Fibre Network and Compression Moulding of Polymer Composites

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Abstract
Squeezing flow of glass-mat reinforced thermoplastics (GMT) material was carried out in this research study to investigate the structure and reorientation of the fibre reinforcement and to develop a better understanding of the material's rheological behaviour. Circular disks of GMT material were squeezed to different final thicknesses, always forming an elliptical shape, and three-point bending tests were carried out on each specimen in the two main directions. The mechanical properties of the specimens confirmed the fibre rotation predicted by Jeffery's equation. It is shown that the fibres rotate in a manner aligning themselves in the major direction of the flow, which makes the material less anisotropic as the flow proceeds.

KEYWORDS: Rheology, GMT, Squeeze flow, Anisotropy, Fibre reorientation.

INTRODUCTION

The squeezing flow of polymer composites (thermoplastic matrix reinforced by continuous glass fibre) was investigated in this work because of the importance of this material in manufacturing industrial parts for automotive and non-automotive applications [1]. The governing viscosities of the flowing material, the filling time, how far the filling is achieved when different and complicated shapes are considered, and the homogeneity and mechanical properties (strength) of the final products are thus urgently important.

GMT-PP 30% glass fibre material manufactured by PCD-Polymere, Austria was investigated in this research study and displayed anisotropic results under lubricated compression moulding as shown in Figure 1. In the previous work [2, 3], the squeeze flow process was modelled accordingly to accommodate this behaviour and a transversely isotropic model was introduced to give a mathematical representation to the squeezing process. The transversely isotropic model was used to characterise two different viscosities in the two in-plane directions because the flow pattern followed an elliptical rather than a circular shape. The model considered that the material is governed by two main viscosities in two orthogonal directions and was solved for the Newtonian case [2] and for the non-Newtonian case [3]. The major assumption in using the transversely isotropic model is that the viscosity through thickness was assumed to be equal to the viscosity in one of the in-plane directions. The two in-plane viscosities were determined depending on the geometrical shape which was elliptical. That enabled the derivation of a relationship between two strain rates and then two viscosities were calculated.

Fibre structure and orientation was also previously studied [4] and showed that the anisotropic flow behaviour is due to the slight orientation of the fibre in one direction more the other. In this work the authors are concerned about the reorientation of the fibre during the flow process, possible fibre-matrix separation and the impact of such behaviour on the flow process and on the mechanical properties of the final moulded parts.

MATERIAL

GMT-PP, 30% glass fibre manufactured by PCD-Polymere, Austria, was used in this research study. The matrix material is polypropylene, and it contains swirled continuous glass fibre (Figure 2), showing the fibre structure of the material after burning off the matrix in a convection oven at 500 °C for 4 to 5 hours. The material (commercially known as Daplen TC-GMT) is available as semi-finished sheets containing 30% glass fibre by weight [5]. The sheets are manufactured by impregnating glass mats with thermoplastic matrix on double belt presses by extruding a thermoplastic melt between two or more chemically or mechanically bonded glass mats.

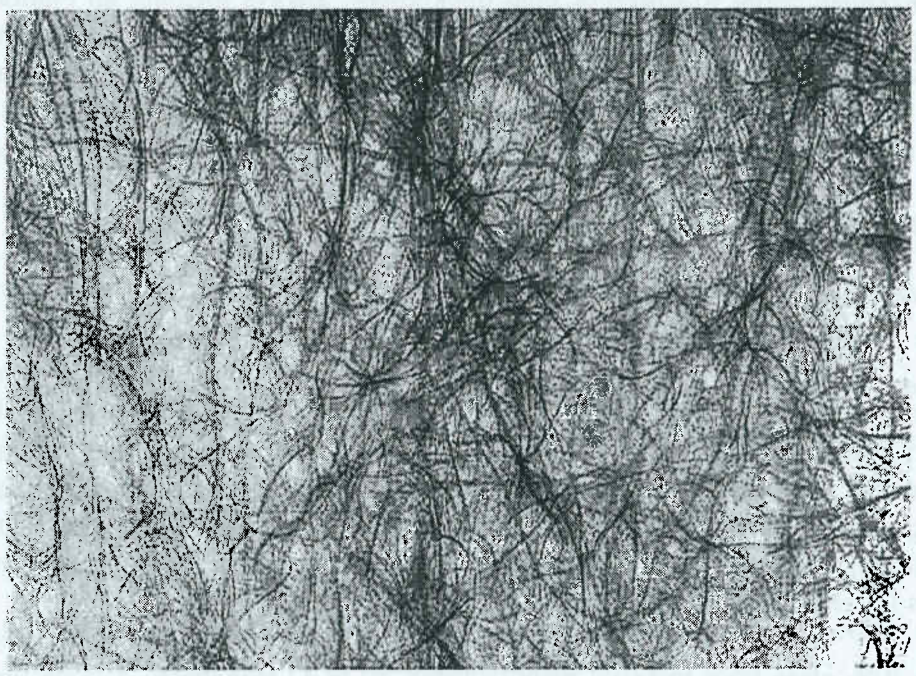

Figure 1: Photograph of the fibre after burning off the matrix

EXPERIMENTAL RESULTS

Squeezing flow behaviour

The fully lubricated, circular mould was set up on a universal 100 kN hydraulic Instron test machine and heated to 180°C. 50 mm diameter circular disks (3.7 mm thick) of 30% continuous glass fibre mat were isothermally squeezed to different final thicknesses as shown in Figure 2. The specimen started as a circle and as it was squeezed the elliptical shape started to appear gradually.

The elliptical shapes (Figure 2) which resulted from squeezing circular disks of GMT can be modelled as shown in Figure 3. To make the modelling process possible and to characterise the material as an anisotropic material the previous relationship [2, 3] between the major and minor axes of the elliptically deformed specimens can be also used in this work:

$$e = \frac{r_1}{r_2} \qquad (1)$$

$$f = \frac{\Delta r_1}{\Delta r_2} \qquad (2)$$

Figures 4 and 5 show how *e* and *f* vary with mould separation distance (*h*). *f* was taken as a constant (Figure 4) at any mould plate separation (*h*) while *e* is a variable with h (Figure 5).

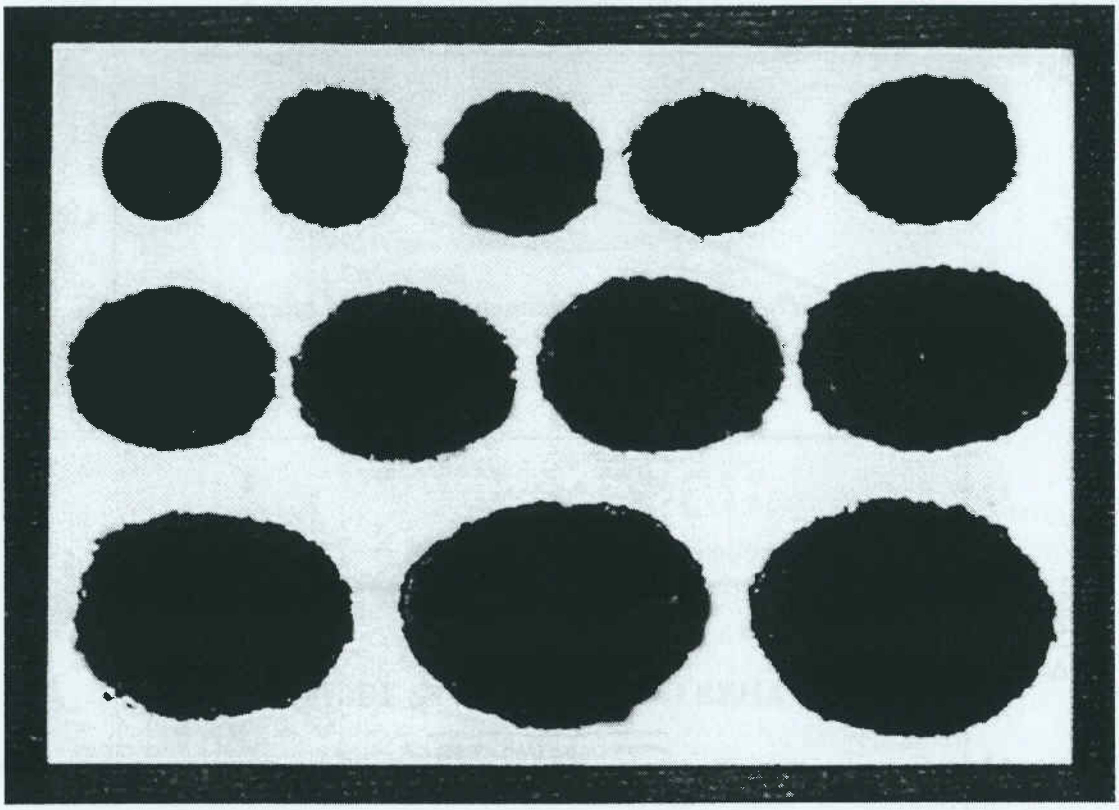

Figure 2: A photograph showing the stages of the elliptical deformation as the specimens were squeezed to different final thicknesses.

Taking into consideration that the material deforms at constant volume and the two parameters *e* and *f*, three different flow velocities can be derived in the three directions x, y and z where x and y are identical to the major and minor axes of the elliptically deformed specimen while z denotes the through thickness direction:

$$V_x = \frac{fux}{(e+f)h} \qquad (3)$$

$$V_y = \frac{euy}{(e+f)h} \qquad (4)$$

$$V_z = -\frac{uz}{h} \quad (5)$$

where u is the mould closing speed (squeezing rate) and h is the plate separation or the gap between the two mould plates.

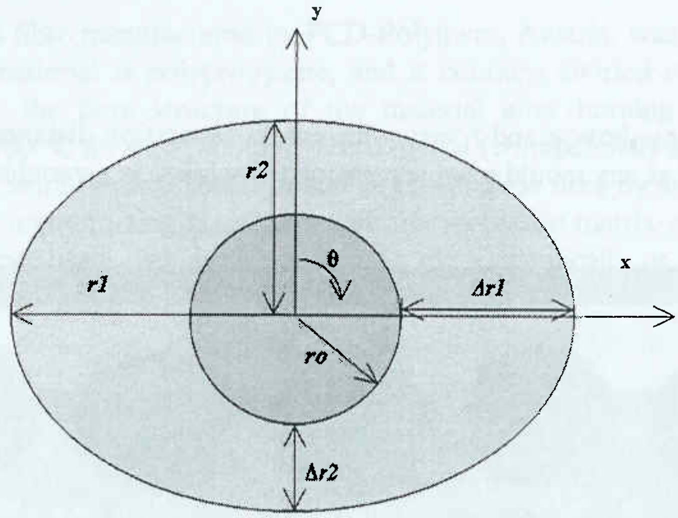

Figure 3: Schematic of the specimen before and after squeezing.

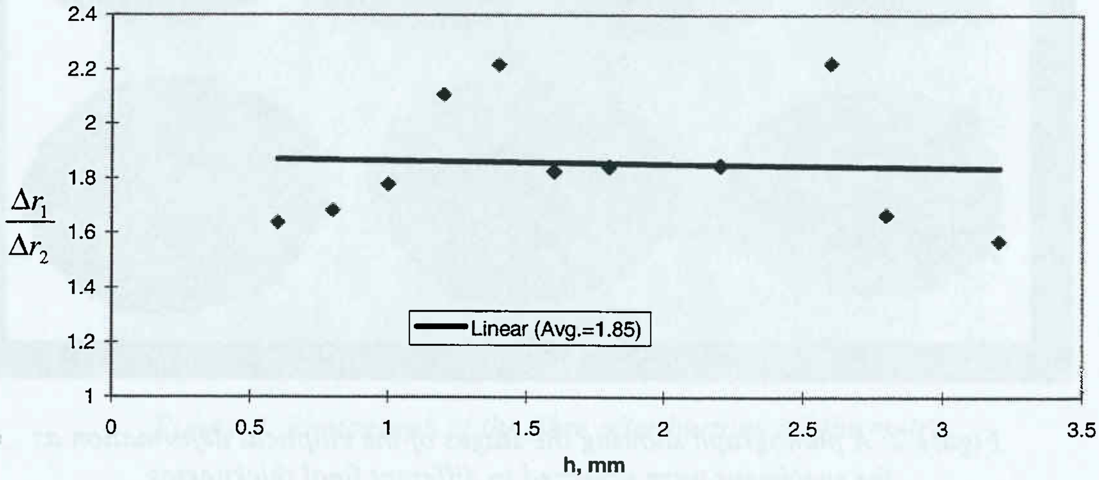

Figure 4: $\frac{\Delta r_1}{\Delta r_2}$ *(f) versus mould separation (h, mm)*

Three point bending test

A 100 kN Zwick screw driven testing machine was used to carry out the three-point bending as a non-destructive test. Each of the specimens shown in Figure 2 was tested twice, each specimen was loaded first in the major axis direction and the load and displacement were kept in the elastic region, then the specimen was rotated by 90° and loaded again within the elastic region. Load versus displacement was recorded for each experiment and the flexural modulus was calculated according to the following equation for the three-point bending:

$$E_x^b = \frac{PL^3}{4bh^3 w_c} \quad (6)$$

where P is load (N), w_c is the mid-point test deflection (m).
L is the span distance; it was taken as 33.23 mm for this case for all the tested specimens.
b and h are dimensions of the cross sectional area of the specimen where the load was imposed.
Table 1 shows the different dimensions of the twelve specimens tested for bending.

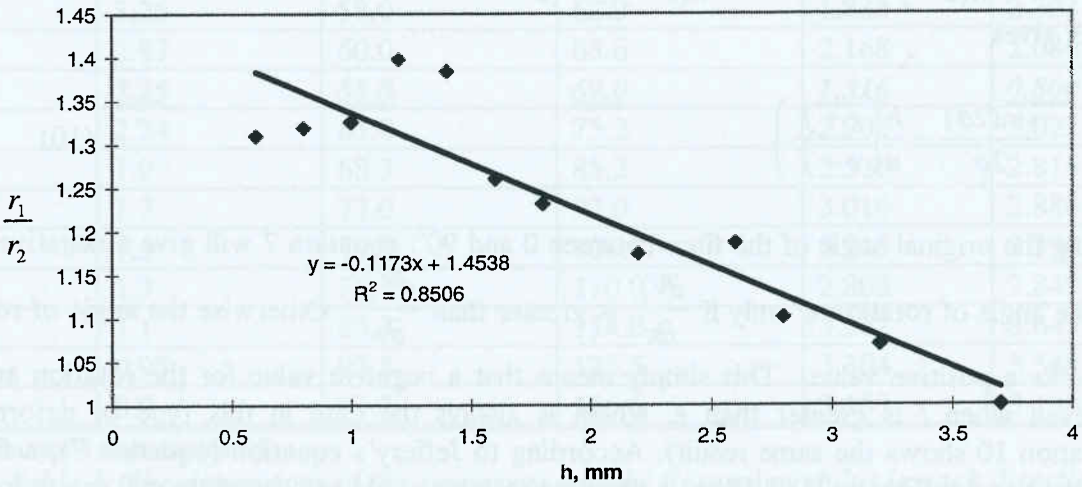

Figure 5: $\frac{r_1}{r_2}$ *(e) versus mould separation (h, mm)*

THE FIBRE STRUCTURE AND BEHAVIOUR

Jeffery's Theory

GMT flow was previously considered to be planar and Jeffery's equation [6] was presented by Meij et al. [7] in the X-Y plane for short diluted fibre as:

$$\phi = -\sin\phi.\cos\phi.\frac{\partial V_x}{\partial x} - \sin^2\phi.\frac{\partial V_x}{\partial y} + \cos^2\phi.\frac{\partial V_y}{\partial x} + \sin\phi.\cos\phi.\frac{\partial V_y}{\partial y} \quad (7)$$

ϕ is the fibre rotation angle. If $\phi = 0$ no rotation takes place and if $\phi \neq 0$ there is fibre rotation as a result of the flow of the material. V_x and V_y are the flow velocities in the X and Y directions as given by equations 3 and 4. To substitute in equation 7 and find a value for the rotation angle, equations 3 and 4 give:

$$\frac{\partial V_x}{\partial x} = \frac{fu}{(e+f)h}$$

$$\frac{\partial V_x}{\partial y} = 0 \tag{8}$$

$$\frac{\partial V_y}{\partial y} = \frac{eu}{(e+f)h}$$

$$\frac{\partial V_y}{\partial x} = 0$$

Equation 7 can be reduced to:

$$\phi = \frac{u(e-f)}{h(e+f)} \sin\phi \cos\phi = \frac{u(e-f)}{h(e+f)} \frac{\sin(2\phi)}{2} \tag{9}$$

which gives

$$\frac{\sin(2\phi)}{2\phi} = \frac{h}{u}\left(\frac{e+f}{e-f}\right) \tag{10}$$

Taking the original angle of the fibre between 0 and 90°, equation 7 will give a negative value for the angle of rotation, ϕ only if $\frac{\partial V_x}{\partial x}$ is greater than $\frac{\partial V_y}{\partial y}$. Otherwise the angle of rotation will take a positive value. This simply means that a negative value for the rotation angle is achieved when f is greater than e, which is always the case in this type of deformation (equation 10 shows the same result). According to Jeffery's equation (equation 7), a fibre in the first quarter (Figure 3) will have a negative rotation angle and therefore will rotate towards the X-axis while a fibre in the second quarter (Figure 3) will have a positive rotation angle (because the cosine takes a negative value in the second quarter). A positive rotation angle in the second quarter also means that the fibre will move towards aligning with the X-axis.

Accepting that the anisotropic behaviour was encountered due to the fibres being aligned in the Y-direction more than the X-direction [4], the findings in this paper shows that the anisotropic behaviour might disappear after squeezing the material to a certain thickness. This means that more fibre will align in the major flow direction, which was initially the direction of less fibres, and such rotation of the fibres should change the flow phenomenon. More experimental investigations are needed to support these theoretical findings.

Three point bending test

The specimens shown in Figure 2 were tested under bending in the two perpendicular directions in order to verify the finding that the fibre directions rotate experimentally and to compare what was established for short non-interacted fibre with the continuous long fibre, which is being tested here. The minor axis of the elliptical specimen was always found to be the zero direction of the original sheets which is the long dimension of the rectangular sheets received from the manufacturer. Some variation in the flexural modulus was reported which also suggests that a change in the fibre direction was taking place as a result of squeezing the GMT material. The full results of the three point bending test are shown in Table 1 below.

Figure 6 shows the flexural modulus $\left(E_x^b\right)$ versus displacement (displacement 0 means that an original specimen with the original thickness was tested) when the specimens were loaded along the major axis of the elliptical specimen. Figure 7 shows the same result for $\left(E_y^b\right)$ where the specimens were loaded along the minor axis of the elliptical specimen. It is shown by the two Figures 6 and 7 that the flexural modulus has increased as a result of increasing the mould displacement (or decreasing the mould separation, h, as the material was squeezed). Since both moduli were seen to increase, the result can only be justified as a result of a fibre-volume increases due to fibre-matrix separation, which were reported in the earlier work [2, 8].

specimen	thickness, mm	width 1 (minor axis), mm	width 2 (major axis),mm	E_x^b, GPa	E_y^b, GPa
1	3.75	50.1	50.1	2.105	3.056
2	3.25	58.0	62.0	1.833	2.703
3	2.87	60.0	65.6	2.168	2.084
*4**	***3.25***	***58.0***	***69.0***	***1.316***	***0.800***
5	2.24	63.0	75.2	2.903	3.022
6	1.9	68.3	85.2	2.576	2.815
7	1.7	73.0	92.0	3.016	2.880
8	1.5	72.2	101.0	2.945	3.390
9	1.3	76.3	110.0	2.808	2.849
10	1.1	85.3	114.0	3.282	3.643
11	0.95	93.5	125.5	3.304	3.548
12	0.9	97.3	126.6	3.187	3.136

* Specimen 4 was eliminated

Table 1: The dimensions of the specimens and the flexural modulus in each direction.

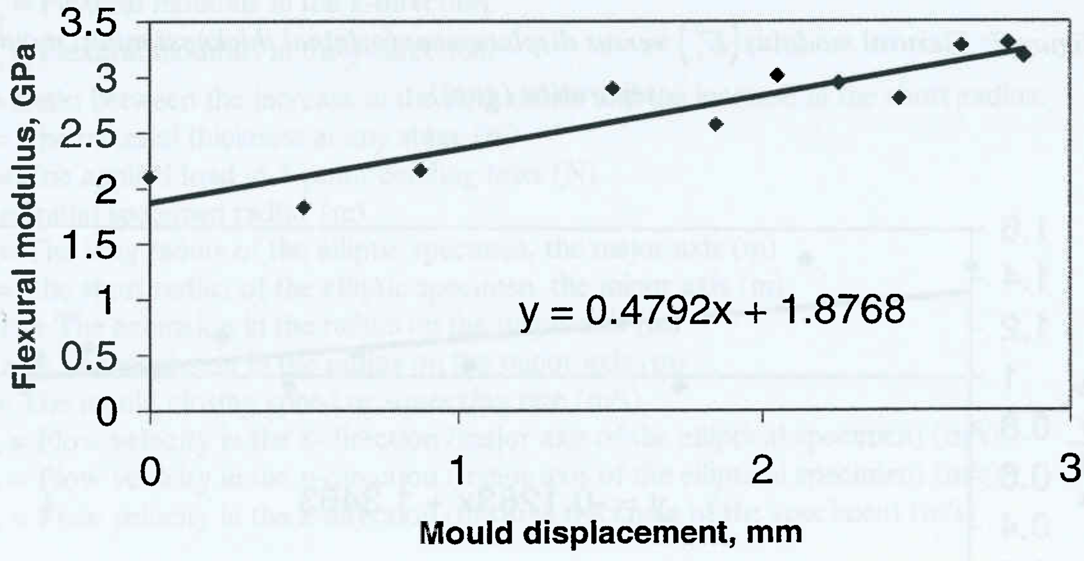

Figure 6: Flexural modulus $\left(E_x^b\right)$ versus mould displacement (original thickness minus mould separation)

The ratio between the flexural moduli in the two perpendicular directions was plotted in Figure 8, in order to be able to establish whether there was also fibre reorientation. Figure 8 shows that the ratio of the flexural modulus in the minor axis to its corresponding value in the major axis direction decreases as the mould separation (h) decreases. The decrease, from a ratio of over 1.35 in the original material, to a ratio of almost 1.0 in specimens squeezed by 3mm, suggests that more fibre rotated towards the main flow direction as the squeezing continued. The initial anisotropy in mechanical properties disappears as the squeezing proceeds, thus suggesting that the initial anisotropy of flow should also disappear.

The finding above agrees with Jeffery's theory for fibre rotation, presented earlier, which predicted that the fibres would tend to align more with the major flow direction. As the specimens start off with more fibre alignment in the minor flow direction, and thus a higher modulus in that direction, the overall effect is for the fibre to re-orient in a more isotropic fashion as the flow proceeds

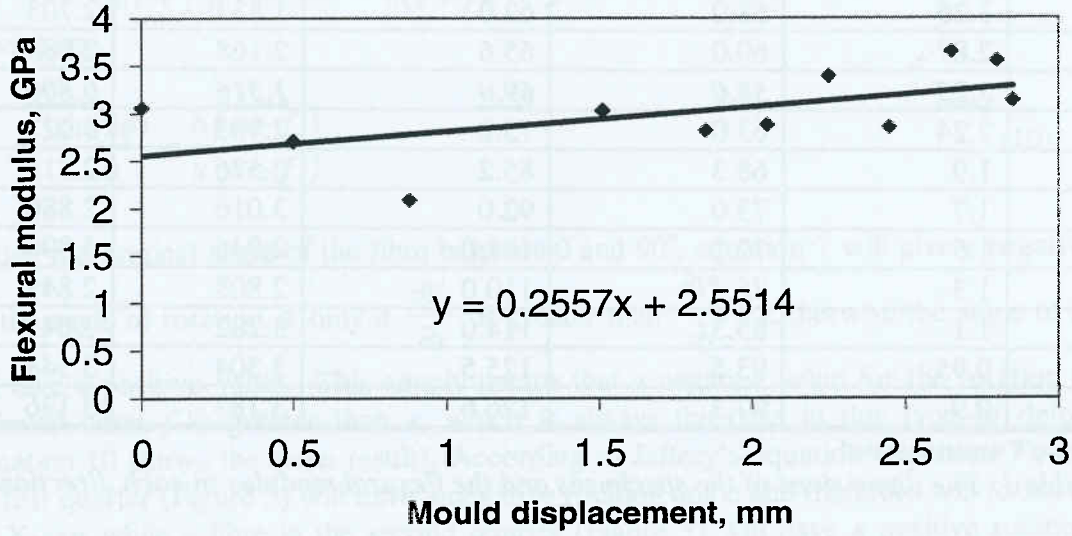

Figure 7: Flexural modulus $\left(E_y^b\right)$ versus displacement (original thickness minus mould separation (gap))

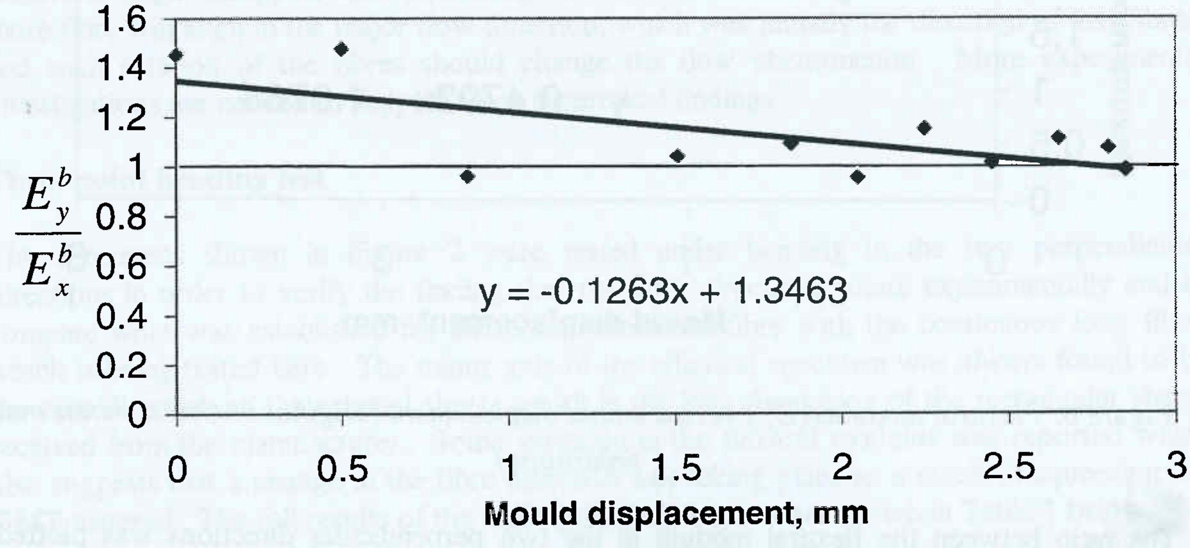

Figure 8: the ratio between the flexural modulus in the two main directions of the elliptical specimen versus displacement.

CONCLUSIONS

The main conclusion of this study is that the GMT material investigated exhibits anisotropic behaviour in both flow and mechanical properties, but that this anisotropy is diminished as the flow proceeds further.

Some other observations add to earlier work in characterisation of the anisotropic behaviour of the GMT as it flows:

The relationship between the minor and the major axes of the elliptically deformed specimens (the ratio $\Delta r_1/\Delta r_2$ may not be constant, as assumed in previous models but may vary as the squeezing flow proceeds, due to fibre re-orientation.

Both modulii (E_x^b) and (E_y^b) increase as the specimens are squeezed more, confirming the observations of higher fibre volume fraction and fibre-matrix separation observed earlier.

The ratio between the two flexural modulii $(E_y^b)/(E_x^b)$ decreases from an original value of over 1.35 to close to 1.0 as squeezing proceeds, thus confirming that the fibres re-orient and the material becomes more isotropic as the squeezing proceeds.

Nomenclature

e = Ratio between the long and the short radius of the elliptical specimen.
E_x^b = Flexural modulus in the x-direction.
E_y^b = Flexural modulus in the y-direction.
f = Ratio between the increase in the long radius and the increase in the short radius.
h = The material thickness at any stage (m)
P = The applied load in 3-point bending tests (N)
r_o = Initial specimen radius (m)
r_1 = The long radius of the elliptic specimen, the major axis (m)
r_2 = The short radius of the elliptic specimen, the minor axis (m)
Δr_1 = The extension in the radius on the major axis (m)
Δr_2 = The extension in the radius on the minor axis (m)
u = The mould closing speed or squeezing rate (m/s)
V_x = Flow velocity in the x-direction (major axis of the elliptical specimen) (m/s)
V_y = Flow velocity in the y-direction (minor axis of the elliptical specimen) (m/s)
V_z = Flow velocity in the z-direction (through thickness of the specimen) (m/s)

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