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CHARACTERISATION OF FINITE LENGTH COMPOSITES: Part IV—Structural studies on injection moulded composites

(Technical Report)

Prepared for publication by

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Characterisation of finite length composites: Part IV—Structural studies on injection moulded composites (Technical Report)

ABSTRACT

The microstructure of discontinuous fiber composites can be articulated by many parameters. In this collaborative study as part of series of parallel projects under the IUPAC Working Party IV.2.1 on Structure and Properties of Commercial Polymers, various laboratories have carried out measurements. Their results and observations are summarised in this paper. In this family of materials, the microstructure should be described by at least three parameters, namely fiber orientation distribution, fiber length distribution and fiber content.
It is shown in this work that there is a profound influence of the processing history on the microstructure for these composite materials, and that there is a considerable challenge in measuring the macro-, meso and microstructural aspects.

1 INTRODUCTION

The materials under investigation in this study are injection moulding engineering plastics which have been prepared by pultrusion compounding. It is now well established that this family of materials can produce significantly longer fibers in the moulded artefact than by traditional extrusion compounded means (**1**). This margin may reach as much as an order of magnitude higher aspect ratio, but depends upon the fiber/matrix system and the manner in which subsequent processing takes place. An optimum number average l/D ratio for extrusion compounded (short) fiber composites would be 100 to 200, but in the case of pultrusion compounded (long) fiber compounds) can reach values in excess of 1000; this very much depends on the processing geometry, flow path and mould cavity design, and actual magnitudes of attainable fibers lengths are always relative to the severity of processing. In some cases, individual residual fibers of the original length (l_o) have been observed in long fiber moulded parts. The conventional injection moulding process is used with effectively similar conditions for both feedstock variants; this forming process offering a limited range of processing variables for altering the microstructure-properties relationship.

Producing components composed of longer fiber reinforced composites provides interesting new challenges for characterisation, and changes in fiber length and orientation are more pronounced. The influence of the flow path and shear history has a profound effect upon the resultant microstructure and hence the ultimate properties. The rheological characterisation of these materials with an insight into the flow induced morphological changes is described elsewhere (**2**).
In moulded panel components, the extent of in-plane anisotropy arising from the injection moulding process can be moderated by the methods reported by Allan and Bevis (**3**). In their novel process development, once the melt is injected into the mould in the normal way, it is pulsed in order to align polymer chains and fibers by shearing the fluid material remaining in the mould cavity by actuating pistons, typically these operate in the direction of mould-fill. Their process is known as the multiple live-feed process.

The microstructure of injection mouldings is extremely complex. It is challenging to attempt to characterise the fiber and molecular orientation of both conventional and multiple live-feed injection moulded samples. It is often convenient to visualise the microstructures attained by the simple model of through thickness skin/core/skin layers, and for this study it was

our intention to produce some variations by influencing the flow and hence the processing conditions which dictate the resultant "structure". If a wide range of moulding conditions are used, and the resultant moulded parts are truly "structures" then there is an implication that each structure requires experimental characterisation of its morphology before any description of the overall mechanical properties and the material properties is possible.

In order to gain this understanding, a move towards a quantitative microstructural characterisation is necessary. This paper is part of a series from the work carried out by IUPAC Working Party IV.2.1 and focuses on the microstructural issues observed by the contributors. In principle, this involves a measurement of fiber length distribution, fiber content and the fiber orientation function.

In exploring the relationship property-structure-processing and rheology we intend to conduct the work with both traditional and multiple live-feed mouldings. Moreover, the materials will have two different fiber systems (glass and carbon, Kevlar® is omitted) and two different matrix materials (polyamide 6,6 and polypropylene). This paper forms part of a series concerned with these materials which describes the mechanical properties, processing routes and flow characteristics of this family of materials. Moreover, the microstructural data in this study is used to model the stiffness in Paper V which follows on in this series of papers.

The following laboratories have contributed to this project under the umbrella of an IUPAC working party (Working Party IV.2.1) and this hopefully adds value to the contribution of this work:-

Laboratory 1 ICI Materials (D R Moore, R S Bailey, G v Bradsky, R S Prediger).
Laboratory 2 Shell Research Arnhem, (A Cervenka).
Laboratory 3 Rhone-Poulenc, (Y Giraud).
Laboratory 4 Huls AG, (H Motz).
Laboratory 5 National Research Council Canada, (T Vu-Khanh).
Laboratory 6 Brunel University, (M J Bevis, P S Allan).
Laboratory 7 B P Chemicals, (M J Cawood, A Gray, with contributions from the A Duckett at the IRC, Leeds University).
Laboratory 8 Institut für Technische und Makromoleclare Chemie, Hamburg (H G Zachmann, G v Krosigk).

2 DESCRIPTION OF SAMPLES

The materials produced for this study were prepared from the following constituent fibers and polymer matrices using either conventional injection moulding or by multiple-feed injection moulding:

fibers:

i) E-glass fibers
ii) HS-type carbon fibers

Polymers:

i) polyamide 6.6 homopolymer
ii) coupled polypropylene homopolymer

The details of the materials and their preparation is reported in the Introductory paper of this series (**4**). Materials containing a polyaramid (Kevlar®) fibers were also manufactured, but are not discussed in this paper. This is because the microstructural investigation is exceedingly difficult owing to the problems encountered in preparing polished sections. Additionally, a different approach would be needed because these fibers tend not to break along their length but rather defibrillate along their axis. So nearly all fibers retain their original fiber length, but change in diameter. Moreover, the fiber orientation can differ significantly along the fiber axis as these fibers cannot be considered as rigid rods. For these reasons these materials have been omitted from analysis.

The processing operations employed were by pultrusion compounding followed by injection moulding. All materials were experimental grades of "Verton®" supplied by ICI plc (now LNP (UK) [a subsidiary of Kawasaki Steel]) and incorporated a notional 40% by weight of

fiber reinforcement. Pultrusion compounding enabled a granule feedstock for injection moulding to be prepared with control of the length of the continuous fiber reinforcement. The fiber length in the granule was either 5 mm or 10 mm.

Two types of injection moulding were used to prepare the samples; a conventional (STATIC) process and a multiple live feed (DYNAMIC) process (**3**). In this paper, these processes are more fully described, however, the aim of the processing operation was for the DYNAMIC procedure to more fully align the fiber in the direction of mould fill, whilst the expectation was for the conventional moulding process to achieve the more traditional "skin-core" structure through the thickness of the moulding. (i.e. the fibers in the "skin" would be aligned in the direction of mould fill, whilst the fibers in the "core" would be aligned transverse to the direction of mould fill). This is an oversimplified description of fiber orientation, included here merely for purposes of describing the mouldings, in practice a skin-core structure was obtained from both processing routes, but significant differences in structure were attained which was the aim.

The moulded test-pieces obtained will be referred to in this work as follows:-
The matrix material is either polypropylene, designated PP or polyamide 6,6 designated PA.
The fiber type is either carbon, designated c or glass designated g
Multiple live feed mouldings are designated type 'DYNAMIC'
Conventional injection mouldings are designated type 'STATIC'.
fiber lengths (in mm) in the pultruded granule (the feedstock for injection moulding) are designated **5** or **10**.

Ten different materials were used in the study, the material combinations studied were:
PA/c 10DYNAMIC and PA/c 10STATIC; PA/g 5DYNAMIC and PA/g 5STATIC; PA/g 10DYNAMIC and PA/g 10STATIC; PP/g 5DYNAMIC and PP/g 5STATIC; PP/g 10DYNAMIC and PP/g 10STATIC.
The plaque mouldings were nominally 6 mm thick and Figure 1 shows their geometry and dimensions. The STATIC process is gated through a single side fan design, whilst the DYNAMIC process employed a fan gate at each end of the mould.

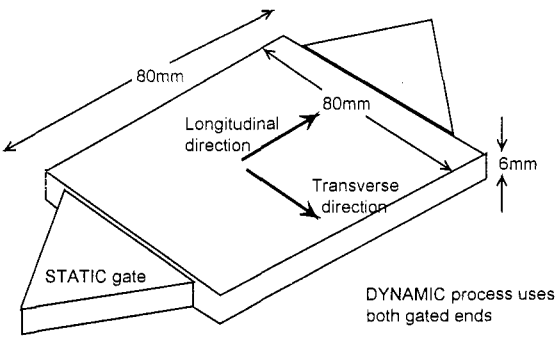

Figure 1. The test mouldings used in this work.

3 OBSERVATION OF THE MICROSTRUCTURE.

3.1 Matrix characterisation

This investigation was performed solely on the glass fiber reinforced polypropylene injection moulded plaques for both STATIC and DYNAMIC processes. Both 5 mm and 10 mm granule length materials were studied.

The orientation of the molecules in the matrix was determined at two locations designated by I and II. Rods having diameter 1 mm by 6 mm were cut out of each plaque as indicated in Figure 2. The orientations referred to in this investigation are MD (Melt flow Direction as longitudinal direction in Fig. 1), TD (Transverse Direction) and ND (Normal Direction - the through thickness direction). Pole figures of the 110 040 and 130 crystal reflections were determined at different distances from the surface by using a new method (**5**) developed for these

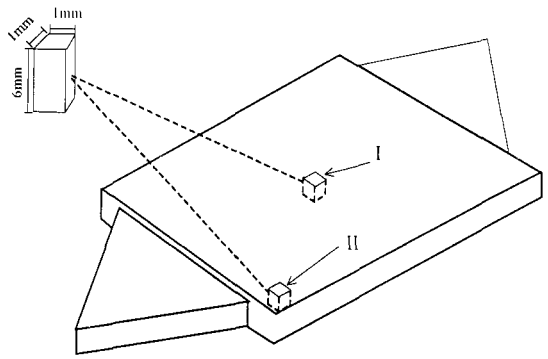

Figure 2. A schematic diagram showing the location of the rods extracted from the middle (I) and edge (II) of the sample.

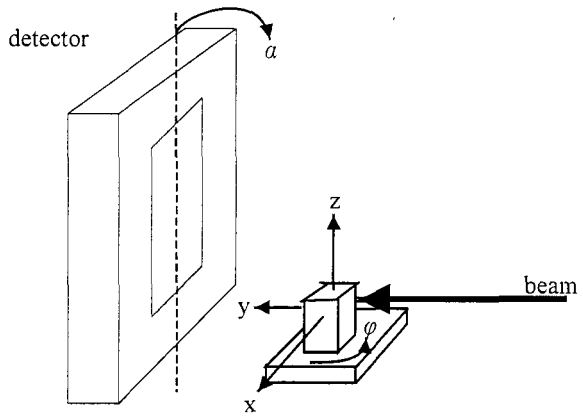

Figure 3. The experimental set-up of the rod in the beam. The rod is moved in the z-direction to measure the pole figures of different locations in the sample. The detector is turned around the y-axis and the sample is turned around the x-axis.

measurements employing synchrotron radiation. The rod was placed perpendicularly to the beam (see Figure 3). The beam had a verticle extension of 0.6 mm and a lateral extension of 2 mm. The rod was rotated about the z-axis in steps of 5°. After each step the X-ray scattering was measured by means of a one-dimensional Gabriel detector having a length of 7 cm, as a function of the angle α indicated in the figure. Different angles of αwere obtained by rotating the detector about the x-axis in steps of 5°. In order to obtain the pole figure as a function of the distance **d** from the surface, the sample was moved in the z direction in steps of 0.5 mm. From the pole figures the Hermans operation functions of the three crystal reflections $\mathbf{f}_{110}$, $\mathbf{f}_{040}$ and $\mathbf{f}_{130}$ with respect to the axis MD, ND and TD were calculated. By means of the equation of Wilchinski (**6**) the Hermans orientation function of the chains $\mathbf{f}_c$ was obtained.

Figure 4 shows an example the pole figures of the sample 5PP/g STATIC I at the distance **d**=0.5mm from the surface. One can recognise that some orientation occurs. In particular, the 040 plane is oriented preferentially parallel to the plaque. Similar pole figures showing various degrees of orientation were obtained for all other samples prepared by DYNAMIC processing. In contrast, in the conventionally injection moulded (STATIC) materials there was no evidence of orientation in the matrix polymer chains.

Figures 5a and b show the Hermans orientation functions of the polymer chains with respect to the MD direction as a function of the distance from the surface of the plaque **d** for the material filled with glass fibers with 5 mm and 10 mm length respectively. One can see that generally, with increasing distance from the surface, the orientation of the chains first increases

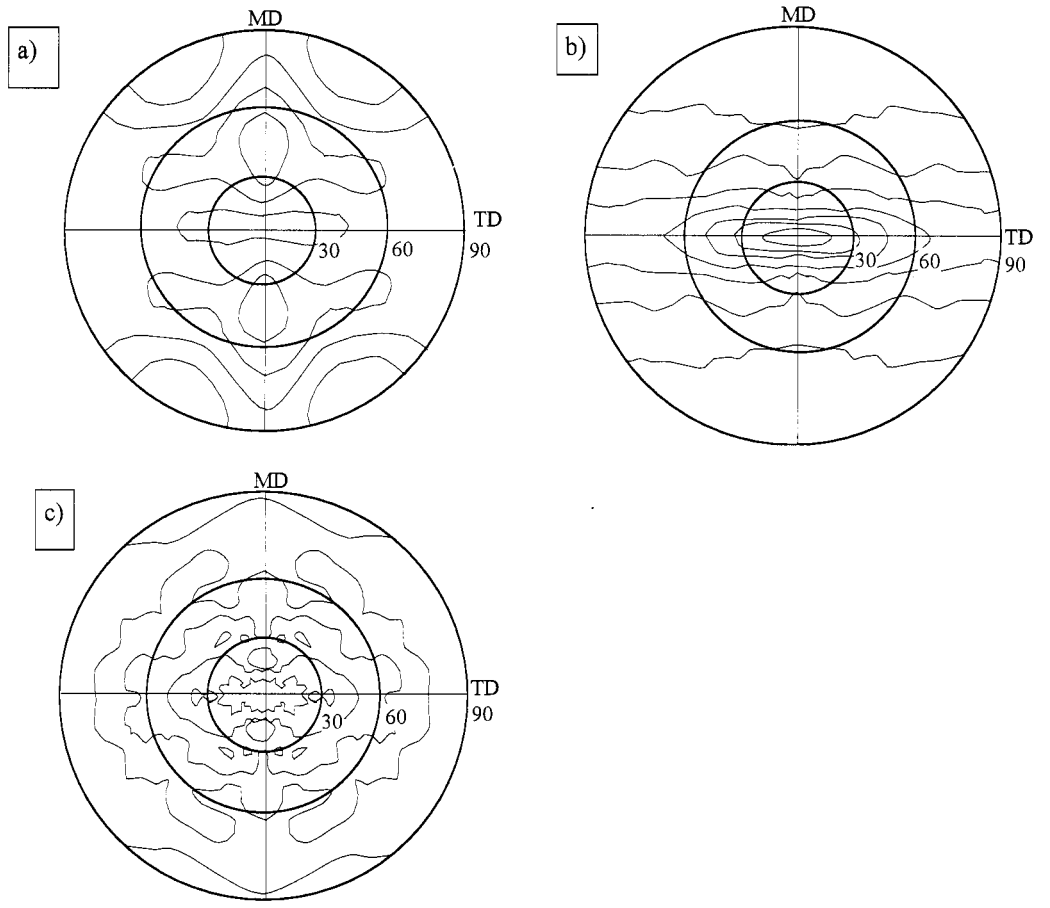

Figure 4. An example of the pole figures from the a) 110-, b) 040-, and c) 130- reflections of polypropylene from the layer at 2 mm depth in sample **PP/g 5 STATIC**.

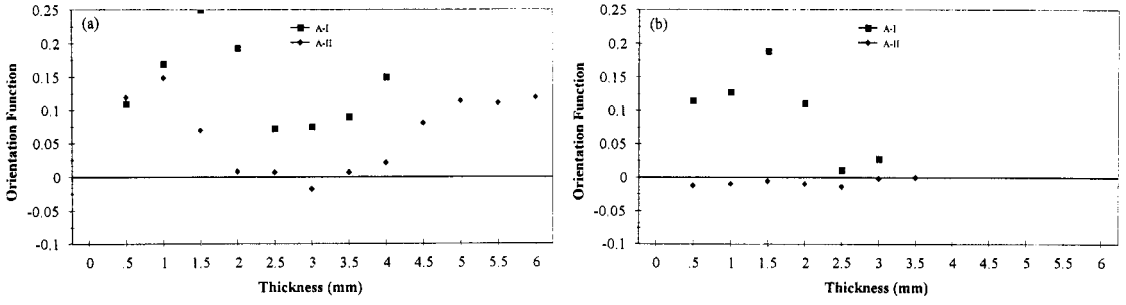

Figure 5. Hermans orientation functions of the chains $\mathbf{f}_c$ with respect to MD as a function of the distance **d** from the surface of the plaque. a) **PP/ g 5 STATIC** b) **PP/ g 10 STATIC** (refer to Fig. 2 for sample location).

and then decreases to almost zero in the middle layer (**d**=3 mm). In the case of both types of fibers the orientation of the sample I, which was taken from the centre of the plaque is greater than that in sample II from the corner. The orientation in these plaques containing the shorter fibers seems to be larger than in that with the longer fibers.

Figure 6 shows the corresponding Herman orientation functions $\mathbf{f}_{040,ND}$ of the normal to the 040 plane with respect to the ND direction. As one can see, close to the surface of the plaque, a slight orientation of the 040 plane parallel to this surface has been found.

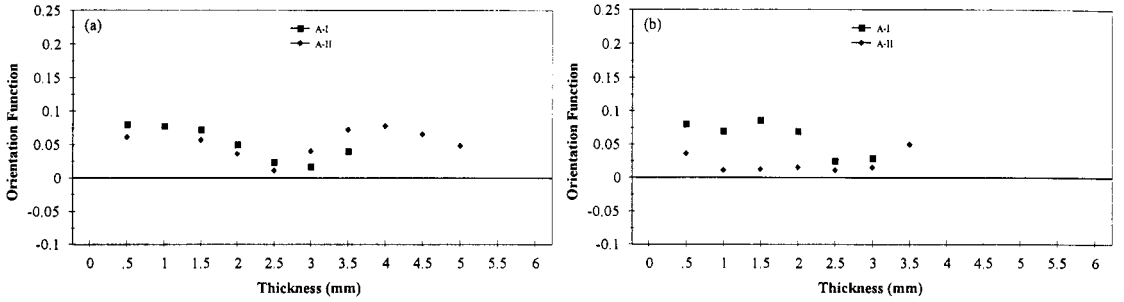

Figure 6. The Hermans orientation functions of the 040-reflection $f_{040,ND}$ with respect to ND as a function of the distance **d** from the surface of the plaque. a) **PP/ g 5 STATIC**, b) **PP/ g 10 STATIC** (refer to Fig. 2 for sample location).

In all of these orientation function plots through the specimen depth (or z-axis), the molecular orientation exhibits a significant drop in the central section, and this corresponds to significant frozen-in orientation in the molecules near the mould walls, and is in line with the observations made on fiber orientations.

3.2 Fiber Orientation Distribution (FOD)

3.2.1 Approach

The measurement of 3D FOD functions within a material is not a trivial exercise and usually necessitates use of Image Analysis techniques for quantitative and statistical data. Consequently, detailed Image Analysis (IA) studies have been carried out only on a limited number of material systems by laboratories possessing the requisite software and hardware (Laboratories 1 and 7). Semi-quantitative information, on the other hand, can readily be obtained from Light Microscopy (LM) or Scanning Electron Microscopy (SEM) images of polished sections of the materials. When sectioned, fibers of infinite length exhibit elliptical cross sections

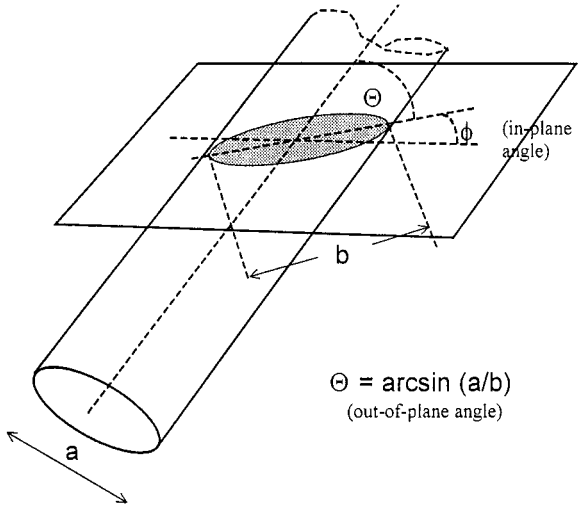

Figure 7. Description of the in-plane and out-of-plane angles.

such that in the sectioning plane, the orientation direction of the fiber is the same as the orientation of the main longer axis of the cross section, as shown in Figure 7.

In other planes, the extremes of orientation present a circular fiber cross section when the fiber is intersected normal to its length (90°) and a rectangular cross section when intersection is parallel to the length (0°). Orientations between these 90° and 0° result in ellipses with varying eccentricity.

It is clear that this method may provide an estimate of the in-plane orientation of the fibers

but does not allow interpretation of the out-of-plane angle of inclination of the fiber to the sectioning plane. This may be calculated from quantitative IA studies (**7**,**8**).

An alternative method of producing images of fiber orientation is using X-ray radiography. The specimen is cut into plates up to 3 mm in thickness and irradiated with the X-ray film in close contact with the specimen. Results were not quantified but qualitative assessments were made.

From SEM analysis of fractured surfaces of the composites, it is also possible to obtain information on FOD. However this is again only qualitative since the direction of the exposed fibers in the fracture plane is not well-defined. All the above techniques have been applied in this investigation, thus allowing the FOD to be characterised both into the depth of the 6 mm thick plaques and along the length of the plaques.

The effect of 5 mm and 10 mm initial fiber lengths on FOD was briefly investigated and did not appear to affect the observations significantly. Consequently, no attempt is made to distinguish between these lengths in the subsequent presentation of FOD observations.

A parallel study (**9**) has made measurements of the mechanical properties from wafer sections machined from identical samples. The fiber orientations observed in this study can be correlated to the wafer properties.

3.2.2 Fiber orientation distribution measurements for glass fiber reinforced polypropylene mouldings

Laboratory 5 polished transverse sections perpendicular to the melt flow for SEM analysis of FOD. Figure 8 represents schematically a model section perpendicular to flow and indicates the multi-layer structures observed for both DYNAMIC and STATIC mouldings central to the plaques. However, the DYNAMIC process resulted in a small core of fibers lying perpendicular (or misaligned with respect) to flow whereas the STATIC process produced a larger core zone. It was noted that some unexpected voiding was observed in both cases probably resulting from the elastic recovery associated with areas of fiber misalignment, as described in the flow studies undertaken on these materials in a parallel project (**2**).

Laboratory 4 analysed FOD qualitatively from X-rays of 1 mm thin strips and 3 mm thin circular plates. Again the DYNAMIC process showed the main portion of fibers present in strips lie parallel to the injection direction, although in circular plates the middle region appeared perpendicular to flow (in the sections taken, but was more probably random-in-plane).

In STATIC mouldings, the central fibers in the strips were clearly aligned perpendicular to the injection direction and near the walls the main fraction was perpendicular to flow. This

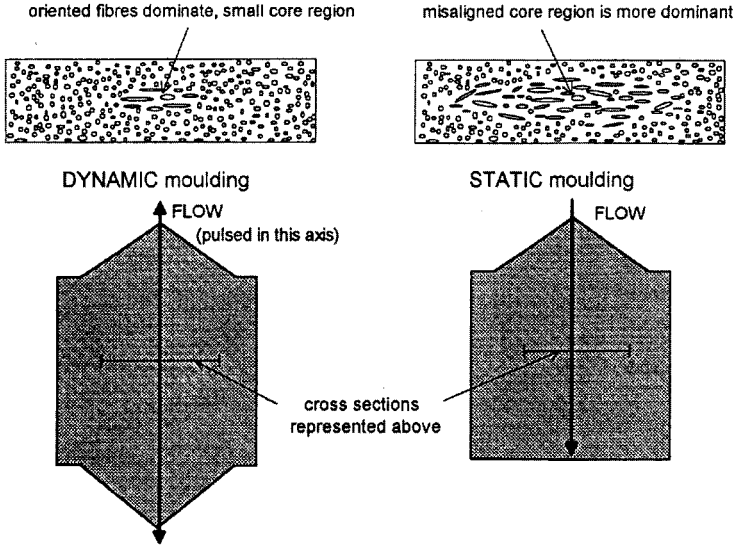

Figure 8. A schematic representation of the two moulded parts and the fiber orientation in the central section shown.

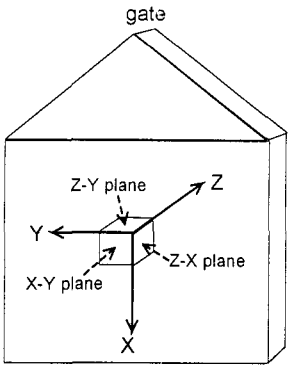

Figure 9. Definition of planes for the characterisation of the 3D fiber orientation functions

observation indicates differences in FOD resulting from different moulding conditions, but the degree of alignment in the skin is consistent with DYNAMIC process.

Laboratory 3 prepared sections from the centre of plaques both parallel and perpendicular to the mould fill direction which were subsequently examined by Light Microscopy, on the basis of which qualitative descriptions of structures were made. Multi-layer structures were observed in specimens from both processes. Dynamic processing generated a 1.6 mm thick random core surrounded by 2 layers at 1.5 mm fully oriented in the injection direction. In addition two 0.5 mm skin layers were apparent, although there was an indication of orientation perpendicular rather than parallel to flow. In total, approximately 50% of fibers appeared fully oriented in the injection direction.

In conventional (STATIC) mouldings, a thin 0.5 mm core was oriented perpendicular to the mould fill direction surrounded by 2 thick layers of random orientation. 10% of these fibers appeared oriented perpendicular to the injection direction.

A fully quantitative method of characterising FOD in 3 dimensions has been used by Laboratories 1 and 7. These methods involve the preparation of sections and viewing by either SEM or Light Microscopy. Sections of three planes may be used as shown in Figure 9. The elliptical sections of fibers presented in Figure 7 can be characterised by 2 angles in three dimensional space with reference to a known axis, which for convenience and uniformity is defined as the primary flow axis from the gate to the opposite wall of the mould cavity (this is easily defined in a plaque moulding). The orientation of the fiber in the sectioning plane ϕ, is equal to the orientation of the longer main axis of the fiber cross section. The angle ϕ describes the in-plane angle the fiber makes with the injection direction (X axis). The angle of inclination of the fiber to the sectioning plane is equal to the angle the fiber projected into the X-Z plane makes with the Z axis. The out-of-plane angle θ is calculated from the ratio of the 2 main axes of the ellipse as shown in Figure 7.

Laboratory 7 has studied PP/g plaques generated in STATIC moulding at 3 different locations along the flow path (it should be noted that the plaques used were of a different geometry with a greater length (>100 mm)). The plane perpendicular to flow (YZ) was chosen for analysis. These observations can be compared with the parallel study observations (**9**).

Study shows that close to the gate, the skin/core/skin boundaries are hard to define. Most of the fibers lie in plane with minimum out-of-plane orientation. Further along the length, the skin/core appeared better defined and the core well-aligned perpendicular to flow with the skin preferentially aligned in the injection direction. The end of the plaque was similar to the centre but with more out-of-plane orientation evident.

From the data generated, it may be calculated that in the central region of the plaque, the alignment of fibers in the skin was approximately 38° in-plane (to X axis) and approximately 13° out-of-plane. Towards the core, these values changed to 67° in plane and approximately 16° out-of-plane.

In summary, it appeared that the fibers lie preferentially in plane perpendicular to the direction of mould fill with an increase in transverse alignment as the distance from the gate increases.

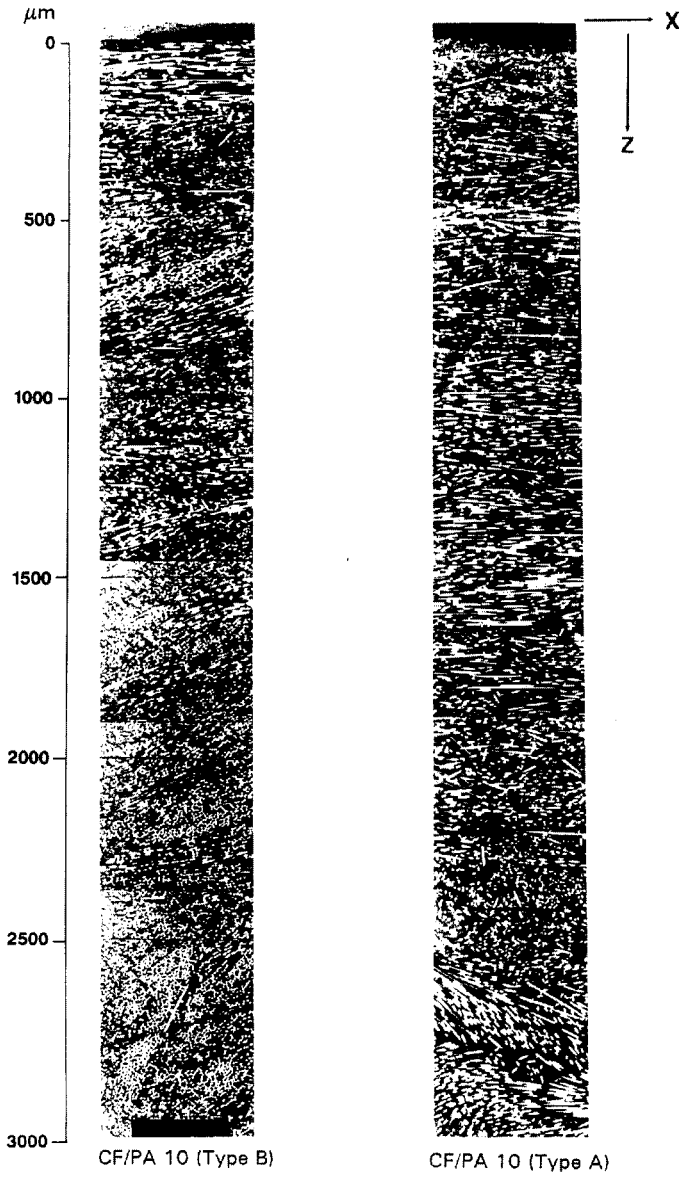

Figure 10. Micrograph sections showing the fibre orientations for **STATIC** and **DYNAMIC** processes in **PA/ c** composites. These sections are in the z-x plane at the centre of the moulded plaque.

3.2.3 Fiber orientation distribution measurements for glass fiber reinforced polyamide mouldings

Polished sections and fracture studies from Laboratory 5 indicated FODs similar to PP/g with again improved alignment apparent for process DYNAMIC over STATIC processing. Laboratory 3 however, suggested a slight difference in microstructure for PA/g using the DYNAMIC process. Although the skin/core structure is still evident, in DYNAMIC moulding a thin core of 0.6 mm was generated with the fibers fully oriented perpendicular to the injection direction. This was in contrast to the 1.6mm random core observed in PP/g. The surface layers appeared fully oriented in the injection direction and in total 90% of fibers were oriented with the flow (compared to 50% for PP/g). Static moulding, on the other hand, appeared to generate a similar structure for both PA/g and PP/g.

Most of the results from Laboratories 3, 4, 5 and 7 suggest little difference in the FOD between PP/g and PA/g systems. Variations in results may be due to slight differences in location of the sections since it is evident that the structure does alter along the length of the plaques.

3.2.4 Fiber orientation distribution measurements for carbon fiber polyamide mouldings

Semi-quantitative studies were made by Laboratory 6. Measurements along the centre line at 42mm from the gate indicated that DYNAMIC processing again produced the greater alignment of fibers. Although the centre plane by depth appears slightly random, more alignment was apparent than for STATIC moulding with 37% present as skin and a core of 63%. These STATIC moulded samples were difficult to separate into skin/core. Most out-of-plane orientation was observed in the core for both processes, although for DYNAMIC, no out-of-plane fiber orientation was observed up to 1.5 mm into the thickness of the sample.

Laboratory 7 examined samples generated by the STATIC process, again at 3 different locations along plaques. Close to the gate, the core is more random but fibers in the skin are preferentially aligned in the injection direction. As for PP/g, the skin/core effects are more marked with progression along the length of plaques such that the skin is still aligned in the injection direction but the core is more aligned perpendicular to flow. At the very end of the plaque, fibers perpendicular to flow predominate. There is no significant difference of FOD values in total along the length of the plaques, although it is clear that the spatial distribution does vary.

In terms of in-plane and out-of-plane (ϕ and θ respectively), for the central section, $\phi = 40°$ and $\phi = 19°$ in the skin; and $\phi = 63°$ and $\theta = 28°$ in the core. Comparing ϕ and θ values with those obtained for PA/g (STATIC) it is clear that the out-of-plane orientation is significantly increased for the PA/c samples.

Laboratory 1 have studied FOD structures for PA/c system comparing both processes using quantitative methods described above to characterise FOD in terms of ϕ and θ. Measurements were taken at the centre of the plaques along the centre line as shown in Figure 9. Analysis was carried out in the plane parallel to flow (XZ). Microstructural section are shown in Figure 10 for the two processes.

The study indicated that a classical skin/core structure did not exist for either process. Very high out-of-plane values (30°) existed in the core of the plaques (STATIC), although the skin was preferentially aligned in the injection direction for both modes. Dynamic moulding generated a more in-plane oriented core compared to random orientation observed for STATIC moulding. The thickness of the core region differed with 70% core/30% skin indicated for STATIC mouldings whereas DYNAMIC processing generated a structure more uniform across the whole thickness. The table below shows the orientation angles for the two moulding routes for central skin and core locations.

	ϕ	θ
DYNAMIC skin	30°	13°
DYNAMIC core	49°	27°
STATIC skin	37°	15°
STATIC core	52°	30°

Table 1. The average in-plane (ϕ) and out-of-plane (θ) fiber orientation angles for the two processing techniques in the two section locations.

Comparison of STATIC data generated at Laboratories 1 and 7 indicate that although results compare reasonably well, some deviation is evident. This may be expected since different planes were analysed, the regions were not identical and Laboratory 7 incorporates a function to correct for intersecting fibers exhibiting different out-of-plane orientations (7). However both sets of results do confirm strongly the increased out-of-plane fibers present in this system.

As other results have shown (**4**, **9**), the fiber orientation structure may vary along the length of the plaque, PA/c samples appeared particularly non-uniform in structure. This may be due to a number of possible factors (compared to glass, carbon fibers have a lower density,

smaller fiber diameter) which may lead to the randomisation of fibers relative to the mould fill direction. The small volume of the mould is unlikely to provide a reasonable opportunity for steady laminar flow to be established, but this is unlikely to differ from glass bearing melts. In addition, the fiber/matrix affinity is believed to be highest for the composites studied.

3.2.5 Summarising Comments from FOD

A number of qualitative observations emerge:
PA/g and PP/g systems behave similarly with well defined skin/core structures.
PA/c system appears more random in structure which may be associated with more turbulent flow during mould filling.

In all cases, DYNAMIC moulding provides improved alignment of fibers in the direction of mould fill resulting in a smaller core region.
In addition, the following observations can be made:
i) Image Analysis allows direct access to in-plane and out-of-plane fiber angles.
ii) For STATIC moulding, the PP/g system contains defined skin/core regions with little out-of-plane orientation.
iii) In the STATIC process mouldings, the PA/c system has a significantly greater out-of-plane orientation distribution.
iv) Comparison of samples from both moulding types confirms the greater alignment of fibers through the thickness of the specimen for DYNAMIC moulding.

3.3 Assessment of Fiber Content.

Fiber content was measured for a number of systems and results are shown in Table 2 below. The glass composites were ashed and a fiber content for the total sample was derived. The results appeared uniform at approximately 20% by volume.

Two methods were used to measure the fiber content for the PA/c composites. From the Image Analysis method, the results indicated that the core contained a higher percentage of fibers compared to the skin.

Significantly, both methods indicate that the STATIC process produces samples which are more heterogeneous in fiber dispersion such that the core is more heavily filled with fibers compared with DYNAMIC moulding.

system	*process*	*skin*	*core*	*total*	*method*
PA/c 5	DYNAMIC	28 (37.4)	31 (40.8)	-	image analysis
	STATIC	25 (33.8)	34 (44.2)	-	"
PA/c 10	DYNAMIC	-	37.6 (48.04)	-	dissolution in m-cresol
	STATIC	-	43.2 (53.84)	-	"
PA/g 5	DYNAMIC	-	-	21 (37)	ashed
	STATIC	-	-	20.5 (36.5)	"
PA/g 10	DYNAMIC	-	-	20 (36)	"
	STATIC	-	-	20.5 (36.5)	"
PP/g 5	DYNAMIC	-	-	20.5 (42)	"
	STATIC	-	-	20 (41)	"
PP/g 10	DYNAMIC	-	-	20 (41.5)	"
	STATIC	-	-	18 (38.5)	"

Table 2. Fiber content by volume % (weight % in brackets).

3.4 Assessment of Fiber Length Distributions

3.4.1 Methods and Presentation of Results

A number of methods exist for determining fiber lengths in reinforced polymers. In this multi-laboratory study we have used the various methods available in the participants' laboratories, rather than determine or even select the most appropriate method. In injection moulded parts it is preferable to describe the fiber lengths present by more than one parameter. In any distribution, it is valid to calculate the number ($\overline{L}_n$ and weight ($\overline{L}_w$ average lengths, and these are calculated using the convention below:-

$$\overline{L}_n = \frac{\sum n_i l_i}{\sum n_i}$$

$$\overline{L}_w = \frac{\sum n_i l_i^2}{\sum n_i l_i}$$

where n_i = the sample frequency with the length increment range l_{i+1} - l_i.

These two mean values if quoted together can be used to describe the distribution in the same way as is applied to molecular weight data. The number average is always the smaller value and is strongly influenced by any small fibers and fragments present, while the weight average is heavily influenced by long fibers. This weight average is significant in these materials since their whole raison d'être is to deliver a portion of longer fibers to the moulded part. However, it is the number average length which is used for comparative purposes in the industry. Moreover, it is this term which is used for property prediction models. Nevertheless, a number average length can be misleading when there is a broad distribution of a non-Gaussian type (**10**).

The measurements of fiber length distributions relate to observations from specimens taken from the exact centre of the plaque mouldings.

Across the participating laboratories, the measurements were made with variations in technique. All laboratories pyrolysed the composite at around 500 °C in air to release the fibers. This technique is ideal for glass fibers where it is generally accepted that below 600 °C fibers are not embrittled. In liberating the carbon fiber composites, dissolution (Formic acid or m-Cresol solvent for polyamides) is the preferred method of releasing the fibers. However, these materials were also prepared by pyrolysis with no deleterious effect on the fibers by oxidation.

Some laboratories have developed a sieving classification step which attempts to separate the longest fibers from the shorter class of fibers (**11**). This step increases the subsequent measurement problem of dealing with such a wide distribution of fiber lengths. Usually this step is accompanied by a weighing operation which provides useful additional information such as giving a proportion of sieved fibers which exceed the sieve dimension.

Once released from the matrix, or after any sieving operation, the fibers are transferred to a microscope either manually or in a dispersion medium. Laboratory 3 used acetone which was allowed to evaporate, thus leaving fibers separate in the microscope field of view. A low magnification is advantageous since it allows a field of view in the microscope which can accommodate a 10 mm length fiber. However this does impair the accuracy of the measurements and reduce resolution.

Image analysis (IA) (either interactive or fully automatic) was used by all participants. Sawyer (**12**) has reviewed the application of this technique to fiber length determination. Some IA instruments have limitations depending on the measurement algorithms they employ. Some microscope configurations are restricted to 3 mm maximum particle size, or are unable to measure curved fibers, as they assume linearity between the fiber ends. In these cases the fiber lengths have been measured in a two stage process encompassing a manual stage for the longest fraction of fibers.

The results of fiber length measurements are summarised in Table 3.

In the carbon fiber reinforced polyamide materials, number average fiber lengths revealed that the fiber lengths resulting from both processing routes gave similar values (0.24 mm for PA/c 10 DYNAMIC and 0.27 mm for PA/c 10 STATIC).

3.4.2 Discussion of Fiber Lengths.

The family of statistical data presented in Table 2 indicates just how difficult it is to attain consensus between fiber length measurement techniques from different laboratories. The common parameter, number average fiber length, is relatively insensitive to changes in distribution because number frequency is used to calculate the term; the short fibers which are in high numbers dominate the result as anticipated, even though they may represent a small weight or volume fraction of the fibers present. The mechanism for attrition in these materials is reported to be a sweeping shear action against the barrel of the injection moulding machine as the granules melt (**13**). This gives rise to lengths of 0.5 mm or less and this is shown in the Figure 11, (Laboratory 6). In conventional injection moulding this behaviour provides a fiber length distribution which characteristically is dominated by the peak described at <1 mm. These data can normally be described by a log-normal shape. The high length tail (>1 mm) is important as these fibers will contribute most to the mechanical performance (especially the strength). In the multi-live feed

DYNAMIC - pulsed injection moulding

Laboratory	PP/g 5 DYNAMIC	PP/g 10 DYNAMIC	PA/g 5 DYNAMIC	PA/g 10 DYNAMIC
Laboratory 3				
(number average)	0.5	0.5	0.6	0.7
500 fibers				
Laboratory 5				
(number average)	0.71	0.66	0.54	0.49
% fibres >3 mm	<1	<1	2	<1
longest fiber	4.5	3.7	3.7	3.5
1000+3000 fibers				
Laboratory 1				
(number average)	0.62	0.59	0.55	1.38
wt./vol. average)	0.83	0.79	0.88	3.73
% fibres >1 mm	15	14	14	71

STATIC - conventional injection moulding

Laboratory	PP/g 5 STATIC	PP/g 10 STATIC	PA/g 5 STATIC	PA/g 10 STATIC
Laboratory 3				
(number average)	1.2	2	0.6	2.3
500 fibers				
Laboratory 5				
(number average)	0.78	0.88	0.54	0.66
% fibres >3 mm	2.6	27	0.4	18
longest fiber	6.7	9.0	6.7	8.8
1000+3000 fibers				
Laboratory 1				
(number average)	1.44	2.47	1.43	2.19
wt./vol. average)	2.87	4.97	2.6	4.73
% fibres >1 mm	14	84	79	83

TABLE 3. Summary of fiber length distribution results

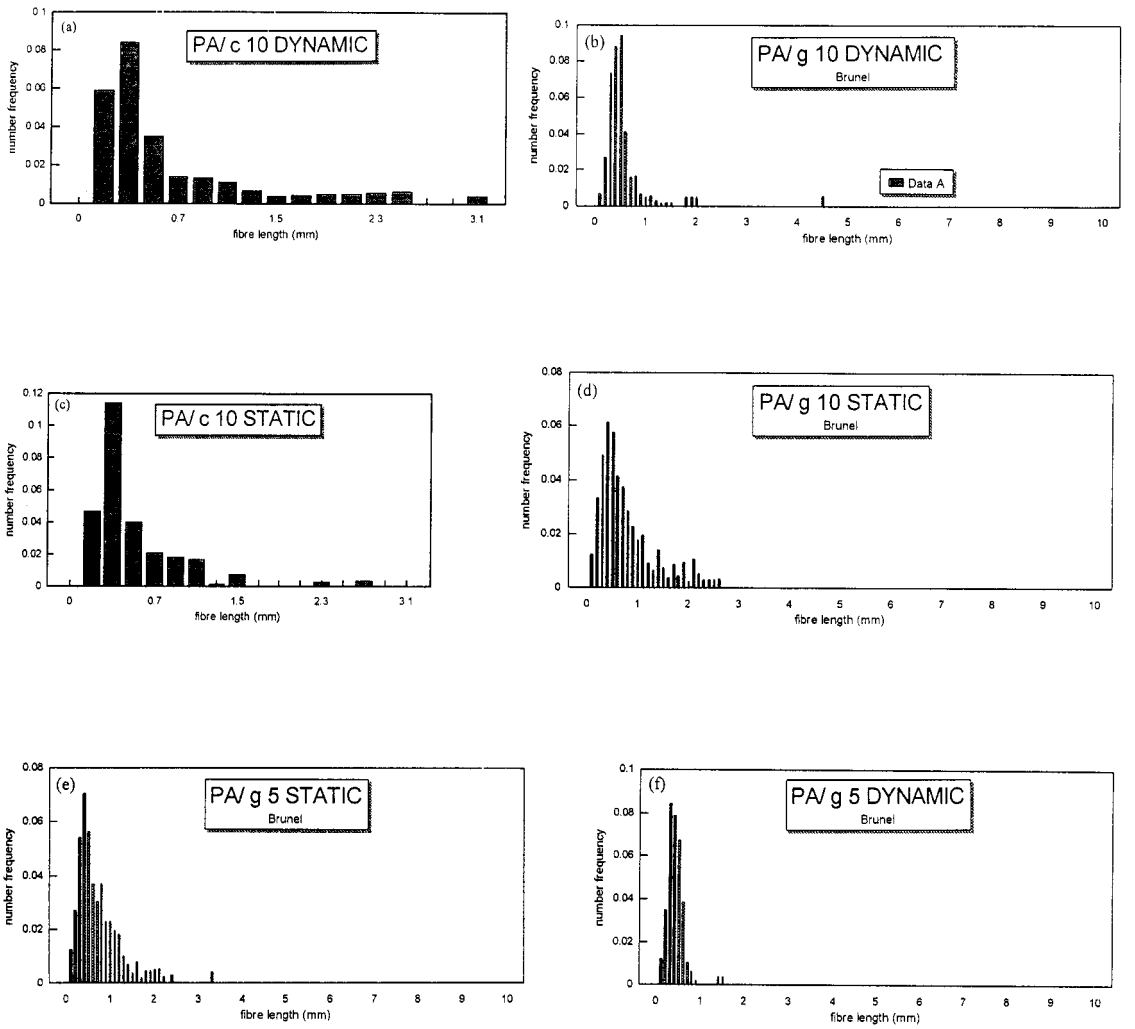

Figure 11. (a - f) Fiber length distributions plotted for polyamide composites (Laboratory 6). The carbon fiber materials are shown with a different fiber length axis. It is clear that the dynamic moulding technique reduces the fiber length statistics compared to the conventionally moulded static process.

injection moulding samples, the fiber attrition has been increased and the mechanism appears to give rise to fibers of similar lengths to the 0.5 mm peak which has generally been reported as arising from fiber breakage through shearing of the melt in the course of screw plasticisation during processing. This implies that the shear experienced by the fibers in the mould cavity is similar to that experienced during the screw plasticisation. Figure 11 illustrates that the higher length tail in the distribution is severely affected (a reduction of a factor of up to 2 in the number average) and this effect is observed in all the materials under study.

The glass fibers are preserved in longer lengths than the carbon fibers. Carbon fibers are of finer diameter and are stiffer. It is generally observed that carbon fibers suffer higher levels of attrition under equivalent melt processing. The results obtained would be consistent with this trend. The granule length seems to have little effect on the number average length: in some cases this parameter was actually higher for the 5 mm granule than the 10 mm equivalent. This indicates that the level of experimental scatter may be high or confirms that the processing conditions were

variable between mouldings. However, the term described as [%>1 or 3 mm] indicates that the 10 mm granule materials retains the greater volume of longer fibers as expected. It is generally observed that longer fibers are retained in polypropylene systems than in polyamides. The pulsed moulding was optimised in order to align the fibers and this may have lead to differences in the fiber length distribution.

This raises the issue of any interrelation between fiber lengths and fiber orientation. Overall, it is evident that in the multi-live feed process, a higher alignment results in shorter fibers because of the additional shear imposed. It is clear that the polymer melt containing the fibers, which arrives at the mould surface first and is frozen there forming the skin, will be similar for both processing routes. Subsequent material filling the mould, will differ between the two processes. In the case of the multi-live feed melt material entering the mould cavity, additional shear forces are imposed which increase as solidification takes place. In conventional moulding the last material to enter the mould is that found in the core region, where fibers are misaligned but have experienced little shear during mould filling and therefore, the core region of STATIC moulding can be expected to contain the longest fibers.

4 CONCLUDING COMMENTS.

We have attempted to describe the way by which the microstructure of discontinuous fiber composites can be articulated. Microstructure should be described by at least three parameters, namely fiber orientation distribution, fiber length distribution and fiber content. It is quite apparent that these are not straightforward measurements and that progress towards simplifying such observations into a skin-core description of heterogeneity through the thickness of a moulding is a gross oversimplification. The fiber length, local fiber fraction, fiber diameter and orientation are all unique for every fiber location in the moulded part, and consequently averaging the measurements is a step in the direction of losing data significance.

Quantitative data have been derived for the purposes of modelling the properties and this is the subject of a further paper (**14**) from this study.

It is shown in this work that there is a profound influence of the processing history on the microstructure for this family of materials, and there is a considerable challenge in measuring the macro-, meso and microstructures. It is clear that across the laboratories involved in this work, there is consensus in establishing trends, but difficulty in agreeing quantitative results. This is perhaps not surprising and is a symptom of the complexity and local variability of the microstructure.

The correlation between the fiber orientation structure and the inferred molecular structure is therefore likely to be even more difficult to undertake. The molecular orientation functions shown through the specimen thickness are however most revealing. The results independently confirm the general underlying skin-core model which is thought to underpin the processing-structure-properties relationship for these composite materials.

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