SHEAR DEFORMATION BEHAVIOUR OF FIBRE-REINFORCED COMPOSITES USING A MODEL COMPOSITE SYSTEM

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ABSTRACT: Experimental methodologies have been employed in the past to determine both longitudinal and transverse steady intraply shear viscosities of molten unidirectional continuous fibre-reinforced composites past resulting in, however, conflicting results. Many physical difficulties are inherently associated with testing for these material properties of thermoplastic matrix based composites due to the elevated testing temperature and fibre entanglement due to manufacturing vagaries. A model composite system has been adopted to overcome these difficulties consisting of long, rigid and virtually inextensible fibres embedded in a room-temperature liquid, Newtonian matrix. Experiments have been carried out to investigate the important rheological parameters associated with the deformation of continuous fibre-reinforced composite materials in steady-shear flow, both along and transverse to the principal fibre direction. To provide a constant shearing motion a custom-built apparatus has been designed and constructed in which a thin, central plate can be drawn out at constant velocity from a test specimen while the shear force exerted by the plate on the specimen is measured. Experimental data has been collected for different pull-out velocities, fibre concentrations, and ply thicknesses and it has been found that the behaviour of the composite varies significantly with changes in these parameters. The observed deformation and results have been carefully interpreted and compared with other published data based on a similar model composite. The viscosities deduced from the experimental data were also compared with values predicted by various mathematical models put forward by several authors. It was concluded that the models grossly underestimate the viscosity values for all values of fibre volume fractions used. Discrepancies between the numerical and experimental results have subsequently been postulated.

KEYWORDS: model composite, unidirectional, intraply shear, viscosity.

INTRODUCTION

The experimental measurement of the intraply shearing viscosities of an advanced composite material, carbon fibre reinforced polyetheretherketone (PEEK), has already been published [1]. The paper describes the experimental difficulties associated with induced a state of steady shear throughout samples of the composite at the forming temperature of the material and highlighted the conflicting results between that study and other published data.
Experimental difficulties would arise by using the advanced thermoplastic composites by virtue of their physical properties i.e. the relatively high processing, hence testing, temperature, and quite significantly, fibre misalignment. The existence of relatively small

fibre misalignments had been noted in commercial composites; through a microscopic study of preimpregnated tapes of APC-2, it has been reported [2] that 83% of the fibres were within ±1° of being perfectly aligned, and measured maximum off-axis deviations of up to ±4° both in-plane of the tape and out-of-plane. The amount of fibres misaligned to 4° was 2-3% approximately, however upon moulding, due to adjacent fibres to reorient co-operatively, this value could typically increase to 10% of fibres. Another imperfection of the internal structure of the composite, namely fibre entanglement, is a direct consequence of this fibre misalignment. Thus, without these complicating factors characteristic of commercial composites, direct comparisons could be made between experimentally determined results and those proposed by numerical models.

Much in-road into establishing a database for the longitudinal and transverse intraply shear viscosities has recently been made using so-called 'model' composite systems, which are utilised to represent or imitate the properties of commercial composites. These model composites consist of a matrix that is a liquid at room temperature meaning that from an experimental point of view there is a significant advantage in that there is no need for a heat cycle. Several researchers [3-7] have made use of model composites consisting of larger diameter fibres than that found in commercial composites. This has the consequence that the fibres are straighter and more rigid which removes the complication of fibre misalignments and entanglements associated with commercial composite systems. There is also the advantage that with larger diameter fibres the model composite can be constructed by hand meaning that the fibre concentration can be controlled. This allows the effect of the concentration of fibres on properties to be investigated.

In this paper a custom-built experimental apparatus is described that was designed to provide experimental values for the longitudinal and transverse viscosities of model composite systems undergoing steady-shear deformation. The apparatus has originally been detailed for use in the elevated temperature tests on APC-2 [1]. The operation of the instrument is based on the principle of drawing a thin, flat plate from a composite specimen using a constant pull-out velocity and measuring the pull-out force as a function of the plate's displacement from its initial position. By aligning the fibres parallel to the pull-out direction or perpendicular to this direction the anisotropic behaviour of the test specimen can be characterised, and through appropriate mathematical analysis, the readings of pull-out force can be converted into corresponding values of longitudinal and transverse shear viscosity.

MODEL COMPOSITE SYSTEM

The material used in the experiments was a model composite that could be assembled by hand, thus avoiding the complications of fibre entanglement and misalignment that occur in the commercial composites. The desired features of the constituents of the model composite were as follows:

i) the matrix to be liquid at room temperature;
ii) the matrix viscous enough to construct a composite without a great deal of sample loss, but 'thin' enough to wet the fibres;
iii) the fibres to be straight, uniform, rigid and inextensible;
iv) both constituents, fibres and matrix, to have similar densities;
v) good adhesion between the matrix and the fibres.

Following previous work done on model composites [4-7], the composite selected consisted of long, rigid fibres of Nylon embedded in a liquid matrix of Golden Syrup. These constituents have density values of approximately 1.134gcm^{-3} and 1.45gcm^{-3} respectively. The Nylon fibres were manufactured using a standard extrusion process (Speciality Filaments Ltd), and supplied to order in a black colour with dimensions of 0.2mm diameter and 50mm length. The matrix was a Newtonian liquid with a viscosity of 70 Pa.s at 20°C. Comprehensive constituent examinations have been documented elsewhere [8].

APPARATUS

An illustration of the front-end of the experimental apparatus, with a sample to be tested *in situ*, is given in Figure 1.

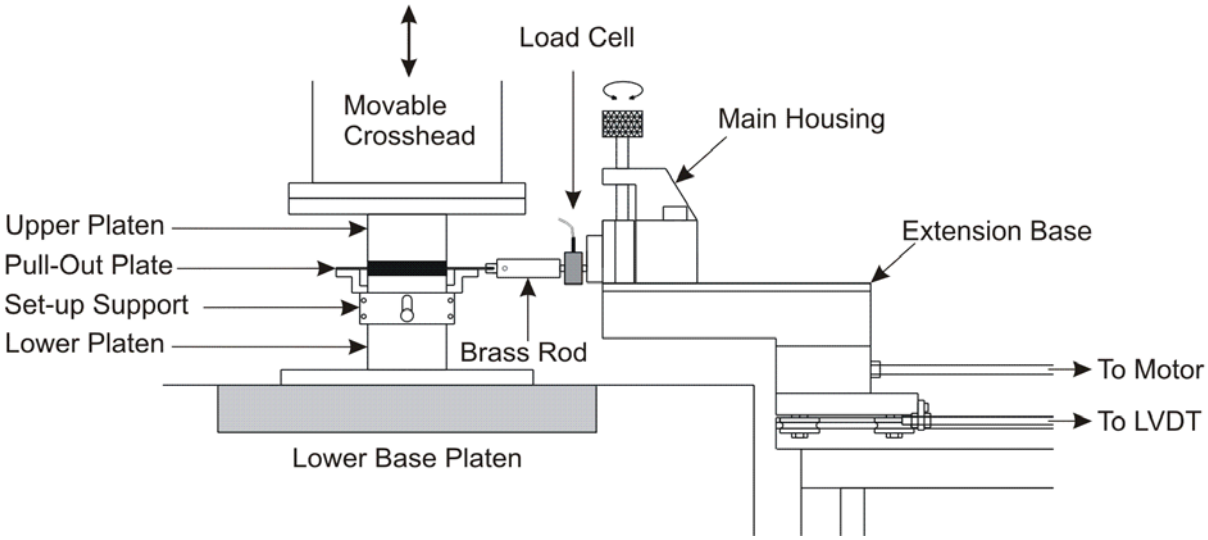


Figure 1: Illustration of the custom-built shear apparatus, indicating main parts of the rig.

The instrument consists of two principle units, one a commercially available Dartec rig to provide vertical motion for gap setting, and the other a side-mounted custom-built shear rig to provide the horizontal pull-out motion. The operating principles and technical details of the instrumentation and drive mechanism of the rig have been published [1, 2].

The temperature at which the test was conducted was of prime importance due to the high thermal sensitivity of the viscosity of the matrix. The surrounding ambient temperature was therefore controlled to within range of temperatures but was accurately recorded so that the resulting pull-out data could be standardised to 20°C. A Comark C9011 digital thermometer with a PP23L insertion probe, was used. The combination of the sensor and probe were calibrated by the manufacturer, according to NAMAS standards, to an accuracy of less than ±0.03% of the reading, or a maximum or ±0.1°C over the full temperature range.

SPECIMEN PREPARATION

Composite specimens were constructed in a delicate, indeed tedious, manner by hand. The fibre wetting procedure involved immersing bundles of the Nylon fibres, gripped by one hand, into a globule of the Golden Syrup. The matrix was worked into the fibre bundle by

careful manipulation of the compound by fingertip until a consistent composition was achieved with full fibre wetting. The procedure to construct the full composite sample and centralising the pull-out plate, attachment to the load cell etc. was a delicate procedure to ensure acceptable alignment, correct fibre volume fraction and so on.

EXPERIMENTAL PROCEDURE

With the composite fully laid-up and in position the test was started and the motor engaged. Figure 2 shows a typical experiment in progress for a composite specimen in which the fibres have been aligned with the pull-out direction. The gap between the pull-out plate and the fixed top and bottom plates, or equivalently the thickness of each composite layer, is 4 mm.

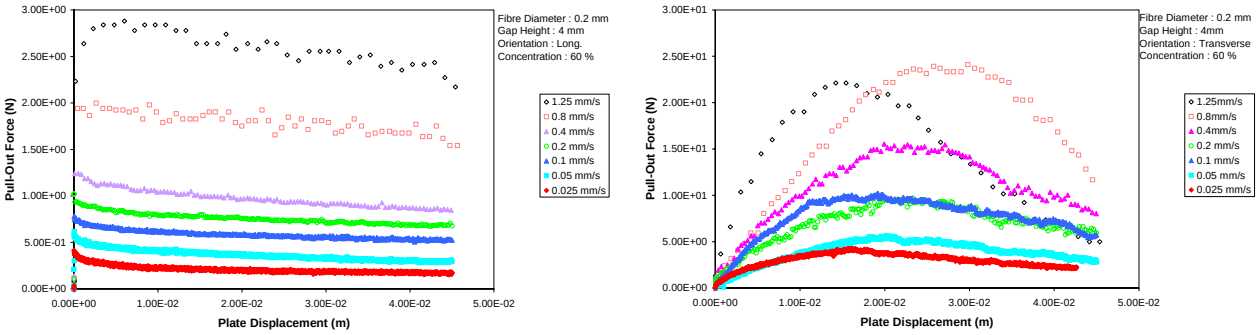
Figure 2: Photograph taken of a typical test in progress.

The photograph shows the classic sheared profile of the specimen through its thickness. The characteristic 'V' shape is evident within the composite showing the through thickness intraply shear.

RESULTS

It should be noted that because of the high temperature-dependence of the viscosity of the Golden Syrup matrix all results for the model composite tests have been standardised to a temperature of 20°C. This allows a direct comparison of graphs and results to be made. To investigate the effect of pull-out velocity a series of tests was carried out in which composite specimens were sheared using a range of different pull-out velocities, starting at the lowest velocity, $0.025 mms^{-1}$, and increasing in steps to the highest velocity, $1.25 mms^{-1}$. The fibre volume concentration in the tests was chosen to be 60% in order to coincide with the high volume concentration of fibres found in a typical commercial composite, while the thickness of each layer of composite was 4mm.

Figure 3 (a) contains a set of force versus displacement curves obtained for specimens in which the fibres were aligned with the pull-out direction. For the lower pull-out velocities, it is seen that the force immediately reaches a maximum point after which it gradually decreases over the remainder of the test. This suggests that at low shear rates an initial yield stress has to be exceeded at the start of a test in order for the material to deform. No such yielding behaviour was observed for the Golden Syrup alone, which implies that for the composite specimens some of the fibres might be coming into contact, thus leading to localised friction.



(a) (b)

Figure 3: Force versus displacement over 45 mm of shear for a range of pull-out velocities – (a) longitudinal direction, (b) transverse direction.

Force versus displacement curves for specimens in which the fibres were perpendicular to the pull-out direction are shown in Figure 3 (b). These graphs indicate a significantly different behaviour, with the pull-out force increasing steadily to a maximum value, before levelling off and starting to decrease.

During these tests it was observed that as the pull-out plate was withdrawn from the material, the fibres initially rolled over one another in what seemed to be a combination of rotational and translational flow, until after about 5-10mm of shear they bunched up as a result of the high fibre concentration. This led to a log-jamming effect (c.f. Figure 4) whereby each layer of composite appeared to become locked. A rapid build up in pull-out force was observed until a critical force was reached which was sufficient to draw the material at the front of the specimen from the enclosure of the fixed top and bottom plates, after which the force started to decrease. Examination of the internal structure of the sheared material showed a marked re-organisation and re-orientation of the fibres, with a distinct 'criss-cross' configuration.

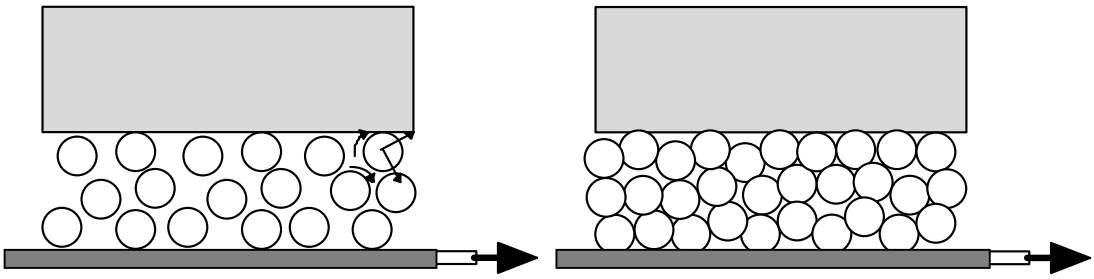


Figure 4: Simplified schematic of the effects on the fibres in a transverse shear test.

The experimental work was extended to determine the effect of fibre concentration on the composite's rheological properties. Tests were carried out on composite specimens of five different concentrations – 20%, 40%, 50%, 60% and 70%. The lowest value of concentration was set at 20% because, firstly, this would be the lower limit to any commercially available composite systems, and secondly, there was very much a practical reason for this value of 20% - below this concentration, it would have been extremely difficult to construct a composite with a somewhat even fibre distribution, although even 20% proved difficult to obtain successfully in practice. All specimens were sheared using a pull-out velocity of $0.1 mms^{-1}$, and the thickness of each layer of composite was 4mm. The longitudinal test results are shown in Figure 5.

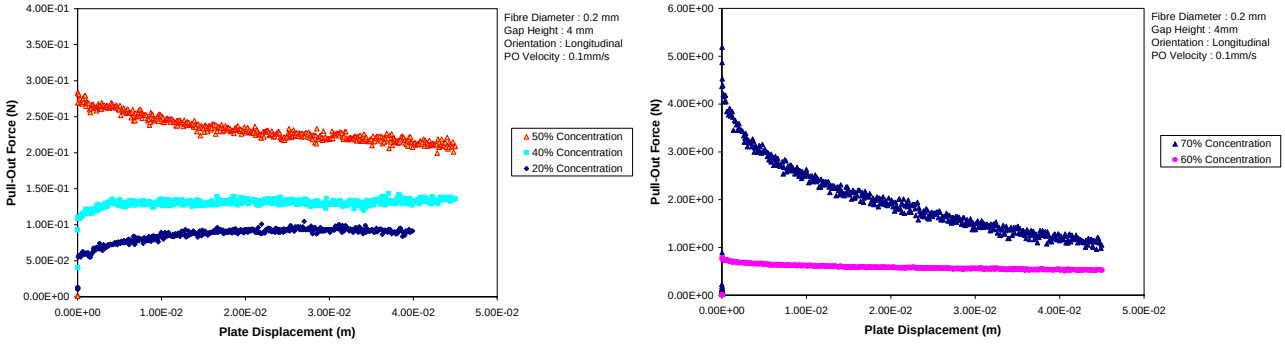


Figure 5: Force versus displacement curves obtained for specimens in which the fibres were aligned with the pull-out direction.

Force versus displacement curves for specimens in which the fibres were perpendicular to the pull-out direction are presented in Figure 6.

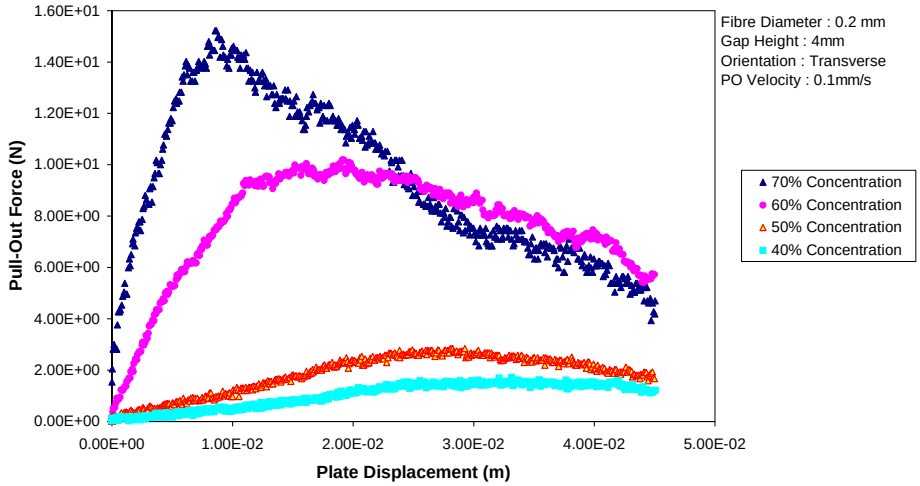


Figure 6: Force versus displacement curves obtained for specimens in which the fibres were aligned normal to the pull-out direction.

DISCUSSION

All of the results collected for 60% fibre volume concentration composites, tested in the longitudinal direction are presented in Figure 7. All shear rates are included in this plot.

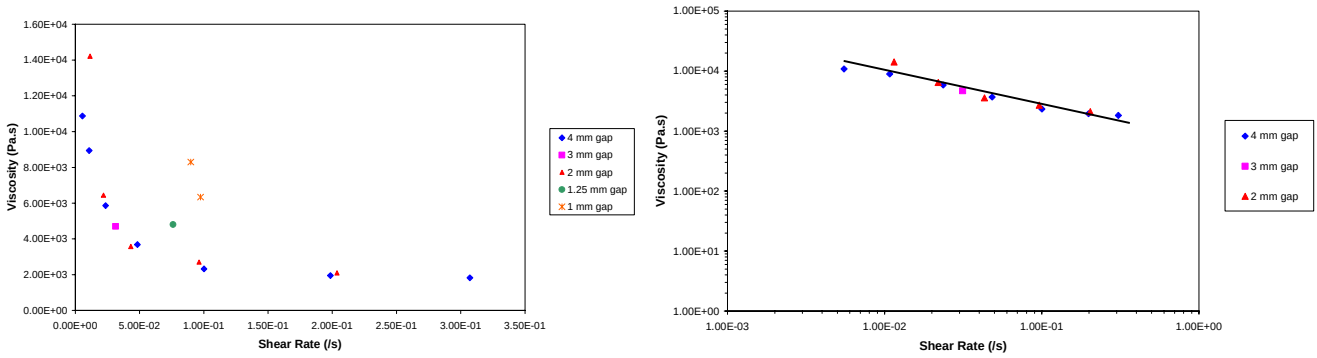


Figure 7: Calculated viscosity values versus shear rate, fibre volume fraction at 60 % - Longitudinal direction and the associated log-log plot.

From this plot it was observed that at the lowest shear rate, the longitudinal viscosity had relatively high values of 14,000 Pa.s for a 2 mm gap height, but, more importantly, ca. 11,000 Pa.s at a 4 mm gap height.[1] As the shear rate increased the viscosity values decrease rapidly before levelling off. This levelling off of viscosity may suggest that there was an identifiable region at the higher shear rates where the behaviour of the composite could be regarded as being approximately Newtonian. However, the shape of the curve is characteristic of pseudoplastic or shear-thinning polymer melts [9]. If one plots the viscosity values versus shear rate on a log-log scale, ignoring the outlying values recorded for the 1.25 and 1.0 mm gap heights, one can clearly see a 'power-law' region, typical of a polymer melt, over that shear rate range. In a similar manner, one may analyse the results derived from the experiments on the model composites, with the same preceding parameters, but with the fibres aligned in the transverse direction. Figure 8 is a plot of the viscosity values versus shear rates.

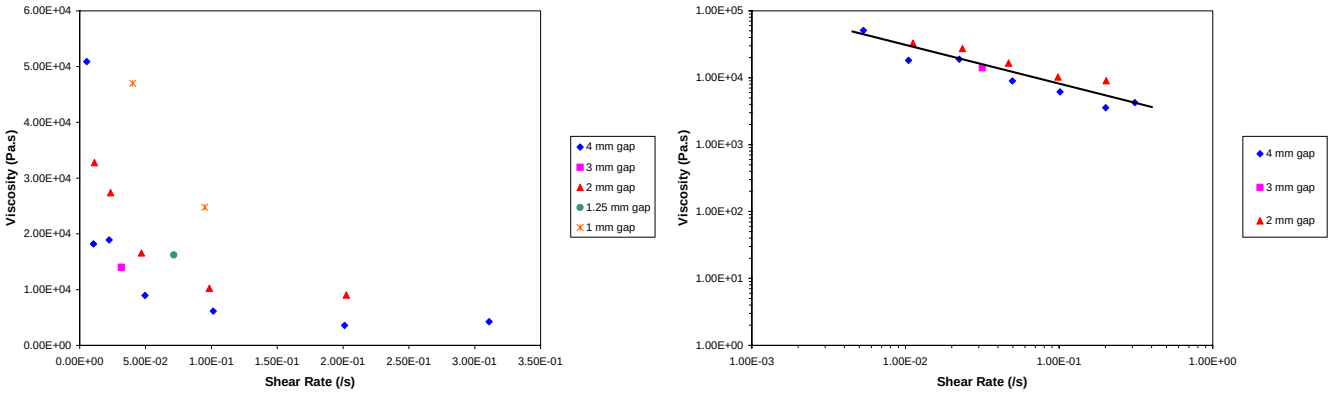


Figure 8:Calculated viscosity values versus shear rate, 60 % concentration - Transverse direction and the associated log-log plot.

Again, pseudoplastic behaviour is evident from this viscosity profile. In this principal direction, the viscosity drops off from in excess of 50,000 Pa.s at the lowest shear rate to approximately 4,000 Pa.s at the highest shear rate. Figure 9 shows the dependence of composite viscosity in both principle fibre directions against fibre concentration. For the lower values of concentration there appeared to be a relatively small dependence.

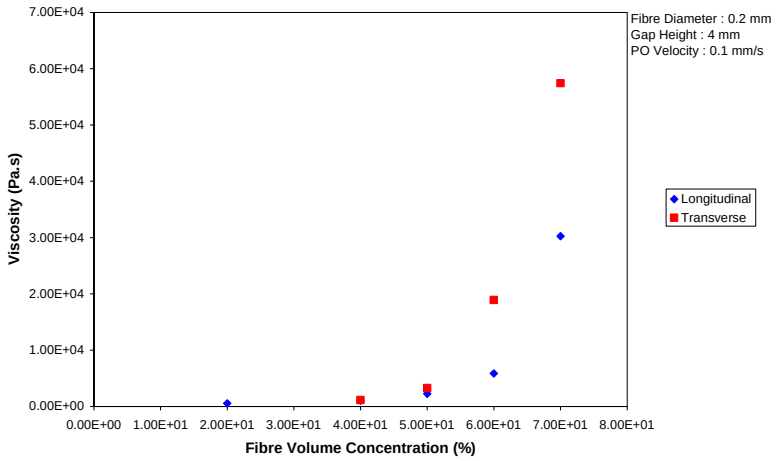


Figure 9: Viscosity versus fibre concentration at a single rate of shear – both longitudinal and transverse directions.

[1] 4 mm Gap Height was taken as the reference thickness when one compares the number of fibres through the thickness to a single ply of APC-2.

However, approaching 50% fibre volume concentration, this dependence was significantly more marked, in particular regarding the transverse behaviour. Throughout the range of fibre volume concentrations, the transverse viscosity values were greater than the corresponding longitudinal viscosity. This difference was again more apparent towards the higher values of concentration.

Roberts and Jones [5], who applied the method of dynamic testing to investigate the same composite system, also identified such a Newtonian region. They found the material to be frequency thinning with sharp decreases in longitudinal and transverse dynamic viscosities up to a given frequency, after which each viscosity levelled off. Roberts and Jones also found that for specimens of 60% fibre concentration, the transverse viscosity was significantly greater than the longitudinal viscosity, which agrees qualitatively with the data in Figure 9. Goshawk and Jones [6] carried out dead-weight loading tests to determine values for η_L and η_T. They also found the transverse viscosity to be much greater than the longitudinal viscosity for specimens of 60% concentration, but it is significant that in these tests η_L and η_T were found to be independent of shear rate, which clearly contradicts the above findings. Goshawk and Jones also reported that none of their specimens exhibited yield-like behaviour, whereas in the previous section clear indications of the existence of yielding behaviour was observed for specimens sheared along the direction of the fibres. The dynamic tests carried out by Roberts and Jones revealed an interesting variation of the ratio of the composite viscosities with fibre concentration. Surprisingly, it was found that the longitudinal dynamic viscosity η'_L was significantly higher than the transverse dynamic viscosity η'_T for fibre concentrations below 55%, while at a concentration of 55% η'_L was equal to η'_T, and for concentrations greater than 55% η'_T became much greater than η'_L. No such cross-over in viscosity values was observed by Goshawk and Jones in their dead-weight loading tests on the same model composite system. It was speculated that the disparity could be due to the relatively large strains imposed on the material in the dead-weight loading tests.

Table1: Comparison of viscosity ratios for model composites of increasing fibre concentrations.

	Current Study		**Roberts et al**		**Goshawk et al**	
Concentration %	$\frac{\eta_L}{\eta_M}$	$\frac{\eta_T}{\eta_M}$	$\frac{\eta'_L}{\eta'_M}$	$\frac{\eta'_T}{\eta'_M}$	$\frac{\eta_L}{\eta_M}$	$\frac{\eta_T}{\eta_M}$
20	7.99	*	2.73	1.93	2.85	3.71
40	14.89	16.25	5.83	4.10	7.43	26.57
50	32.14	46.38	7.76	6.71	*	*
60	83.38	268.97	11.04	12.61	134.29	262.86
70	430.23	816.63	21.37	38.44	*	*

In Table 1, the results of these previous investigations are shown along with viscosity values determined from the current study. For comparison purposes, the viscosities quoted are relative viscosities calculated by dividing the measured viscosity of the composite by the corresponding matrix viscosity η_M at the test temperature.

From the figure, it can be seen that the relative viscosities calculated in the current study and those derived by Goshawk and Jones are of the same order of magnitude, with the relative transverse viscosity greater than the relative longitudinal viscosity over the entire range of concentrations. It is significant that in the present study the ratio of the relative viscosities at 40% concentration is much closer to unity than the ratio for the data obtained by Goshawk and Jones, suggesting that at lower concentrations there might be a cross-over of viscosities.

Several authors [10-12] have put forward analyses that are concerned with determining formulae for the longitudinal and transverse shear viscosities of composites in terms of the fibre volume fraction and the shear viscosity of the matrix. The basic configuration considered in each analysis quoted is that of a composite/suspension comprising perfectly aligned, rigid fibres embedded in an incompressible Newtonian viscous liquid matrix. The fibres are understood to have a regular packing arrangement and have a large aspect ratio. Moreover, the composite system is undergoing steady shear in either along or transverse to the main axis of the cylinders. The experimental values for viscosity collected during the current study on the fibre concentration dependence are plotted together with the values predicted by the three analyses in Figure 10 and listed in Table 2. Throughout the range of fibre concentrations considered, the longitudinal viscosities are less than the transverse viscosities, which agrees qualitatively with the theories put forward by Christensen [11] and Pipes [12].

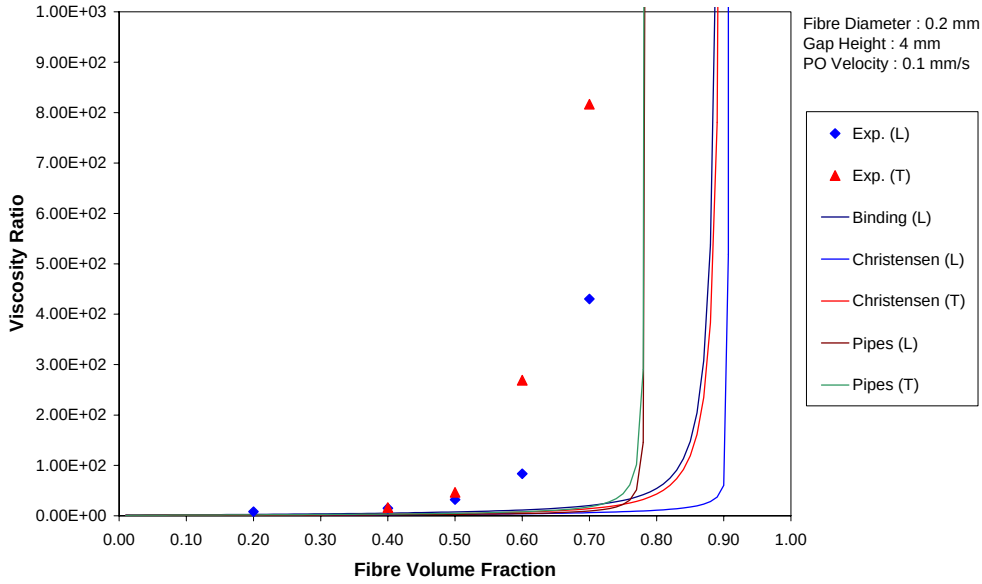


Figure 10: Comparison of the experimental and theoretical fibre concentration viscosity dependence.

Table 2: Comparison of experimental viscosity ratios with those arrived at by theoretical models, for the 0.2 mm fibre diameter model composites.

	Experiments		**Binding**	**Christensen**		**Pipes**	
Concentration %	$\frac{\eta_L}{\eta_M}$	$\frac{\eta_T}{\eta_M}$	$\frac{\eta_L}{\eta_M}$	$\frac{\eta_L}{\eta_M}$	$\frac{\eta_T}{\eta_M}$	$\frac{\eta_L}{\eta_M}$	$\frac{\eta_T}{\eta_M}$
20	7.99	*	2.84	1.50	1.58	1.51	2.02
40	14.89	16.25	5.32	2.37	2.89	2.25	3.49
50	32.14	46.38	7.54	3.08	4.31	2.97	4.95
60	83.38	268.97	11.49	4.20	7.14	4.47	7.94
70	430.23	816.63	20.34	6.20	14.23	9.44	17.88

Clearly, the values of η_L and η_T predicted by the models increasingly underestimate the measured values as the fibre concentration increases. The disparity between theory and experiment can be attributed to two factors. First, the models are based on geometrical arguments and do not take fibre-fibre interaction into consideration, which certainly does occur during the experiments and increases with fibre volume fraction. The fibre-fibre

interaction on a microscopic scale, i.e. two fibres moving with a relative velocity with an intervening Newtonian liquid, is the basic building block. This is representative of the macroscopic deformation behaviour of the whole composite. Secondly, in the models it is assumed that the fibres lie within the planes of shear and this will not necessarily be so during an experiment. If fibres become misaligned with the shear planes then a velocity gradient will exist along their length. Since the fibres are inextensible, there will be an increase in the drag on the fluid, which would translate into a higher measured viscosity. All of the theoretical models referred to base their assumptions on the idea of the continuum, which the experimental evidence certainly contradicts. Further, the viscosity ratios arrived at during a separate investigation on a real commercial composite [8], APC-2 indicate that the longitudinal value is approximately 1200 and the transverse value 500. These value are significantly greater than those predicted by the mathematical models for a fibre concentration of 60%.

CONCLUSIONS

A custom-built shear apparatus has been successfully developed and applied to investigate the steady-shear deformation of a model composite system consisting of straight, rigid fibres in a liquid, Newtonian matrix. The rheological behaviour of the composite has been investigated for different pull-out velocities and fibre concentrations with the following key results:

For high fibre concentrations, the longitudinal and transverse viscosities decrease rapidly with increasing pull-out velocity, before eventually levelling off to an approximately constant value. Calculations of the viscosities indicate that the transverse viscosity is much greater than the longitudinal viscosity over the entire range of velocities considered. The values of viscosity, for both the longitudinal and transverse directions, showed significant shear-thinning typical of a polymer melt.

Yield-like behaviour can occur when specimens are sheared along the direction of the fibres. This is most pronounced at high concentrations and low shear rates, and is believed to be caused by localised friction between neighbouring fibres. At high shear rates, these frictional contacts are easier to overcome and there is a transition to Newtonian behaviour.

Extensive re-organisation and re-orientation of fibres is observed for specimens sheared perpendicular to the fibre direction. With increasing concentration, there are indications that the properties of the composite become more solid-like.

The deformation of the composite at high shear rates and high concentration might be assisted by the formation of thin, resin-rich layers, resulting in a plug flow in which relatively little shearing occurs through the composite thickness.

The results from the model composite test series were also compared to those predicted by mathematical models. When the experimentally determined viscosity values for each fibre concentration were plotted against those predicted by the models, the models consistently underestimated the viscosities, in particular towards the higher levels of fibre concentration. It was speculated that the models describe purely idealised composite materials regarding their initial fibre distribution and the flow behaviour of perfectly aligned cylinders in a Newtonian matrix. As was evidenced during the experiments, a great deal of fibre-fibre interaction may take place during the shearing action of the composite, resulting in much larger shear forces being developed throughout the material, thus increasing the viscosity. Moreover, no model can predict the extremely complex nature of the fibre movement in the transverse direction, including the combined rotation and translation eventually leading to localised bunching and so on. Thus, the models derived to predict the viscosity of a composite, relative to the matrix viscosity, are quite limited in their characterisation of model composites, and hence, more significantly would be of limited benefit to predicting the shearing behaviour of a real commercial high fibre volume unidirectional composite system.

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