Rheological characterisation of continuous fibre composites in oscillatory shear flow

R.W.Roberts and R.S.Jones,
Department of Mathematics,
University of Wales,
Aberystwyth,
Dyfed, SY23 3BZ.

Abstract

The anisotropic rheology of a model composite consisting of a temperature-sensitive viscous liquid matrix reinforced by aligned and virtually inextensible fibres of Nylon has been studied using a custom-built linear oscillator. The composite was characterised dynamically both along and transverse to the fibre direction over a wide range of frequencies for different fibre volume concentrations. Towards the limit of zero-shear the composite was found to exhibit a yield stress. The temperature dependence of the matrix dynamic viscosity was used to study the dependence of the longitudinal and transverse dynamic viscosities of the composite on the dynamic viscosity of the matrix. The dependence of the composite dynamic viscosities on fibre volume concentration are compared with models of the steady shear dependence for aligned fibre systems that have been reported in the literature.

Keywords : *anisotropic* ; *fibre volume concentration* ; *yield stress.*

1 Introduction

One inherent property of a continuous fibre reinforced composite is that of anisotropy. The extent of the anisotropy is governed by the degree to which the fibres can be stretched together with their orientation within the matrix phase. In the special case where the fibres are highly inextensible and aligned the composite is *strongly* anisotropic with principal directions along and transverse to the fibres.

In an earlier paper [1], we reported on experiments to characterise the rheology of a strongly anisotropic model composite consisting of Nylon fibres in a viscous liquid matrix of Golden Syrup. In those experiments samples of the composite were subjected to small amplitude oscillatory shear both parallel and transverse to the fibre direction using the linear oscillator designed and built by Jones and Wheeler [2]. Longitudinal and transverse dynamic moduli were measured over two decades of frequency ($1rad/s - 100rad/s$) for different fibre volume concentrations. Preliminary results indicated that the ratio of the longitudinal dynamic viscosity η'_L to the transverse dynamic viscosity η'_T varied significantly with the volume concentration of the fibres. At a concentration of fifty-five percent, η'_L was found to be approximately equal to η'_T whereas for lower concentrations η'_L was greater th and vice-versa for higher concentrations. These observations showed consistency with experimental findings of Groves et al [3]. The dependence of η'_L and η'_T on fibre vol concentration were compared with models of the steady shear dependence for aligned fi systems put forward by Christensen [4] and Pipes [5].

The key objectives in this paper are to clarify the above results and to extend experimental work to incorporate lower frequencies. The behaviour of the Nylon fi reinforced Golden Syrup towards the limit of zero-shear is of particular interest as composite appears to exhibit a yield stress. A similar yield stress has been observed for hi modulus carbon fibre reinforced thermoplastic composites in their molten state [3], [6]-[1

2 Experiments

2.1 Apparatus

The linear oscillator used for the experimental investigations is shown in Fig. 1; a detail account of the way it operates has been given by Jones and Wheeler [2]. The instrument h

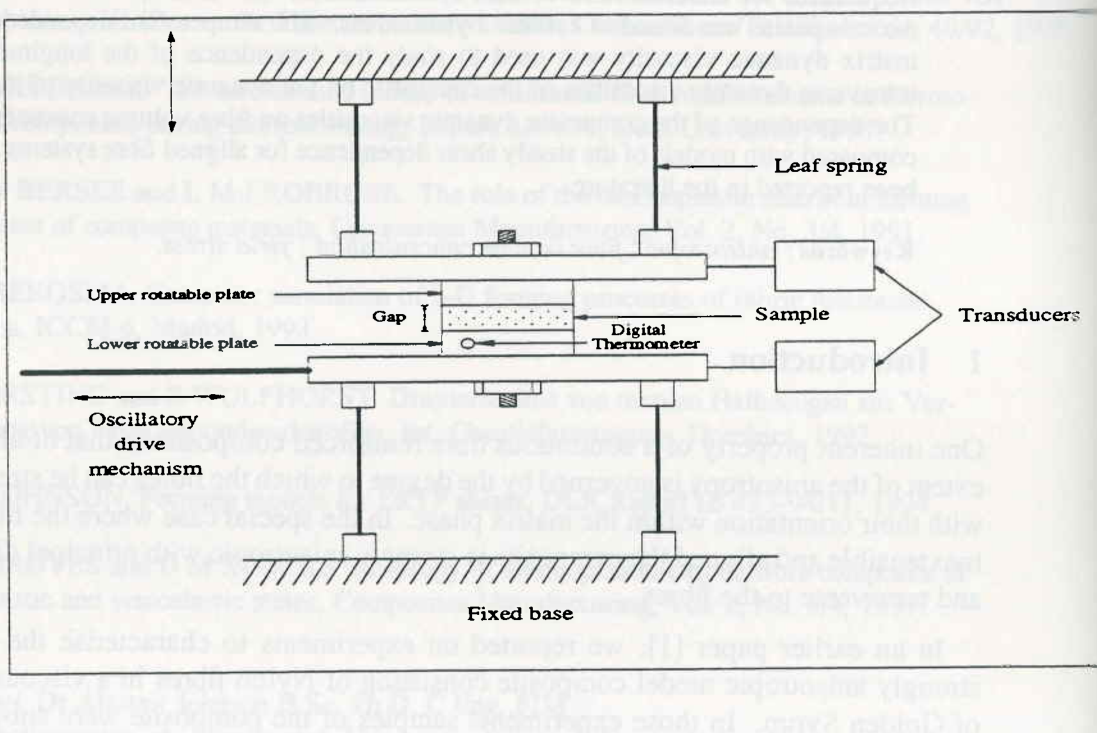

Figure 1: Schematic diagram of linear oscillator

been tested with isotropic materials and the results show excellent agreement with data giv by standard commercial rheometers.

2.2 Fibre and matrix materials for the model composite

The basic ingredients for the model composite were a temperature-sensitive Golden Syrup liquid, Newtonian in behaviour and of viscosity $70 Pa.s$ at $20°C$, and an abundant supply of straight, inextensible Nylon fibres with mean diameter $0.2 mm$, length $39 mm$ and density $1.35 g cm^{-3}$.

2.3 Sample construction

A sample of the composite was constructed by coating a pre-determined quantity of the Nylon fibres with Golden Syrup, before carefully arranging the assembly on the lower plate of the linear oscillator such that it was uniformly distributed and free from fibre misalignments. The upper plate of the instrument was then lowered to the chosen gap, and it was ensured that the sample and the plates made proper contact. By choosing an appropriate quantity of fibres it was possible to construct a sample of any chosen fibre volume concentration.

2.4 Sample linearity

Before the dynamic moduli of a sample could be correctly evaluated and interpreted it was first necessary to determine the range of input amplitudes over which the response of the sample was linear. This was achieved by examining the displacement waveforms of the upper and lower plates.

A sample of 60% fibre volume concentration and thickness $3mm$ was constructed and subjected to longitudinal and transverse shear at $20°C$ using input amplitudes $\epsilon = 0.18mm$, $0.36mm$ and $0.72mm$ (This covered the entire range of input amplitudes possible with the linear oscillator). For each value of ϵ, photographs of the displacement waveforms of the upper and lower plates were taken at frequencies $\omega = 78.91 rad/s, 1.25 rad/s$ and $0.079 rad/s$ (Figs. 2 and 3). Where the waveforms were sinusoidal measurements were carried out at different amplitudes, and it was found that the dynamic response was independent of the amplitude indicating a linear response.

- Longitudinal shear

  In this case it can be seen that for each value of ϵ the displacement waveform of the upper plate is sinusoidal at the high frequency ($\omega = 78.91 rad/s$), where the response is linear, but flat-topped at the low frequencies ($\omega = 1.25$ and $0.079 rad/s$), an effect consistent with a yield stress [11]. The yield stress was estimated from the output waveforms and found to be approximately $50 Pa$.

- Transverse shear

  At the smallest input amplitude (0.18mm) the response is linear for each frequency. However for $\epsilon = 0.36mm$ and $\epsilon = 0.72mm$ the output displacement waveforms are non-sinusoidal at the low frequencies, and the sample appears to behave non-linearly. This non-linear behaviour at large strain levels is in line with the theory for oscillatory shear [12].

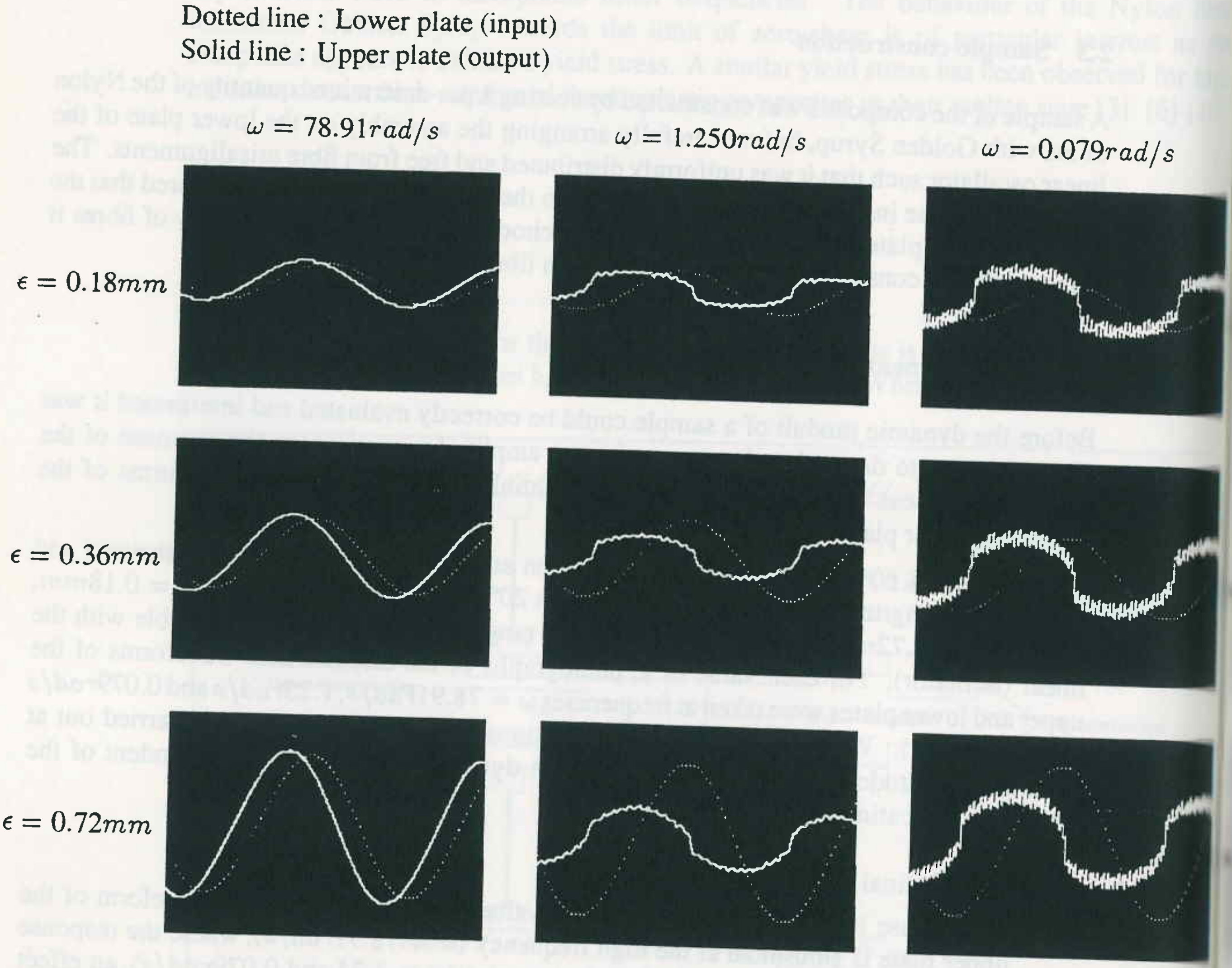

Figure 2: Displacement of linear oscillator plates (Longitudinal shear)

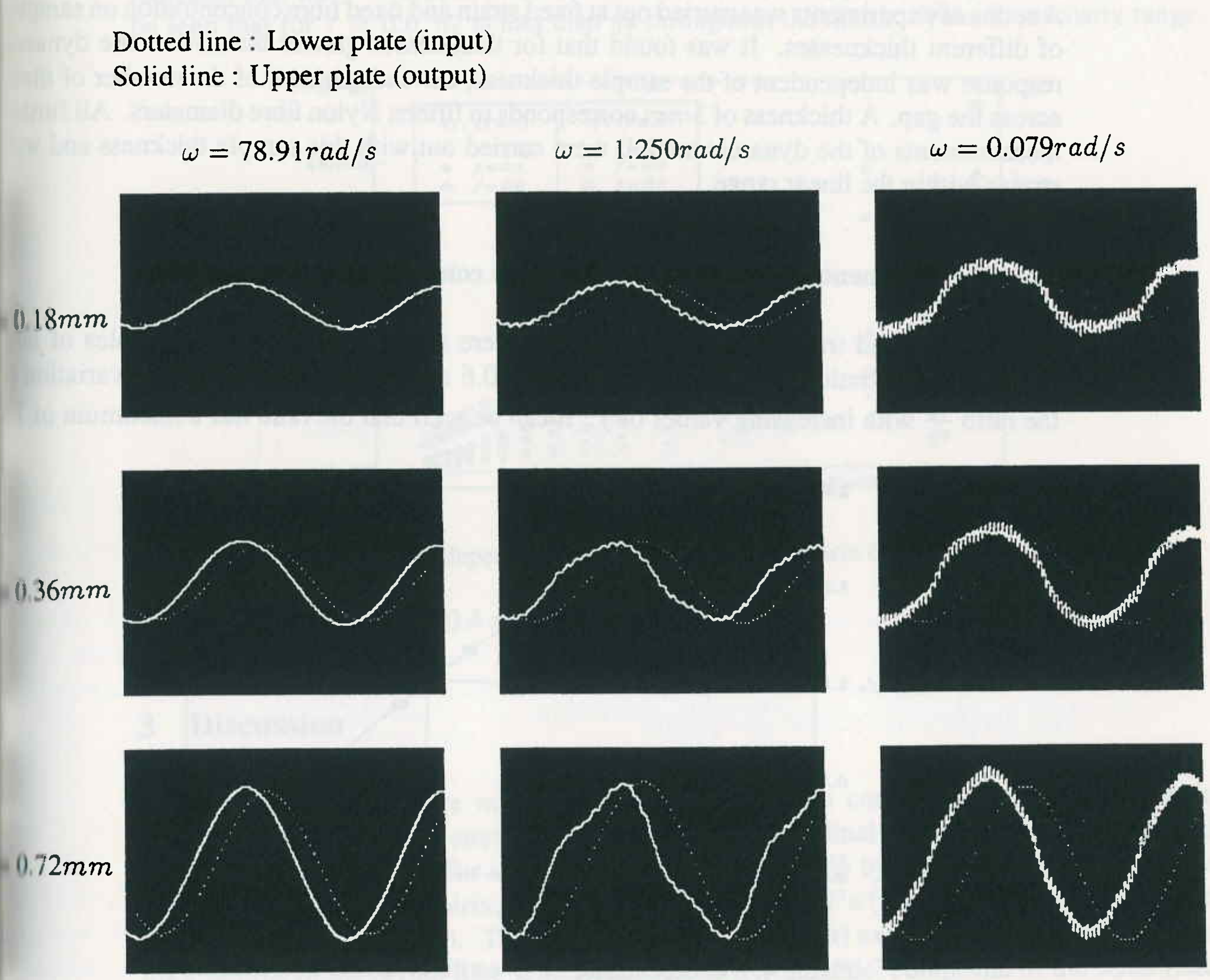

Figure 3: Displacement of linear oscillator plates (Transverse shear)

Similar results were obtained for a sample of 40% fibre concentration, which was f to have a yield stress of around $9Pa$. No significant yield stress was observable for a sam of 20% fibre concentration.

2.5 Conditions for measuring dynamic moduli

A series of experiments was carried out at fixed strain and fixed fibre concentration on sam of different thicknesses. It was found that for thicknesses greater than $3mm$ the dyn response was independent of the sample thickness, *i.e.* independent of the number of fi across the gap. A thickness of $3mm$ corresponds to fifteen Nylon fibre diameters. All fur measurements of the dynamic moduli were carried out with this sample thickness and w strains within the linear range.

2.6 Measurements for different fibre volume concentrations

Longitudinal and transverse dynamic moduli were measured at 20°C for samples of fi volume concentrations f = 0.2, 0.3, 0.4, 0.5, 0.6 and 0.7. Fig. 4 shows the variation the ratio $\frac{\eta'_L}{\eta'_T}$ with increasing values of f. It can be seen that the ratio has a maximum of

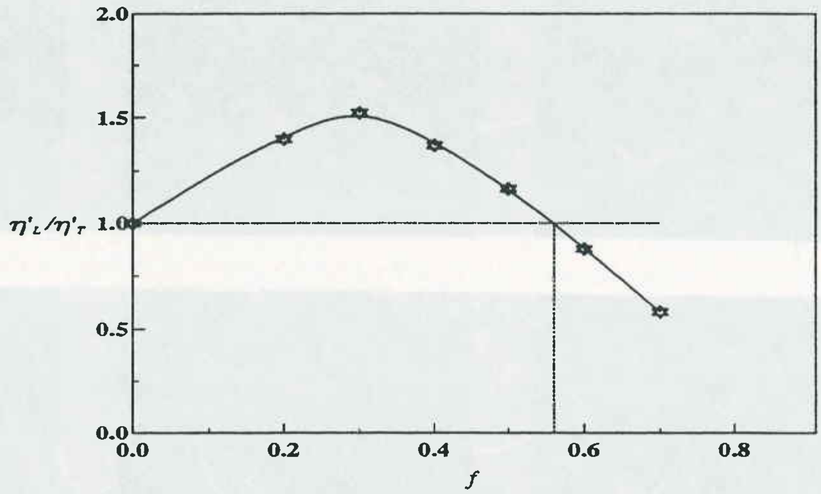

Figure 4: The dependence of $\frac{\eta'_L}{\eta'_T}$ on fibre volume concentration

when $f = 0.3$, reduces to 1 when $f = 0.55$ and becomes less than 1 for higher values of The ratio of the longitudinal dynamic rigidity G'_L to the transverse dynamic rigidity G'_T, w found to be less than 1 for all the fibre concentrations.

Intermediate dynamic moduli between longitudinal and transverse were also measured rotating the plates of the linear oscillator *in situ*. For each value of f, it was found that change in moduli from longitudinal to transverse shear was virtually uniform.

2.7 Changing the matrix dynamic viscosity

The dynamic response of Golden Syrup is very sensitive to temperature and is well characterised [1]; temperature can therefore be used as a means of varying the matrix viscosity. Measurements were carried out on the composite at different temperatures and the dependence of the dynamic moduli of the composite on the matrix dynamic viscosity was obtained. Fig. 5 indicates that for $f = 0.2$, 0.4 and 0.6 the relationship is approximately linear. It can also be seen that for $f = 0.6$, η'_L is less than η'_T throughout the matrix dynamic viscosity range.

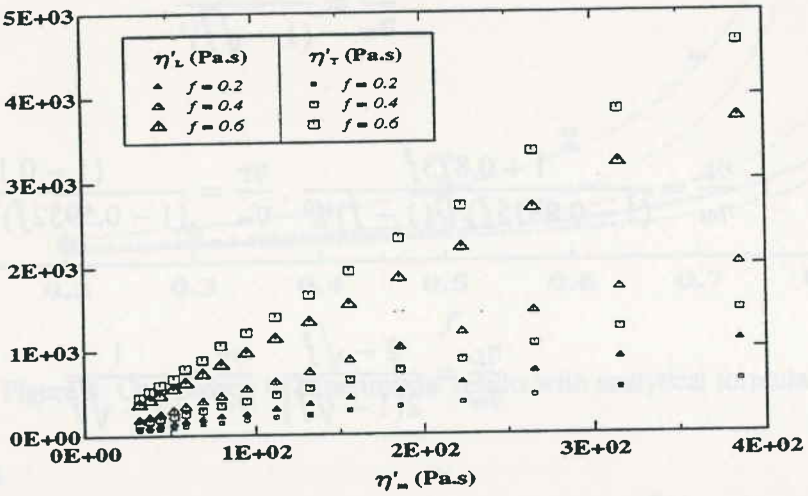

Figure 5: The dependence of η'_L and η'_T on the matrix dynamic viscosity

Similarly for $f = 0.2$ and 0.4 η'_L remains greater than η'_T.

3 Discussion

In the literature, there are numerous reports that molten continuous fibre thermoplastic composites exhibit a yield stress when subjected to longitudinal shear [3], [6]-[10]. Reported values of the yield stress for a composite consisting of 60% by volume of carbon fibres in a polyetheretherketone matrix (CF/PEEK) range from $450Pa$ (Scherer and Friedrich [9]) to $1000Pa$ (Groves et al [3]). This range ($450Pa - 1000Pa$) exceeds our estimate of $50Pa$ for a sample of the Nylon fibre reinforced Golden Syrup model composite of the same fibre concentration. It must be noted however that there is a significant difference between the dynamic viscosity of the molten PEEK matrix (approximately $200Pa.s$) and the Golden Syrup matrix ($70Pa.s$).

Groves et al [3] also measured the dynamic moduli of the 60% fibre concentration CF/PEEK and their results show that $\eta'_L < \eta'_T$ and $G'_L > G'_T$. However for a glass fibre reinforced polypropylene composite composite of 35% fibre volume concentration they found that $\eta'_L > \eta'_T$ and $G'_L > G'_T$. More recently Sengupta and Mukhopadhyay [13] carried out dynamic tests on carbon fibre reinforced polypropylene composites of fibre concentrations 13%, 24% and 35%, and they found that $\eta'_L > \eta'_T$ and $G'_L > G'_T$ for each concentration.

From these investigations it appears that the dependence of the dynamic viscosities on fi concentration is consistent with our experiments (Fig.4) but the dependence of the dyna rigidities is not.

Analytical formulae relating the longitudinal and transverse *steady* shear viscositie and η_T of an aligned fibre composite to its fibre volume concentration f have been deriv by Binding [14], Christensen [4] and Pipes [5]. For a hexagonal array of fibres, the sugges formulae are

$$\frac{\eta_L}{\eta_m} = \frac{1-f}{(1-\sqrt{\hat{f}})^2}$$

(1

(Bindin

$$\frac{\eta_L}{\eta_m} = \frac{1+0.873\hat{f}}{(1-0.8815\hat{f})^{1/2}(1-\hat{f})^{1/2}}, \quad \frac{\eta_T}{\eta_m} = \frac{(1-0.193\hat{f})^3}{(1-0.5952\hat{f})^{3/2}(1-\hat{f})^{3/2}}$$

(

(Christens

$$\frac{\eta_L}{\eta_m} = \frac{2-\sqrt{\hat{f}}}{2(1-\sqrt{\hat{f}})}, \quad \frac{\eta_T}{\eta_m} = \frac{1}{1-\sqrt{\hat{f}}}$$

(

(Pipe

where $\hat{f} = \frac{2\sqrt{3}}{\pi} f \; (0 \leq f \leq \frac{\pi}{2\sqrt{3}})$ and η_m is the matrix viscosity. These expressions can b compared with the experimental data provided it is assumed that the fibre arrangement in th samples was approximately hexagonal and that the dependence of the dynamic viscosities o fibre volume concentration has the same form as that for the steady shear viscosities. Th comparison is given in Fig. 6 from which it can be seen that the prediction of Binding [1 best fits the experimental data points.

4 Summary

(i) The response of the model composite when sheared parallel to fibre direction ca be described as visco-plastic. Below a critical yield stress the composite behav like an elastic solid, while above the stress the composite flows and its behaviour i comparable to an elastico-viscous liquid. In transverse shear the composite exhibi non-linear behaviour at large strain levels.

(ii) The ratio $\frac{\eta_L'}{\eta_T'}$ has a maximum of 1.5 when $f = 0.3$, reduces to 1 when $f = 0.55$, an becomes less than 1 for higher values of f.

(iii) For a given fibre volume concentration, the longitudinal and transverse dynami viscosities of the composite appear to depend linearly on the matrix dynamic viscosit

(iv) The dependence of the dynamic viscosities on fibre volume concentration have bee compared with theoretical models.

Ref
[1]
[2]
[3]
[4]
[5]
[6]
[7]
[8]
[9]
[10]
[11]
[12]

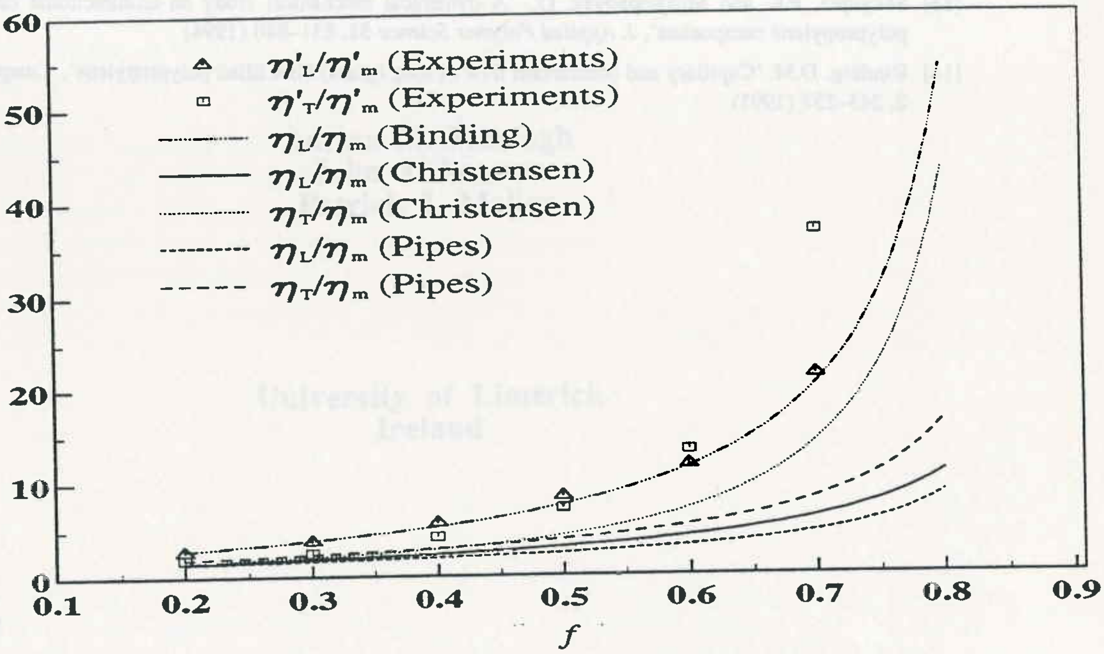

Figure 6: Comparison of experimental results with analytical formulae

References

[1] Jones, R.S. and Roberts, R.W., 'Anisotropic shear flow in continuous fibre composites', *Composites* **25**, 171–176 (1994)

[2] Jones, R.S. and Wheeler, A.B., 'A characterization of anisotropic shear flow in continuous fibre composite materials', *Composites Manufacturing* **2**, 192–196 (1991)

[3] Groves, D.J., Stocks, D.M. and Bellamy, A.M., 'Isotropic and anisotropic shear flow in continuous fibre thermoplastic composites', *Proc. Golden Jubilee meeting of the British Society of Rheology and Third European Rheology Conference*, Edinburgh, UK, 190–192 (1990)

[4] Christensen, R.M. 'Effective viscous flow properties for fibre suspensions under concentrated conditions', *J. Rheology* **37**, 103–121 (1993)

[5] Pipes, R.B. 'Anisotropic viscosities of an oriented fibre composite with a power-law matrix', *J. Composite Materials* **26**, 1536–1552 (1992)

[6] Groves, D.J., 'A characterization of shear flow in continuous fibre thermoplastic laminates', *Composites* **20**, 28–32 (1989)

[7] Groves, D.J., Bellamy, A.M. and Stocks, D.M., 'Anisotropic rheology of continuous fibre thermoplastic composites', *Composites* **23**, 75–80 (1992)

[8] Cogswell, F.N. 'The processing science of thermoplastic structural composites', *Int. Polymer Processing* **1**, 157–165 (1987)

[9] Scherer, R. and Friedrich, K., 'Inter- and intraply-slip flow processes during thermoforming of CF/PP-laminates', *Composites Manufacturing* **2**, 92–96 (1991)

[10] Mulholland, A.J., Monaghan, M.R. and Mallon, P.J., 'Characterisation of consolidation flow processes in continuous fibre reinforced thermoplastic composites', 13^{th} *International Conference and Exhibition*, European Chapter, SAMPE, May 11^{th}–13^{th} (1992)

[11] Nguyen, Q.D. and Boger, D.V., 'Measuring the flow properties of yield stress fluids', *Annual Rev. Fluid Mech.* **24**, 89–115 (1992)

[12] Walters, K.W., 'Rheometry', Chapman and Hall (1975)

[13] Sengupta, P.K. and Mukhopadyay, D., 'A dynamical mechanical study on unidirectional carbon fibre-rein polypropylene composites', *J. Applied Polymer Science* 51, 831–840 (1994)

[14] Binding, D.M. 'Capillary and contraction flow of long (glass) fibre filled polypropylene', *Composites Manufactu* 2, 243–252 (1991)

.bstract

in the pre
formed in
or by a rul
rubber mo
surface ur
and meas
GF-fabric
materials.
platens, be
sheared fr
velocity sh
also built
across a h
pressure,
adhesive b
By varyin
surface v