The Effect of Impregnation on the Microstructure of Long Fibre Reinforced Thermoplastic Composites and their Consequence on Deformation and Toughness

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CONTENTS

	Page
ABSTRACT	172
1. INTRODUCTION	172
2. SAMPLE PREPARATION	173
3. MORPHOLOGICAL CHARACTERISATION	174
3.1. Pultrudate	175
3.2. Injection Moulded Plaques	176
3.2.1. Fibre Dispersion	176
3.2.2. Fibre Length Distributions	177
3.2.3. Fibre Orientation Structures	182
4. MECHANICAL PROPERTY MEASUREMENTS	183
4.1. Toughness Characterisation	184
4.2. Tensile Dilatometry	185
4.2.1. Theory for Volume Change during Deformation	185
4.2.2. Experimental Measurements Using Tensile Dilatometry	186
4.3. Creep and Recovery	189
5. CALCULATION OF STIFFNESS	192
6. CONCLUDING COMMENTS	192
7. ACKNOWLEDGEMENTS	194
REFERENCES	194

ABSTRACT

Long fibre reinforced composites can be manufactured by a variety of processing routes. In this study the effect of varying the method of feedstock production (from a pultrusion type method to a wire encapsulation type method) has been monitored in relation to the microstructural characteristics and the mechanical properties of the two compounds. The microstructural characterisation showed clear differences in the degree of fibre dispersion and volume fraction for the compounds. Mechanical tests dominated by the response of the fibres in each material showed little difference between the compounds. An extension to the theory of volume strain analysis has enabled the distinction between normal and cavitation type mechanisms in a material under deformation. Using a volume strain approach, the wire coated type material showed a pronounced cavitation with increasing deformation, a result confirmed by subsequent creep and recovery experiments, whereas the pultrusion type material showed a more conventional response.

KEYWORDS

Long fibre composites; mechanical properties; deformation, toughness, injection moulded compounds

1. INTRODUCTION

Fibre reinforcement of thermoplastics provides added-value properties. The addition of fibre reinforcement in conventional extrusion compounded feedstocks infers enhanced stiffness, strength and toughness. Therefore, optimisation of this enhancing mechanism should be profitable and to this end manufacturers have developed technology for the provision of longer fibre reinforcements /1/, thus moving towards an optimisation of properties per unit cost. Inevitably, there are choices in terms of how such long fibre feedstocks for injection moulding can be prepared and there are well patented approaches in terms of how such technologies can be achieved.

Nevertheless, the granular feedstock itself and an injection moulded component from the feedstock can be made available. It is therefore quite appropriate to ask the question as to how one can judge the quality of the feedstock/moulding in objective terms and in so doing to seek confirmation that the properties per unit cost have been optimised.

A judgement on the quality of feedstock/moulding should not be an artificial distinction between "good" or "bad". It would of course be possible to generate production optimised materials for comparison with an amalgam of all that can be imagined to be bad practice in production. Such a comparison would be both possible and pointless since only one of the materials carries any credibility. A much more informed comparison would emerge by the preparation of "production possibilities": materials that are made on a production line and where both represent a possible approach to manufacture of the feedstock. Indeed, it might even be possible that such materials represent competitive alternatives through different approaches to production methods by different manufacturers.

The purpose of this paper is to create two "production possible feedstocks" in the form of continuous glass fibre reinforced nylon 6,6 from the same production line with the same starting materials. This feedstock will then be chopped to granules of 10mm length and used as the feedstock for the preparation of some plaque injection moulded pieces. The granules and the plaques will then form the subjects of a study to establish and discuss the types of parameters that are important in achieving a "high quality" product. The differences in the feedstock will relate to the degree of fibre dispersion during the precompounding process. This is achieved by operating a pultrusion compounding unit in two regimes. First, the compounding line was set up to achieve a pultrudate where the fibres are uniformly dispersed. Second, the line was set up to produce a wire-coated or encapsulation type pultrudate which contained undispersed fibre bundles. The details of sample preparation are given later.

The objective of the study will then be to measure and describe characteristics of both the granular feedstock (i.e., prior to injection moulding) and the properties and morphology of the injection moulded

parts in order to articulate the character of the materials. In particular, it is desired to establish what features are beneficial and what features are disadvantageous. One of the areas of interest will be the link between the original materials and the properties of the injection moulded parts. We will be attempting to address the issue of whether the two stages of processing, namely precompounding and injection moulding, progressively dilute the early choices and decisions in manufacture or not.

One of the complications of such a study is bound up in the anisotropic and heterogeneous nature of injection mouldings prepared from discontinuous fibre composites /2/. It is well known that injection mouldings constitute a layered structure through their thickness /3/. The surface "skin" layers contain fibres that are mainly aligned with the direction of mould fill and the "core" region contains fibres approximating to 90° with respect to the mould fill direction. Moreover, the ratio of thickness for the "skin" to "core" layers depends on several parameters such as processing, mould thickness, fibre volume fraction and other factors /4/.

A further complication arises from the reduction in fibre length as the continuous fibre granule is processed in the injection moulding machine. In fact there is a change from a distinct length to a distribution of fibre lengths /5/. Assessment of an injection moulding should therefore accommodate a measurement of this fibre length distribution function. Moreover, it is important to comment on the mechanical performance of the moulding in relation to the mean fibre length in the moulding relative to the critical fibre length for the material.

Any general assessment of the behaviour of an injection moulded component must reflect the actual nature of the moulding. Therefore, we believe that our characterisation of the properties of the injection moulded plaques must reflect the involved nature of their mechanical behaviour in order for us to focus on aspects of behaviour that depend on the nature of the feedstocks. With this in mind we have selected quite a wide and detailed cross-section of tests that aim to characterise mechanical performance. Inevitably, some of these tests are dominated by the fibre orientation within the mouldings where the process dependent features cannot be resolved. Other tests do

enable the process dependent features to emerge. We have deliberately accommodated both types of test in this study, because it is seldom self-evident that any particular test can resolve process dependence at the outset.

The work is presented in three parts:

- (i) The preparation of the pultrusions and their characterisation in terms of fibre dispersion and matrix impregnation.
- (ii) The preparation of the injection moulded plaques and their morphological characterisation in terms of fibre content, fibre orientation distribution, through thickness heterogeneity, fibre length analysis and impregnation.
- (iii) Mechanical characterisation of the mouldings in terms of stiffness and toughness.

In all aspects of the work it is our intention to establish a property-structure link in the evaluations with the specific aim of understanding performance of the materials. This in turn leads to a clear indication of those features of the feedstock that are desirable in end product performance.

2. SAMPLE PREPARATION

The objective of this work was to produce and compare materials which exhibited extremes of fibre dispersion as a result of the precompounding operation. In order to achieve this, pultrusion compounding was used in two régimes. Firstly the compounding line was arranged in order to produce pultrudates containing fibres which are uniformly distributed through the circular cross section of the rod.

The second configuration was arranged in order to produce a wire-coated or encapsulation type pultrudate which contained undispersed fibre bundles. In producing such materials a lower bound exists for the degree of retaining the integrity of the fibre bundles in the pultrudate. This boundary condition is dependent upon the subsequent chopping stage to granules for injection moulding. Below a critical level of fibre dispersion and wet-out the pultrudate cannot be cut satisfactorily into granules.

It is the aim of this exercise to produce two pultrusion compounded materials which offer as

diverse morphological attributes as possible whilst employing the same raw materials and compounding technology.

Two nominal 50% w/w (35% by volume) E-glass fibre reinforced nylon 6.6 compounds were prepared by pultrusion compounding and were chopped to 10mm granule lengths for subsequent injection moulding. The E-glass fibres are nominally 17 μ m in diameter.

The materials are identified as "dispersed" fibre compound and "undispersed" fibre compound.

Injection moulding was carried out using a Battenfeld 700/300 machine with a screw diameter of 45mm. The injection moulding conditions were optimised for both materials in order that the moulding cycle was as similar for each as possible. The mould cavity employed was a 160 x 160 mm x 3mm thickness coathanger gated plaque, the test piece used is shown schematically in Fig. 1. The moulding

conditions were selected in order that the test pieces would retain desirable fibre length distributions by minimising the screw back pressure during the preplasticising stage of the cycle. The moulding conditions are tabulated in Table 1.

Table 1. Moulding conditions for both the "dispersed" and "undispersed" compounds for the Battenfeld 700/300 injection moulding machine

Settings	Compound Types	
	"dispersed"	"undispersed"
Barrel temperatures [°C]		
Zone 1	285	285
Zone 2	290	290
Zone 3	290	290
Zone 4 (nozzle)	295	295
Mould	90	90
Pressures [%]		
Injection	50	50
Holding 1	35	35
Holding 2	40	40
Back	5	5
Injection speed [%]		
	50	50
Times [s]		
Holding pressure 1	5	5
Holding pressure 2	15	15
Screw rotation [dig.]		
	5	5

Fig. 1: Schematic representation of the coat hanger plaque, used in all of the tests.

3. MORPHOLOGICAL CHARACTERISATION

It is anticipated that the moulding process induces further impregnation of the fibres. In order to monitor this process, and indeed to identify any additional factors, the plaque test pieces were subjected to similar characterisation techniques as the feedstock materials for fibre dispersion.

The test piece morphologies were further characterised by studying material taken from the exact centre of the plaque. This location coincides with the region used in the mechanical test program.

3.1. Pultrudate

The pultrudates were characterised by two techniques:

- i) Immersion ultrasonic C-scanning.
- ii) Fibre dispersion by image analysis from scanning electron micrographs of polished sections.

Determination of fibre dispersion on a macro-scale can be obtained from an ultrasonic immersion C-scanning method. The ultrasonic immersion C-scanning instrument employed was a Branson Wells Krautkramer. The scans produced are threshold representations in two dimensions of the local ultrasonic signal attenuation level. A 6" x 3" panel of the pultrudate materials were compression moulded

using a very mild compression moulding consolidation cycle in order to assess impregnation in the pultrudate by the ultrasonic C-scan. Figures 2 and 3 show the scans for the two materials under study; the legend at the side of the raster scan showing the corresponding signal attenuation bands (dB).

The uniform shaded area of Fig. 2 is indicative of a relatively consistent and high level of impregnation of the fibres. The patchy C-scan of Fig. 3, however, is an indication of some poor and certainly variable impregnation of the fibres. The mild consolidation procedure has had little influence on the level of impregnation and consequently we may relate these observations to the impregnation of the original pultrusions.

Fibre dispersion by image analysis was conducted on pultrudate rods mounted in room temperature curing epoxy resin and metallographically polished using a Struers Pedemin (220, 1000 grit SiC paper, 6 and 1 μ m diamond polish). The subsequent specimens were gold sputter coated and micrographs prepared by scanning electron microscope (Hitachi S520). In this

Fig. 2: Ultrasonic immersion C-scan plot for the "dispersed" compression moulded pultruded rod.

Fig. 3: Ultrasonic immersion C-scan plot for the "undispersed" compression moulded pultruded rod.

way it was possible to enhance the contrast between the fibres and the polymer matrix sufficiently to apply image analysis techniques (Seescan Solitaire) to make local area fraction measurements in the composites on the microscale. The micrographs are shown in Figs. 4 and 5. In order to measure the local area fractions and hence the degree of fibre dispersion a grid was imposed over the micrographs and the measurements made within each square. The resultant area fractions, once normalised by the area of composite within each square, was plotted into histograms as shown in Fig. 6. The two histograms represent considerably different area fraction distributions. The "dispersed fibre" sample exhibits a Gaussian distribution around a mean of 35% area fraction, while the undispersed fibre" sample exhibits a wider, less Gaussian distribution with a mean area fraction of 38%; this is interpreted as being significantly higher. It is clear from these figures that the "undispersed" material contains dramatically more fibre rich and resin rich regions than the "dispersed" material, as expected.

3.2. Injection Moulded Plaques

3.2.1. Fibre Dispersion

In order to determine the effect of the injection moulding process on the degree of fibre dispersion in the mouldings, the C-scan and area fraction by image analysis techniques described above were applied.

The C-scan measurements were made using the same conditions and are shown in Figs. 7 and 8. As before, it is clear that the level of impregnation of the dispersed sample is higher than that for the undispersed sample.

Fibre dispersion assessments by the image analysis technique (area fraction measurements) were also made at the mid-point of the test plaques (corresponding to the location for all other measurements) and were obtained at 90° planes with respect to the flow direction. Such measurements can often give misleading results owing to the extrapolation of areas to volumes, although the

Fig. 4: Scanning Electron Micrograph showing the fibre dispersion through the thickness of the "dispersed" pultruded rod. The grid has been superimposed to allow local area fraction measurements.

measurements do correlate well with volume fractions calculated from weight loss measurements during an ashing procedure. It is also possible to compare the effect of fibre dispersion within the skin and core regions in the microstructure. The micrographs are shown in Fig. 9 and the corresponding histograms in Fig. 10. Fig. 11 shows the local area fractions measured through the section thickness.

Unlike the area fraction measurements made on the pultrudate sections, the moulding sections exhibit a more similar Gaussian distribution for both materials. One noticeable feature is the fibre rich values for the "undispersed" material. These relate to unimpregnated fibre bundles retained through the

entire injection moulding cycle. In Fig. 11, the local area fractions are displayed. The trend is for the core to be slightly higher than the average area fraction.

Once again, the area fraction for the "undispersed fibre" sample is significantly higher than for the "dispersed fibre" sample.

3.2.2. Fibre Length Distributions

Fibre lengths were measured by the image analysis/optical microscopy method (Seescan Satellite/Olympus Stereo Microscope) from fibres

Fig. 5: Scanning Electron Micrograph showing the fibre dispersion through the thickness of the "undispersed" pultruded rod. The grid has been superimposed to allow local area fraction measurements.

released from the polymer matrix by pyrolysis. The results are given in Table 2 and the fibre length distributions are shown in Figs. 12 and 13. The fibres remaining from pyrolysis of this type of material retain their structure as imposed by the injection moulding process. As a result, the skin and core can be identified as distinct layers and fibres may be extracted from each region with ease. This serves to increase the relative accuracy of the fibre length results and to provide more morphological information regarding the degree of fibre attrition

resulting from the shear history, and enables the local fibre length data to be used directly in modelling the microstructure.

The fibre length distributions are very similar for the two materials. The data are within experimental error of the measurement indicating that the moulding cycle caused similar levels of fibre attrition in each material. The correlation between the skin and core regions is in agreement with the typical trend with core fibres on average being longer than the skin fibres.

Fig. 6: Histogram showing local fibre area for both the "dispersed" and "undispersed" pultruded feedstock.

Fig. 7: Ultrasonic immersion C-scan plot for a typical "dispersed" moulded test plaque.

Fig. 8: Ultrasonic immersion C-scan plot for a typical "undispersed" moulded test plaque.

Fig. 9: Scanning Electron Micrograph showing the cross section through both the "dispersed" and "undispersed" plaques about the centre plane.

Fig. 10: A plot of the fibre volume fraction taken from moulded plaques (see Figure 9) showing the various of fibre loading through the thickness. In both materials the core appears to have a higher fibre fraction.

Fig. 11: A plot of local area fractions taken from moulded plaques (see Figure 9) showing the variation of fibre loading through the thickness. In both materials the core appears to have a higher fibre fraction.

Table 2. Number average fibre lengths (in mm) in both the skin and core regions for the "dispersed" and "undispersed" compounds

Compound Type	Skin Number Average (mm)	Core Number Average (mm)
"dispersed"	0.86	1.59
"undispersed"	0.79	1.51

Table 3: Percentage core in the "dispersed" and "undispersed" compounds taken from the micrographs in Figure 9.

Compound Type	% Core
"dispersed"	24
"undispersed"	23 - 26

3.2.3. Fibre Orientation Structures

Skin-core ratios: The orientation structure can be considered in two parts, firstly the extent of the skin-core structure, and secondly the orientation with these régimes. For the purposes of simplification and later modelling this can be represented by a number of layers representing half the moulding thickness (i.e., twice this number of layers for the full moulding). The definition of these layers and, more particularly, the orientation of a fibre within each layer, is simply to describe the fibre orientation function through the thickness of the moulding. This is required in order to model and describe by theory the stiffness of the moulding /6/ for comparison with measurements. This is discussed in a later section.

The orientation of the fibres will be significantly different in the skin and core regions. Therefore, the relative sizes or proportions of these regions should be defined by measurement before the number of layers of fibre orientation can be defined. Table 3 shows the percentage of core morphology for the two samples.

Fig. 12: A histogram showing the fibre length distribution in the skin and core regions for the "dispersed" compound.

Fig. 13: A histogram showing the fibre length distribution in the skin and core regions for the "undispersed" compound.

Fibre orientations within the skin-core structures can be measured by image analysis. In order to aid modelling of the mechanical properties and to accommodate the relative skin to core sizes, the structure can be subdivided into 16 layers, which are assumed to be symmetrical around the centre line of the plaque moulding. Therefore, only 8 layers need definition of the fibre orientation. The results of these orientations are shown in Table 4. The angles ($\theta_{1 \text{ to } 8}$) are relative to the direction of mould fill.

To summarise the morphological investigation, the results indicate that the microstructure arising from the injection moulding process, i.e., the fibre length and skin-core structures, are approximately the same for both undispersed and dispersed materials. The different precursor pultrudates were characterised and confirmed as being significantly different in "impregnation" in terms of local area fraction and C-scan results. However, in the moulded plaques, these measurements confirm that the difference in fibre dispersion is still significant despite the tendency of the injection moulding cycle to homogenise the compound.

4. MECHANICAL PROPERTY MEASUREMENTS

The consequence of the degree of impregnation at the

pultrusion stage has been examined in terms of its influence on mechanical properties of injection moulded samples. To this end, a range of stiffness and toughness experiments have been performed by selection and control of test features that enable resolution of the level of deformation and the failure process. These principally involved fracture toughness

Table 4: Fibre orientation distribution through the half-thickness of a typical injection moulding showing symmetry about the centre of the specimen.

Layer	Average orientation angle w.r.t. injection
Outer surface	
θ_1	30°
θ_2	20°
θ_3	20°
θ_4	10°
θ_5	20°
θ_6	30°
θ_7	50°
θ_8	65°
Centre of section	

measurements at initiation and during crack propagation, tensile deformation measurements and creep and recovery experiments.

In recognizing the possible influence of moisture on these polyamide 6,6 compounds, all samples were stored dry prior to testing. All measurements were made at 23°C.

4.1. Toughness Characterisation

The toughness of the materials was measured by two quite separate methods: instrumented falling weight impact (IFWI) and a linear elastic fracture mechanics (LEFM) test. IFWI provides a measure of impact toughness by simply measuring the energy absorbed by a specimen in achieving some specific failure or fracture event. This is conducted on some specific geometry and the results reflect the selected geometry. The LEFM approach measures some aspect of toughness which is independent of the test geometry.

IFWI tests were performed on 75mm² specimens cut from the half of the coathanger plaque nearest to the gate. All tests were performed at 5m/s and 23°C using a hemispherical impactor of diameter 10mm and a ring support of diameter 40mm. Ten specimens were tested for each material.

The force-displacement signal for these tests is relatively complex and a detailed interpretation of the principal features of the signals has been reported elsewhere [7,8]. This enabled us to identify the energies associated with crack initiation and with total energy prior to contributions from friction as the impactor probe pushes through the specimen. (This later energy is called "peak" energy because it is associated with the maximum force position on the force-displacement signal.)

The impact results for the dispersed and undispersed materials are shown in Table 5 with the values given in the brackets relating to the standard deviations.

The IFWI test is dominated principally by bending stresses which are controlled by the "skin" layers in the composite. Since the mean fibre lengths and orientations in the "skin" region are approximately the same for both the dispersed and undispersed compounds, it is expected that the toughness values would

Table 5: Instrumented Falling Weight Impact results for both compounds, tested at 5 m/sec speed and at 23°C. The mean results together with the standard deviations quoted in brackets are from ten specimens.

Compound Type	Initiation Energy (J)	Peak Energy (J)
"dispersed"	0.59 (0.07)	7.02 (0.97)
"undispersed"	0.51 (0.05)	5.99 (1.09)

be dominated by the amount of fibre present in the materials. Based on the information contained in Figs. 10 and 11, the dispersed material has a lower volume fraction in both the "skin" and "core" regions than the undispersed material. Consequently the impact toughness based on both the initiation (associated with the initiation toughness) and peak (associated with the total energy involved in failure) energy values for the undispersed compound should be larger than the dispersed compound. After analysing the data in Table 5 using a student t-test, the initiation energies were found to be 95% likely to be different, indicating that the dispersed material is slightly tougher than the undispersed material. Therefore, it is clear that the level of dispersion of the fibre is critical to the toughness of the material and that good dispersion is favourable. Although these observations only relate to the "skin" portion of the two samples, the conclusion is nevertheless important.

In an attempt to gain a geometry independent and a broader picture of the toughness of these materials, fracture mechanics tests were performed on compact tension specimens according to the protocol developed by Williams [9]. Compact tension specimens were selected for these measurements in order to provide values for both initiation toughness [9] but also propagation toughness. Only K_{Ic} values can be determined for this geometry since we had no accurate means of determining displacement in the line of applied force. The force was applied at 90° to the direction of mould fill.

Three specimens were tested for each material at 23°C and 1mm/min. The crack length was measured during the tests using a crack foil gauge bonded to the size of the specimen. This enabled initiation and

Table 6: Fracture mechanics test results performed using compact tension specimen geometry for both compounds, tested at 1 mm/min speed and at 23°C. The mean results together with the standard deviations quoted in brackets are from three specimens.

Compound Type	Initiation K_{Ic} (Mn/m ^{3/2})	Propagation K_{Ic} (MN/m ^{3/2})
"dispersed"	5.77 (1.30)	10.5 (0.87)
"undispersed"	5.96 (0.53)	12.7 (1.21)

propagation values to be calculated for the fracture mechanics parameters K_{Ic} . In each case, the initiation point was deduced from the crack gauge signal. The results are given in Table 6.

Interpretation of the toughness from the CT specimens is unclear. The results in Table 6 show a similar toughness at initiation for the two materials but apparently slightly higher toughness during propagation for the sample with the undispersed fibres. Unfortunately, the complications that accompany these results prevent a clear conclusion from the results, as we shall now show.

The Compact Tension test drives the crack in the direction of mould fill and hence along the interface between fibres and the matrix in the "skin" region, but across fibres in the "core" region. The fracture mechanisms occurring in the "core" regions will include fibre fracture which is a highly energy absorbing process. Consequently the "core" region contributes quite strongly to the overall toughness. As discussed previously, the higher fibre concentration for the undispersed sample will mean more fibres in the "core" and therefore a high energy per unit volume contribution from the fibre fracture process. This will result in an expectation of higher toughness for the undispersed sample. This is not observed at initiation and is marginally observed during propagation. However, it leaves the results unresolvable and no clear indication emerges on whether dispersed fibres aid toughness or not.

4.2. Tensile Dilatometry

4.2.1. Theory for Volume Change During Deformation

The measurement of volume strain during a tensile test provides an insight into the various deformation mechanisms that govern material behaviour, as well as monitoring the usual stress-strain, modulus-strain functions and measuring the lateral contraction ratios for the material.

The volume strain can be expressed by

$$V_s = (1 + \epsilon_A)(1 + \epsilon_W)(1 + \epsilon_T) - 1 \quad (1)$$

where the subscripts A , W and T refer to the axial, width and thickness directions in the tensile deformation.

The samples under consideration display a large degree of anisotropy, so that the fundamental elastic constants such as tensile modulus E and lateral contraction ratios ν are direction dependent. The lateral contraction ratios in the width and thickness directions are defined by

$$\nu_W = - \frac{\epsilon_W}{\epsilon_A} \quad (2)$$

$$\nu_T = - \frac{\epsilon_T}{\epsilon_A} \quad (3)$$

Consequently, Eq. 1 can be rewritten by substituting Eqs. 2 and 3

$$V_s = (1 + \epsilon_A)(1 - \nu_W \epsilon_A)(1 - \nu_T \epsilon_A) - 1 \quad (4)$$

On expansion of this equation, the volume strain V_s can be expressed as

$$V_s = \epsilon_A(1 - (\nu_W + \nu_T)) + \epsilon_A^2(\nu_W \nu_T - (\nu_W + \nu_T)) + \epsilon_A^3 \nu_W \nu_T \quad (5)$$

Thus it can be seen that the volume strain, V_s , contains first, second and third order terms that are based on the axial strain and the width and thickness lateral contraction ratios ν_W and ν_T . It is possible to predict the volume strain response for a material at any

applied axial strain if both the width and thickness lateral contraction ratios are known, using Eq. 5.

It is possible to monitor the mechanisms involved in bulk deformation by comparing the change in the volume strain with an increasing stress. The major contribution involves the normal elastic response due to hydrostatic tension in the bulk of the material under stress, defined by Eq. 5. In addition other mechanisms will contribute to the volume strain increase as the stress is increased. The basic shape of the volume strain/axial strain plot enables the distinction between the normal state of hydrostatic deformation, and further mechanisms of cavitation or yield to be distinguished as shown in Fig. 14. The volume increasing mechanisms include crazing, voiding, microdebonding; shear yield mechanisms include shear band formation (see /11-14/). The region of hydrostatic deformation can be calculated for the compounds under examination by inserting the small strain ($<0.2\%$) values for the width and thickness lateral contraction ratios into Eq. 5. The onset of

cavitation can be associated with the departure from the predicted volume strain behaviour in the region of hydrostatic tension, under increasing strain. Yield type behaviour can be seen as an eventual limiting value of volume strain with an increasing axial strain.

4.2.2. Experimental Measurements Using Tensile Dilatometry

Tensile deformation of parallel sided specimens machined from the centre of the coathanger plaque mouldings allowed the measurement of tensile modulus and volume strain for each type of material. The specimen geometry tested was labelled 90° (with the stress aligned perpendicular to the mould fill direction), so that a majority of fibres in the "skin" regions were oriented between 60° to 90° to the applied stress. Each specimen had a width and thickness of approximately 12.5 mm by 3 mm and a gauge length of 100 mm. The deformation was

Fig. 14: Schematic representation of the volume strain vs axial strain relationship shown diagrammatically. The shape of the two types of curve, showing volume increasing (cavitation) or deviatoric (shear banding) are shown, together with the region of dilational behaviour.

Table 7: Tensile modulus, width and thickness lateral contraction ratios (all measured at 0.2% applied strain) for both compounds, tested using 90° oriented specimens, at 1 mm/min speed and at 23°C. The mean results together with the standard deviations quoted in brackets are from five specimens. The volume strain measurements at 1.0% applied strain are also quoted.

Compound Type	Modulus 90° Specimen (GPa)	ν_W	ν_T	Volume strain at 1.0% axial strain
"dispersed"	8.23 (0.75)	0.23 (0.01)	0.50 (0.09)	0.28 (0.11)
"undispersed"	9.50 (0.90)	0.20 (0.03)	0.44 (0.01)	0.45 (0.04)

Table 8: Predicted versus measured volume strain response at 1.0% applied strain for both the "dispersed" and "undispersed" compounds, tested at the 90° orientation. The measured data is taken from Table 7.

Compound Type	V_s Predicted ($\epsilon_A = 1.0\%$)	V_s Measured ($\epsilon_A = 1.0\%$)
"dispersed"	0.26	0.28 (0.11)
"undispersed"	0.34	0.45 (0.04)

performed using a screw driven 6025 Instron at a cross head rate of 1 mm/min, with strain measurements being monitored in three mutually perpendicular directions, together with the applied stress. The equipment and method for calculating the volume strain are described more fully by Johnson et al. [8]. The measured values for modulus (taken at 0.2% axial strain), lateral contraction ratios in the width and thickness directions (determined from the slope of the lateral/axial strain up to 0.5% axial strain) and the volume strain (determined at 1.0% axial strain and approximately 75 MPa stress respectively) are shown in Table 7.

The difference in modulus for the two compounds is principally due to the volume fraction differences in the feedstock, as determined from data in Fig. 11. The dispersed compound had a mean fibre volume fraction of 32%; the undispersed compound had a mean fibre volume fraction of 43%. The mean modulus values for the two compounds reflect these ratios, with the higher volume fraction leading to a higher modulus.

There is a clear difference in the value for the width and thickness lateral contraction ratios for each of the compounds. This is to be expected from the microstructure of both compounds. The values for the lateral contraction ratios for the dispersed and undispersed compounds in the width direction are the same within experimental error. The thickness lateral contraction ratios for the dispersed and undispersed compounds are different; this is due to slight variations in the microstructure in the materials.

These values for the lateral contraction ratios can be used to estimate the volume strain-axial strain function, using Eq. 5. The predicted values for the volume strain at 1.0% applied axial strain are shown in Table 8.

The predicted result for the dispersed compound is in good agreement with the measured value for the volume strain, indicating that the deformation at 1% axial strain is primarily dilational in nature. For the undispersed compound, the predicted result is somewhat smaller than the measured value for the volume strain. This indicates that an extra volume increasing mechanism is occurring over the strain range considered. A likely explanation lies in some form of cavitation process, which might include debonding at the fibre matrix interface and/or an increase in voiding due to poorly wetted fibres. Figs. 15 and 16 show the volume strain-axial strain plots for the dispersed and undispersed compounds tested in the 90° direction. The volume strain plot for the dispersed compound follows the predicted line as described by Eq. 5. The volume strain-axial strain plot for the undispersed compound shows departure from the predicted line at about 0.5 - 0.8% axial strain. This

Fig. 15: Volume strain vs axial strain plot for the "dispersed" compound, measured with the stress applied 90° to the direction of melt impregnation.

Fig. 16: Volume strain vs axial strain plot for the "undispersed" compound, measured with the stress applied 90° to the direction of melt impregnation.

indicates that a cavitation type process begins at this value of deformation.

4.3. Creep and Recovery

Application of stress to a plastic will result in a consequential strain; removal of the stress will enable deformational recovery to occur. This recovery process may continue until all residual deformation has been removed or will never be complete because irreversible damage has occurred during the loading period. Therefore a recovery strain subsequent to loading will be a function of time, original level of deformation and failure mechanics; the latter will be influenced by the micromechanics of the material. This recovery function together with the characteristics of the time dependent strain during loading (i.e., the creep function) can be used in order to investigate the influence of fibre dispersion and the degree of impregnation.

Creep and recovery tests were performed on a series of both 0° (with the stress aligned parallel to the mould fill direction) and 90° (with the stress aligned perpendicular to the mould fill direction) creep coupon specimens, for both the dispersed and undispersed compounds according to the methods described by Turner /15/. The specimens were machined from the centre of the coat hanger plaque panels and had nominal widths of 10.0 mm and thicknesses of 3.0 mm. The measurements were performed on lever loading creep machines with the strain measured using optical extensometry. Before the full creep experiment a short series of loading (up to 100 seconds) and unloading (up to 400 seconds) tests were performed on separate specimens, to establish the 100 second isochronous modulus strain curve. These "mini" isochronous stress-strain measurements were conducted to ensure that no strain damage was introduced by the applied load on the creep specimens once the full creep and recovery test began. This was confirmed by observation of the final recovery strain.

The creep programme was conducted for 10⁵ seconds for both the "dispersed" and "undispersed" compounds, with an approximate strain equal to 0.3% and 0.35% after 100 seconds for the 0° and 90° specimens, respectively. After 10⁵ seconds had elapsed

the recovery stage commenced, with the approximate 10⁵ creep strains equal to 0.4% and 0.5% for the 0° and 90° specimens, respectively. The recovery strain was then monitored as a function of time. Following the methods developed by Turner /15/, results are presented as fractional recovered strain (ϵ_{FRS}) versus reduced time, where

$$\epsilon_{FRS} = 1 - \frac{\epsilon(t_r)}{\epsilon_{max}} \quad (6)$$

with $\epsilon(t_r)$ the strain after (t_r) seconds of recovery and ϵ_{max} the maximum strain at the end of the creep experiment.

The fractional reduced strain ranges from 0.0 (no recovery from the creep strain of ϵ_{max} at a creep time of t_0) to 1.0 (full recovery from the creep strain of ϵ_{max} at a creep time of t_0).

The reduced time under load is given by

$$\text{Reduced Time} = \frac{t_r}{t_0} \quad (7)$$

with t_0 the associated creep test duration.

The recovery and creep duration time were up to 10⁵ seconds, so that the reduced time ranged from 10⁻⁴ to 1 (10 seconds of recovery to 10⁵ seconds of recovery, respectively).

The 100 second isochronous modulus data for the dispersed and undispersed compounds, loaded in both the 0° and 90° directions relevant to the mould fill direction, are shown in Fig. 17. The modulus values at 0.2% applied strain detailed in Table 9 follow the expected pattern, with the 0° specimens showing larger values than the 90° specimens for both types of material. The undispersed compound also had a higher modulus than the dispersed compound, reflecting the difference in fibre volume fraction between the two compounds.

The creep response for these specimens is shown in Fig. 18. The 0° orientation specimens had approximately a common level of strain at 100 seconds, as had the 90° orientation specimens. After 10⁵ seconds, the creep strain for each specimen was similar for both types of compound, indicating that a similar degree of viscoelastic existed. This enabled recovery to start from a common level of deformation.

Table 9: 100 seconds isochronous modulus values taken at 0.2% applied strain and at 23°C for both the "dispersed" and "undispersed" compounds tested using 0° and 90° specimens.

Compound Type	100 second isochronous modulus at 0.2% strain, 23°C	
	0° (GPa)	90° (GPa)
"dispersed"	10.9	8.5
"undispersed"	13.5	8.6

Table 10: Fractional recovered strain for both the "dispersed" and "undispersed" compounds after 10⁵ seconds recovery. A value of 1 for the fractional recovered strain indicates complete recovery, a value of 0 indicates no recovery.

Compound Type	Fractional recovered strain after 10 ⁵ seconds recovery	
	0° orientation	90° orientation
"dispersed"	1.000	0.935
"undispersed"	0.896	0.865

The fractional recovered strain (calculated using Eq. 9) is plotted against the reduced time (as calculated from Eq. 10) in Fig. 19 for all the specimens. The pattern for recovery is markedly different for the dispersed, compared to the undispersed samples at both orientations. The fractional reduced strain after 10⁷ seconds recovery is detailed in Table 10.

The dispersed compound tested in the 0° orientation displayed full recovery at the reduced time of 1, unlike the undispersed compound at the same orientation. The dispersed compound showed a better degree of recovery tested in the 90° orientation compared to the undispersed compound.

It is clear from the data shown in Fig. 19 that an extra mechanism is involved in the creep of the

Fig. 17: 100 second isochronous modulus strain measured with the stress applied at 0° and 90° to the direction of melt impregnation, for both the "dispersed" and "undispersed" compounds.

Fig. 18: Creep strain measured with the stress applied at 0° and 90° to the direction of melt impregnation, for both the "dispersed" and "undispersed" compounds.

Fig. 19: Fractional recovered strain versus reduced time under load for both the "dispersed" and "undispersed" compounds, applied at 0° and 90° to the direction of melt impregnation.

undispersed compound, compared to the dispersed compound. This is so because the dispersed compound recovers from deformation more than the undispersed compounds. The fractional recovered strains reflect the recovery from time dependent creep and any other mechanism of deformation that may occur during creep. If a deformational mechanism is occurring during creep that causes permanent damage (e.g., a voiding or debonding mechanism) then recovery of strain subsequent to such a mechanism will not occur. Therefore, the dispersed compound is less susceptible to an irreversible deformation mechanism than that for the undispersed compound. It is noticeable that the 90° specimen exhibits more unrecovered strain. For these specimens the stress field can readily encourage debonding around a fibre interface because of the alignment of the stress relevant to the larger volume of unaligned fibres in the 90° specimen. It is therefore apparent that better impregnation and/or fibre bonding is occurring for well dispersed compounds.

5. CALCULATION OF STIFFNESS

We have presented a description of the microstructure of the two materials as well as accounting for some stiffness and toughness properties. In so doing, it has been possible to associate certain aspects of fibre dispersion and fibre impregnation with properties. A more comprehensive way of linking the microstructural observations with mechanical properties would be to predict the properties. If this can be performed successfully, then it confirms the accuracy and relevance of the observed microstructure. Property prediction is possible for modulus and these calculations are now described.

The moduli values for the dispersed and undispersed materials have been modelled using the analysis described by Bailey et al. /6/, using the microstructural information detailed in Sec. 3. The model calculates the stiffness using a non interacting layer approach, where each layer contains fibres with a given orientation, length distribution and volume fraction. A sixteen layer model based on the microstructural details contained in Sec. 3 was used to predict the tensile modulus for both the dispersed and undispersed materials. However, the resolution in

describing fibre volume fraction through the thickness prompted the 16 layers to be reduced to 8 (i.e., 4 layers for the half thickness). The model for the two materials paid particular attention to the volume fractions for each material and to the fibre length distributions. The details for the materials properties and microstructural parameters are displayed in Tables 11 and 12.

The data described in Tables 11 and 12 produced the predictions for the 0° and 90° tensile modulus as shown in Table 13.

The predicted results are in good agreement with the experimental data, successfully showing the increased stiffness with the principal direction of the mould flow. These results also predict that the stiffness for the undispersed material is greater than the dispersed material, where this difference is due principally to the increased volume fraction of the undispersed material and to a lesser degree to the fibre length distribution (the fibre orientation distribution being held constant for both models). The predictions of the model are quite sensitive to changes in the microstructural input, thus serving as a good indication that our observations of the microstructure of the two materials are an accurate reflection.

6. CONCLUDING COMMENTS

This study has shown clear differences in impregnation levels between the two types of long fibre reinforced thermoplastic compounds. Mechanical tests that are dominated by a fibre response (such as fracture toughness) have been

Table 11: Material properties used in the stiffness calculations for both the "dispersed" and "undispersed" compounds

Material Properties	"dispersed"	"undispersed"
Fibre tensile modulus (GPa)	76	76
Fibre diameter (μm)	17	17
Matrix tensile modulus (GPa)	2.8	2.8
Matrix tensile modulus (GPa)	1.0	1.0
Composite LCR	0.4	0.4

Table 12: Microstructural properties used in the stiffness calculations for both the "dispersed" and "undispersed" compounds

Data for "dispersed" compound			
Layer Type	Orientation Angle (°)	Layer Volume Fraction (%)	Fibre Length Distribution (Figure No.)
"Outer Skin"	10	28	Figure 12
"Skin 2"	20	32	Figure 12
"Skin 3"	20	36	Figure 12
"Core"	80	42	Figure 12

Data for "undispersed" compound			
Layer Type	Orientation Angle (°)	Layer Volume Fraction (%)	Fibre Length Distribution (Figure No.)
"Outer Skin"	10	46	Figure 13
"Skin 2"	20	40	Figure 13
"Skin 3"	20	40	Figure 13
"Core"	80	53	Figure 13

Table 13: Experimental data (taken from Table 9) compared to the calculated values for tensile modulus measured in the 0° and 90° orientation

Compound Type	0° Tensile Modulus at 23°C (GPa)	
	Predicted	Measured
"dispersed"	11.0	10.9
"undispersed"	13.6	13.5

Compound Type	90° Tensile Modulus at 23°C (GPa)	
	Predicted	Measured
"dispersed"	8.7	8.5
"undispersed"	8.9	8.6

difficult to resolve in terms of the influence of fibre dispersion and level of impregnation on a property. However, tests designed to measure the interfacial response (such as volume strain measurements and creep and recovery) have shown that improved properties are seen with compounds that have better levels of impregnation. A proper model for the growth of the cavitation mechanisms detected during deformation in the volume strain and creep and recovery tests would be needed to confirm the qualitative nature of these measurements. A full description of the resultant microstructures has been obtained that has allowed for the prediction of the stiffness of the compounds.

It is clear from the study performed that the enhanced impregnation gained from using a well dispersed pultrudate is retained subsequent to an injection moulding process. The material produced by the encapsulation process showed a deterioration of

property performance at both the pultruded and injection moulding stages, compared to the well dispersed compound, despite the fact that the undispersed compound had a higher fibre volume fraction.

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