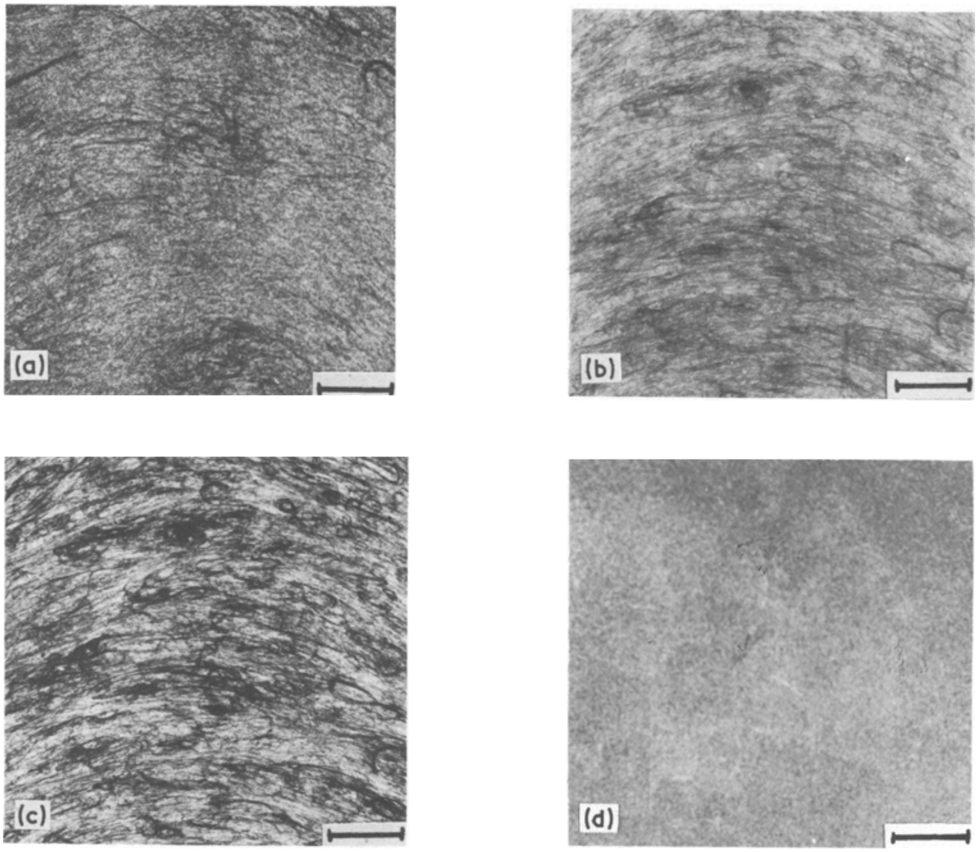

Figure 3 Macroradiographs of regions of (a) disc B, (b) disc C, (c) the core layer of disc C and (d) disc C*. In all cases the plane of the radiograph is the 2–3 plane with the 02 direction horizontal. Bar represents 1 cm.

known and available. Some good micrographs by Bowyer and Bader [16] of polished surfaces of an injection moulded GFPA-66 bar show the difficulty of assessment of orientation intensity by this technique.

It is possible to determine the orientation of the fibre to the polished surface by measuring the orientation and length of the major and minor axes of the elliptical fibre section. However, these measurements are frequently made difficult and unreliable due to damage to the fibre ends during polishing. An additional problem of lack of contrast between certain matrices and the glass fibres may also occur.

The fibre damage can easily be seen in scanning electron micrographs of polished surfaces [7]. The difficulty in defining the extent of the elliptical polished surfaces (in optical micrographs) when the intimate contact between the matrix and fibre has been destroyed by the polishing process can also be appreciated from these electron micrographs. In utilizing ellipticity for the assessment of the three-dimensional orientation of a fibre it should also be noted that an exactly circular cross-section of the glass fibre cannot be assumed [17].

Whether this metallographic polishing technique is used for qualitative or quantitative assessment of fibre orientation distribution, it suffers from the drawback that only fibres intersecting a plane are observed and thus too few fibres may be sampled. The problems associated with this have been illustrated elsewhere [3].

4.4. Contact micro-radiography

The use of contact micro-radiography (CMR) overcomes most of the problems encountered with metallographic sectioning and polishing procedures. The CMR technique produces an image of all the fibres contained in a slice approximately 100 μm thick. Darlington and McGinley [3] have previously described the technique in

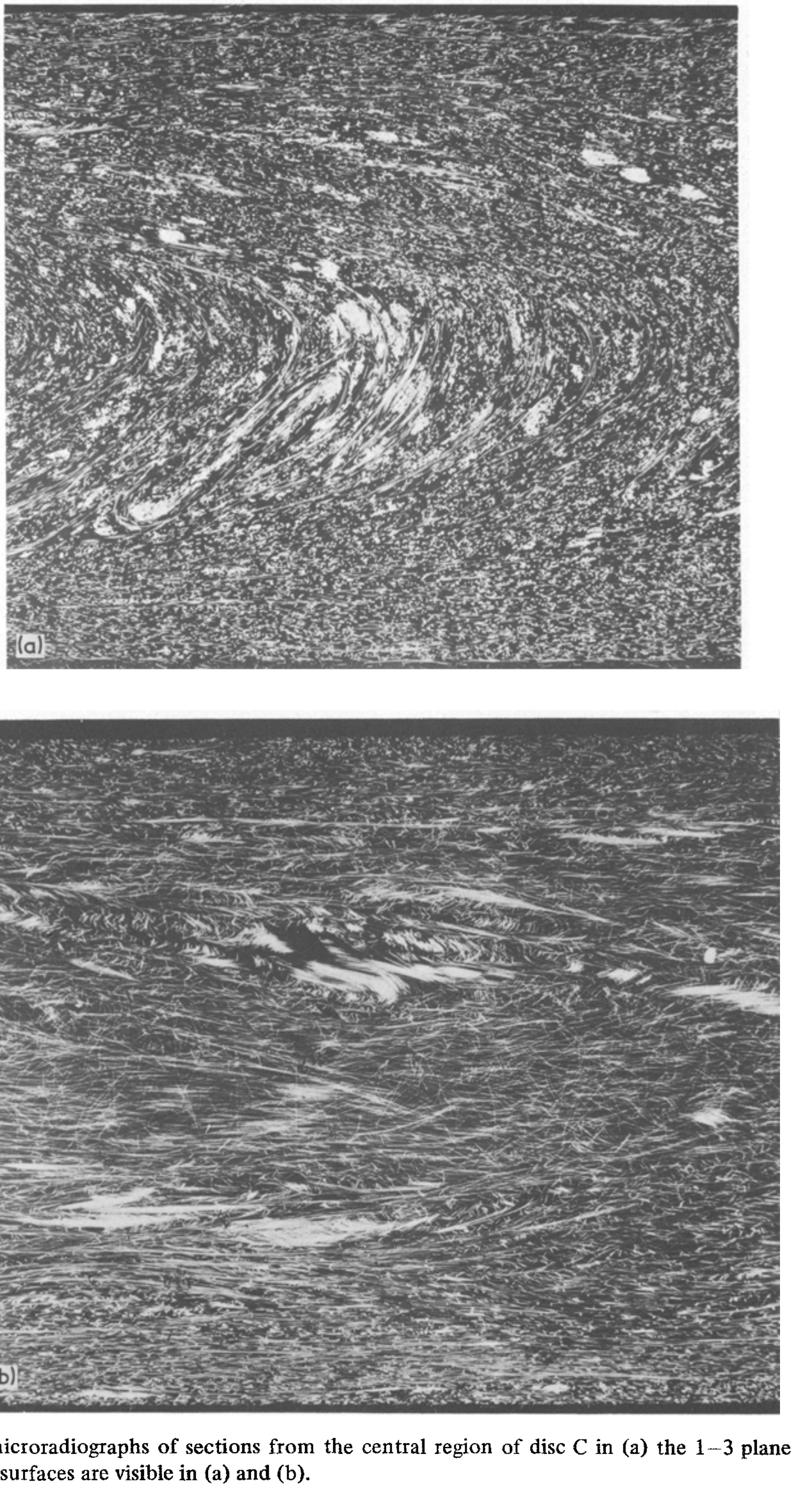

Figure 4 Contact microradiographs of sections from the central region of disc C in (a) the 1–3 plane and (b) the 1–2 plane. Both mould surfaces are visible in (a) and (b).

detail and a direct comparison has been made with the results from metallographic polishing.

In Fig. 4 the central region of disc C is examined using the CMR technique. Poor dispersion of the fibres is apparent, with large clumps or bundles of fibres clearly visible. It can be seen in these radiographs, and it has been verified by other observations in this laboratory, that most of the fibre clumps are confined to the core region of the moulding.

From this limited CMR information a qualitative explanation of the mechanical anisotropy of disc C and its core specimen can be advanced: the central zone of the disc contains fibres highly aligned into the 1–2 plane. Within this plane the fibres are predominantly oriented parallel to the 02 direction. Thus the core tensile specimens show a high anisotropy (modulus ratio 1: 1.77) with the low modulus direction (03) very close to the stiffness of the unreinforced polymer. The surface layers have a similar appearance in both (a) and (b) of Fig. 4, with few of the fibres oriented out of the plane of the disc. This near random array within the 2–3 plane will contribute equally to the modulus in the 02 and 03 directions. Thus the level of anisotropy in the 6 mm thick specimen would be expected to be slightly less than that of the core, as is observed.

At this point the relative magnitude of the moduli for the disc and the "core only" deserve comment. In particular it is perhaps surprising that the modulus in the 02 direction is considerably less for the core than for the whole disc, since the disc response is then supposedly the sum of a layer in which the fibres are highly aligned in the direction of the applied stress (the core layer) plus two (surface) layers in which the fibres are oriented approximately randomly in the plane of the disc. It would normally be expected that this would produce a modulus in the 02 direction for the whole disc that was less than that for the core only.

Possible contributory factors to the low core stiffness could include the following:

(i) there is a significant degree of voiding in the central layer of the 6 mm thick disc [7] with negligible voiding in the surface layers;

(ii) the incidence of fibre clumps is highest in the core layer. Such clumps may severely reduce fibre efficiency in reinforcing the matrix;

(iii) more fibres lie out of the plane of the disc in the core layer than in the surface layers;

(iv) the fibre concentration in the core differs from that in the surface layers. (Limited measurements to date do not support this.)

The examination of these possible causes is under investigation at present.

5. Use of CMR to resolve anomalous stiffness behaviour

In many cases the qualitative assessment of fibre orientation distribution demonstrated in Section 4.4 will be adequate. In those cases where a quantitative assessment of fibre orientation distribution is necessary, the CMR technique can be readily extended to provide this information. This will be demonstrated below using discs D, E and F. These three discs have relatively low levels of mechanical anisotropy and the anisotropy of disc E is reversed compared with discs D and F. Qualitative assessment of these discs in the manner outlined in Section 4.4 proved inadequate to distinguish these differences in mechanical behaviour. A detailed assessment of the fibre orientation distributions in these three discs, therefore, provides a useful illustration of the extent to which the CMR technique can be made quantitative.

5.1. Properties of the discs

The mechanical data of discs D, E and F (Table I) shows apparently anomalous behaviour. Discs D and E come from the same mould with disc D produced in GFPP and disc E in GFPA-66. The GFPP moulding has an anisotropy ratio of 1: 1.23 (with the 02 direction the stiffer) while in the GFPA-66 moulding the ratio is 1.19: 1 (03 direction stiffer). So, for the same mould with different materials (and process variables), a reversal of anisotropy is observed.

A change of thickness in the two GFPA-66 mouldings (discs E and F) from 3 to 6 mm is also accompanied by a reversal of the stiffness anisotropy from 1.19:1 (03 stiffer) in disc E to 1: 1.13 (02 stiffer) in disc F. The change in disc diameter from 100 to 115 mm is not considered to be an important variable.

5.2. Measurement of fibre orientation distribution

Inspection of contact micro-radiographs of sections cut perpendicular to the plane of the discs shows that the fibres lie predominantly in the plane of the discs (see Fig. 6). In quantifying the fibre orientation distribution, the major step is, therefore, to determine the orientation of fibres in this plane.

The orientation of the fibres in a slice cut parallel to the plane of the disc (2–3 plane) may be readily determined by the CMR technique. An example is given in Fig. 5. Fig. 5a shows *part* of the micro-radiograph used to determine the orientation distribution shown in Fig. 5b. From Fig. 5b, an average fibre orientation direction and an

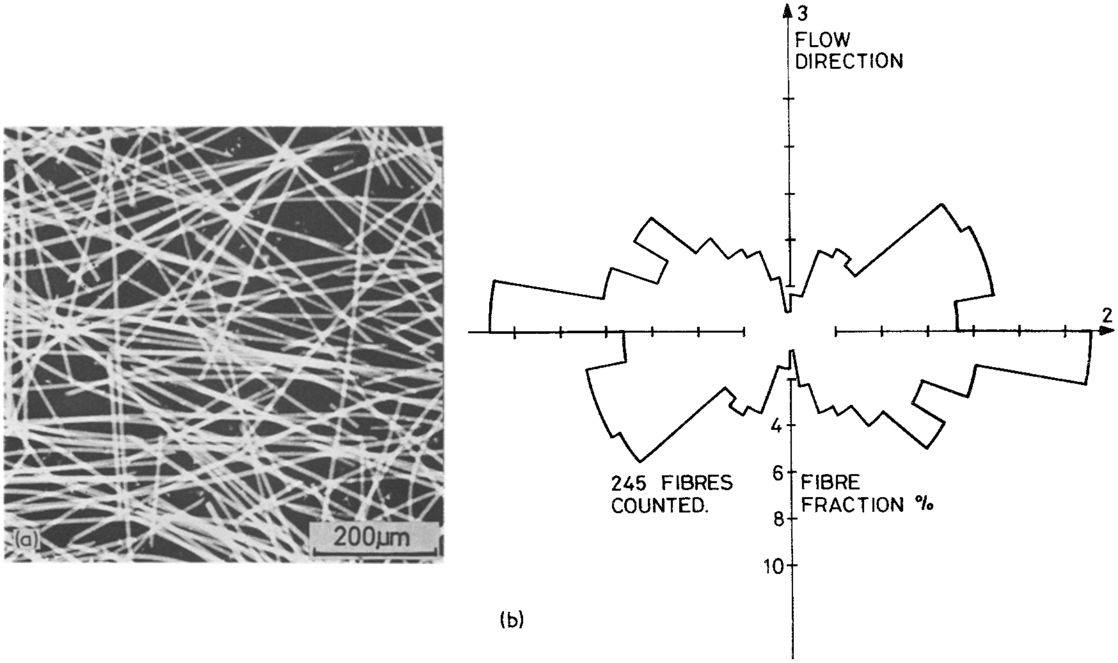

Figure 5 Determination of the fibre orientation distribution of a slice parallel to the 2–3 plane. (a) Part of a contact microradiograph of the surface layer of disc D (direction 02 horizontal). The orientation of a selection of fibres as measured on the micrograph is plotted on the normalized histogram (b).

indication of the intensity of the orientation may be obtained.

A series of slices similar to that in Fig. 5a have been taken through the thickness from the central area of each of the three discs. The spacing between the slices was such that between 13 and 16 slices were obtained from each disc with the first and last slice containing a surface layer of the disc. The fibre orientation distribution was measured for at least nine of the slices of each disc and an average direction and intensity of orientation were calculated by computer for each slice. The results are displayed in Fig. 6 where the orientation and length of the arrows represent the direction and intensity of fibre orientation in the plane of the disc at locations indicated in the adjacent micrograph.

In each disc, the central core region has the fibres oriented transversely to the major flow direction (the 03 direction). The thickness of the core region varies from approximately 20% of the total mould thickness in disc E to 50% in disc F. Discs E and F have the surface layer fibres oriented parallel to the flow direction while in disc D there is only a narrow intermediate layer between the surface and core layers which has fibres oriented parallel to the flow direction.

5.3. Fibre distribution and anisotropy of stiffness

The information in Fig. 6 on fibre orientation direction and intensity may be used in a visual assessment of the relative effects of surface and core layers to estimate the resulting macroscopic mechanical anisotropy. This approach emphasizes the frequently opposing effects of surface and core regions, but the visual summation can be difficult.

An alternative method is to sum the normalized distributions of the layers (i.e. to sum the set of distributions of the type depicted in Fig. 5b) from the surface to the centre of the core region. The resultant fibre orientation distribution pattern represents the total distribution but conceals the variations through thickness. This single distribution pattern greatly simplifies the assessment of the effect of fibre orientation upon stiffness; this being primarily dependent upon the total fibre orientation distribution. The total through-thickness distributions for discs D, E and F are shown in Fig. 7 in the form of smoothed curves.

Disc D shows a 15° off-axis orientation of the major orientation direction which could be a genuine assymetry of the flow pattern of the whole disc or a local effect in the region in which orientation measurements were made. Local

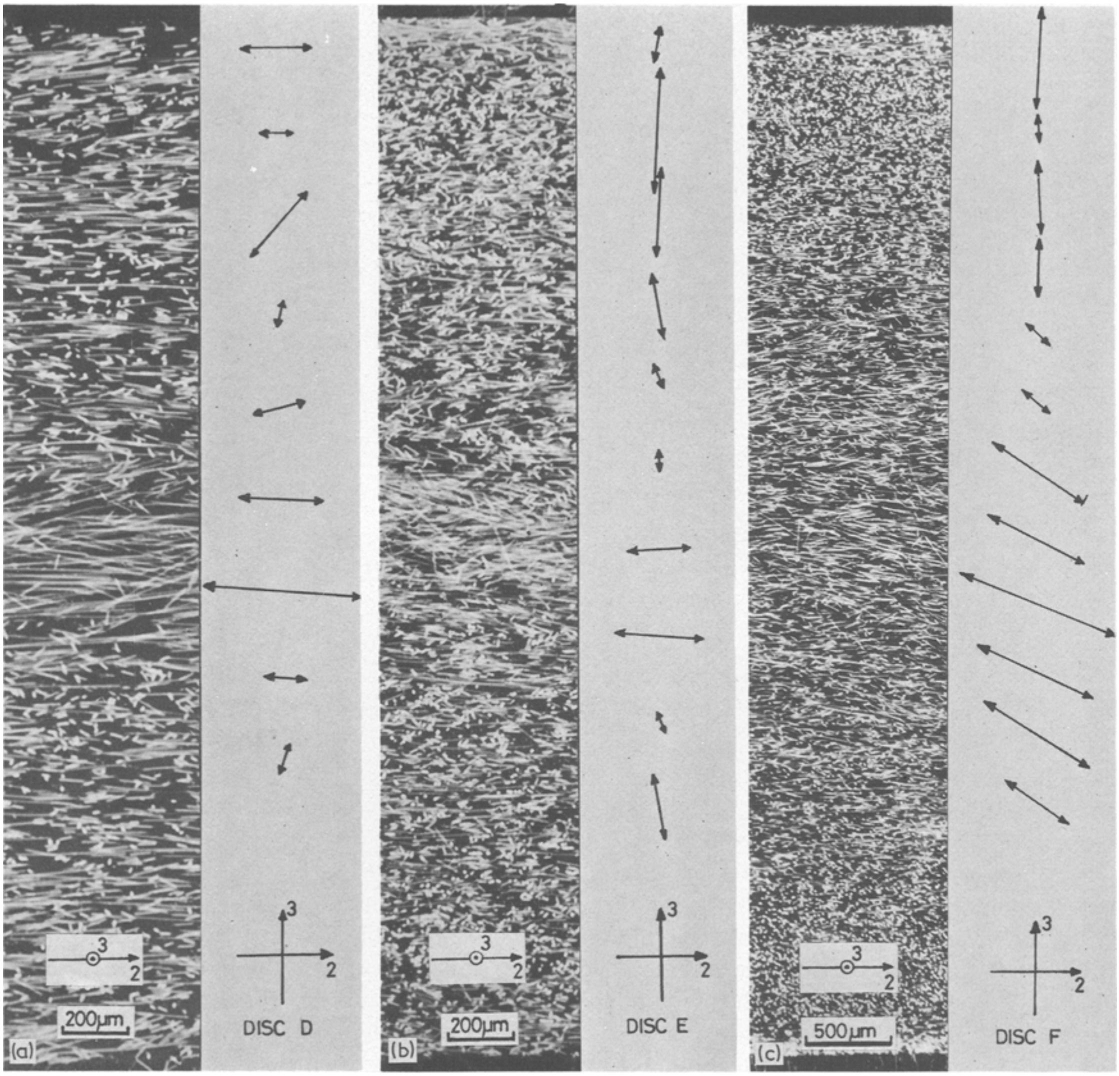

Figure 6 Variation of fibre orientation through the thickness of discs D, E and F. The microradiographs show views of slices cut parallel to the 1–2 plane, extending from the upper to the lower mould surface. Each double-headed arrow represents the average direction and intensity of the fibre orientation on the 2–3 plane for layers located at various depths through the thickness of the moulding.

variations in the position of the narrow transversely oriented core region were also observed in this disc. Typically, the through-thickness location of the core fluctuated by 0.5 mm over a distance of 2 mm.

Disc E has a symmetrical distribution that is centred on the 03 axis so that the measured anisotropy should be the maximum possible for this moulding.

There is a marked assymetry of the distribution in disc F. The surface layer fibres of disc F are seen in Fig. 6c to be aligned parallel to the 03 direction while the core is aligned at approximately 70° to the 03 direction. Superposition of two symmetrical distributions at these orientations and of appropriate intensity produces an orientation pattern similar to that of disc F.

The reversals of mechanical anisotropy noted earlier between discs D and E and discs E and F can now be accounted for in terms of the fibre orientation distributions. It is clear from Fig. 7 that in discs D and E the stiffer directions should be 03 and 02 respectively as is observed (Table I). The difference between the discs lies in the orientation of the surface layers since the core regions are of a similar size and have the same orientation

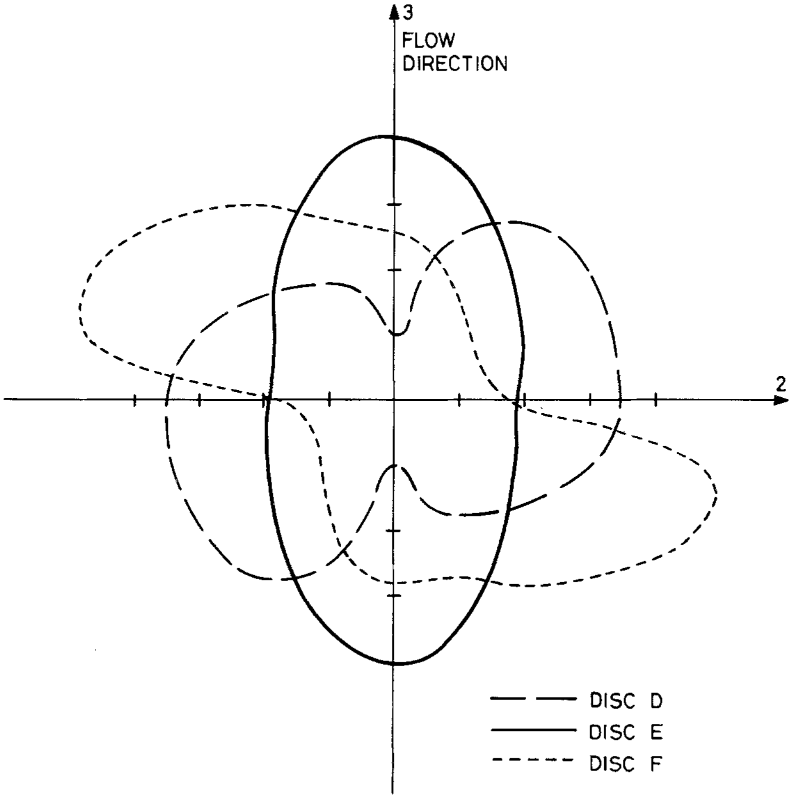

Figure 7 The total through-thickness fibre orientation distribution in the 2–3 plane for discs D, E and F. (Smoothed curves from normalized histograms.)

(see Fig. 6). For the GFPP moulding (disc D), the surface regions have assumed the transverse orientation of the core region, whilst in both the GFPA-66 mouldings (discs E and F) the fibres in the surface layers are aligned parallel to the major flow direction (03).

In discs E and F the observed reversal of anisotropy is due to the relative thicknesses of the transversely oriented core regions. In disc E the core region occupies approximately 20% of the total thickness so the surface orientation predominates; whereas in disc F the core is approximately 50% of the total thickness and is able to overcome the effect of the surface layers. The surface layers are of a similar absolute thickness in each disc, with the increased mould thickness in disc F allowing the core region to expand in absolute thickness and as a fraction of the total. The anisotropy of disc F is low because of the off-axis orientation of the core region. Even in Fig. 7 it is not obvious which of the principal directions should be the stiffer as the contribution of the off-axis fibres are difficult to assess by eye.

While Fig. 7 is extremely useful to indicate which will be the stiffer direction, it cannot of itself give an indication of the absolute magnitude of the stiffness since this will depend upon the stiffnesses of the matrix and fibres, the fibre concentration and length and the matrix–fibre coupling efficiency. It is, however, the first step in such calculations and current work is proceeding in this direction. A complete analysis must, of course, be based on the three-dimensional fibre orientation distribution.

6. Conclusions

The contact micro-radiography technique is shown to be capable of quantitative determinations of fibre orientation distributions in short glass fibre-reinforced thermoplastics. It is considered to be superior to the other commonly available techniques reviewed here. It is *not* restricted to simple mould shapes, translucent or unpigmented matrices or well-coupled systems.

For the discs examined in this paper, the core region is characterized by a fibre alignment that is transverse to the major flow direction and may have a significant component of the orientation that is out of the plane of the moulding. The thickness of the core depends upon a number of variables including the thickness of the moulding. Increasing the thickness of the moulding is more likely to lead to a stiffer modulus in the direction perpendicular to the major flow direction since the thickness of the surface layers remains constant and the relative contribution of the core is increased [18].

It has been suggested that in the past [19] that it is not possible to have significant orientation of fibres without mechanical anisotropy. However, lack of stiffness anisotropy does not necessarily imply random orientation. In injection moulded specimens it is more likely to mean a balance between the orientation in the surface and core regions. The relative effect of the two regions determines the degree of anisotropy and the direction of greater stiffness.

This balance of fibre orientation in the core and surface layers is of considerable importance when comparing flexural stiffness and tensile stiffness and when dealing with the strength of these materials.

Acknowledgements

Grateful acknowledgement is made to the Science Research council for a grant in support of this work. Dr C. Page provided the computer programme used in the calculation of the orientation intensity. Most of the mouldings were kindly supplied by ICI Ltd, Plastics Division. The variation of stiffness with mould thickness in edge-gated injection moulded discs was first brought to our attention by C.M.R. Dunn and S. Turner of ICI Plastics Division, to whom thanks are also due for helpful discussions in the initial stages of this work.

References

1. H. S. LOVELESS and D. E. McWILLIAMS, *Poly. Eng. Sci.* **10** (1970) 139.
2. C. M. R. DUNN and S. TURNER, "Composites – Standards Testing and Design", N.P.L. April 1974. Proceedings (IPC Science and Technology Press, 1974, Guildford) p. 113.
3. M. W. DARLINGTON and P. L. McGINLEY, *J. Mater. Sci.* **10** (1975) 908.
4. M. W. DARLINGTON and D. W. SAUNDERS, *J. Phys. E: Sci. Instrum.* **4** (1970) 511.
5. M. W. DARLINGTON and D. W. SAUNDERS, Anisotropic creep behaviour, in "The structure and properties of oriented polymers", edited by I. M. Ward (Applied Science, 1975, London) Ch. 10.
6. S. TURNER, *Trans. J. Plast. Inst.* **31** (1963) 60.
7. M. W. DARLINGTON and G. R. SMITH, *Polymer* **16** (1975) 459.
8. D. C. FUCCELLA, 27th Annual Technical Conference of Reinforced Plastics/Composites Institute, S.P.I., Section 11 – F (1972).
9. V. KARPOV and M. KAUFMAN, *British Plastics* **38** (1965) 498.
10. E. M. ROWBOTHAM, S. A. E. Automotive Engineering Congress, Detroit, Michigan, 740264 (1974).
11. J.–M. CHARRIER and M. J. SUDLOW, *Fibre Sci. Technol.* **6** (1973) 249.
12. E. JINEN and M. SUZUKI, *Kobunshi Kagaku* **30** (1973) 144.
13. A. NEMET, A. D. BLACK and W. F. COX, *Trans. J. Plast. Inst.* **30** (1962) 192.
14. W. R. SCHLICH, R. S. HAGAN, J. R. THOMAS, D. P. THOMAS and K. A. MUSSELMAN, *S.P.E.J.* **24** (1968) 43.
15. A. D. S. DIGGWA and R. H. NORMAN, *Plast. Polym.* **40** (1972) 263.
16. W. H. BOWYER and M. G. BADER, *J. Mater. Sci.* **7** (1972) 1315.
17. D. V. BADAMI, private communication.
18. C. M. R. DUNN and S. TURNER, private communication.
19. G. W. WEIDMANN and R. M. ORGORKIEWICZ, *J. Mater. Sci.* **9** (1974) 193.

Received 19 August and accepted 25 November 1975.