5th Intl. Conf. Flow Processes in Comp. Materials, Univ. Plymouth, 12-14th July, 1999

RHEOLOGICAL CHARACTERISATION OF DISCRETE LONG GLASS FIBRE (LGF) REINFORCED THERMOPLASTICS

S.F. Bush, F.G. Torres, J.M. Methven, S. Al-Araj and G. Lik Chin
UMIST Polymer Engineering Research Laboratory
PO Box 88, Manchester M60 1QD, England

Abstract

In the study reported here two experimental techniques have been employed to assess the extensional behaviour of LGF reinforced thermoplastics : the Squeeze Load Test (SLT) and the Bubble Inflation Test (BIT). Both tests are analogues of extensional flows in important full-scale processes : compression moulding (SLT), blow moulding (BIT) and thermoforming (BIT) in particular. For comparison shear flow results obtained from rotational viscometry are also reported, since such flows are invariably present at some point in all processes.

1. INTRODUCTION

Long glass fibre reinforced melts exhibit particular rheological characteristics which differentiate them from unreinforced polymer melts and from their short fibre analogues. In the design of manufacturing processes for LGF composites, it is highly desirable if not indispensable to have a good description of the rheological behaviour of the material under different flow conditions. It is well known that the presence of fibres in a polymer matrix increases the viscosity of the unreinforced polymer in both shear and extensional flow (Bush, 1993, 1997, Torres, 1997). Short fibres (0.1-1.0mm), widely used as reinforcement for thermoplastic matrices, tend to align themselves in the flow direction and the increase in melt viscosity relative to the unreinforced polymer is less dramatic than in the case of long fibres. When assisted by fibre management devices (Bush 1993,1995) long fibres form coherent mat structures in the melt. This can be made to occur for all the conventional processing techniques (Bush 1997) under different types of flow conditions. The formation of these fibre mats depends on the average number of fibre touches N given by (Bush 1993)

$$N = A.c.l/d \qquad (1)$$

where A is approximately $8/\pi^2$ in the case of a 2D random distribution, c is the fibre volume fraction, l and d are the length and diameter of the fibres respectively.

In the following experiments c= 0.03 or 0.06, l is typically around 5mm and d is 17μm. Eq. (1) for the random case then gives an average number of touches N, 7 or 14. 5 touches is about the minimum for a coherent mat structure.

The following sections include a variety of rheological techniques used to assess the rheological behaviour of LGF reinforced thermoplastics under different flow conditions. Section 2 presents the Squeeze Load Test (SLT) which combines shear and extensional flow. However as discussed later in this paper, this technique can be used to deduce extensional properties on their own. Section 3 presents the Bubble Inflation Test (BIT) which simulates the inflation of a membrane under controlled conditions and is used in the simulation of both the thermoforming and blow moulding processes. In Section 4 for comparison we present simple shear viscosity results obtained in a cone and plate viscometer.

2. SQUEEZE LOAD TEST (SLT)

2.1 Background

The squeeze load test (SLT) has been used in recent years in the compression moulding simulation of sheet moulding compounds (SMC), glass mat thermoplastics (GMT), and other fibre reinforced materials (Kotsikos et al 1996),(Michaeli and Starke, 1993), (Oelgarth, 1997). The main advantages of the method are its similarity to the compression moulding process and its operational simplicity. The main disadvantage is that both shear and extensional flow take part in the deformation of the material under compression, and that it is not easy to determine the relative importance of each type of flow. In the case of discrete LGF polymer composites, this technique allows for a good and simple comparative measurement of the rheological properties of the composite taking into account the macroscopic flow of the reinforcing fibre mat structure (Torres 1997).

2.2 Experimental

The test has been designed as isothermal and axisymmetric. Circular plates of 100mm of diameter have been used. The plates were heated with cartridge heaters (200W per plate) and the temperature was controlled using independent self-tuning three phase controllers. The time response of the controllers had to be matched accurately to the load of the system in order to achieve stable temperature readings. Thermocouples were inserted into the plates. To enhance the extensional flow characteristics release agent was sprayed on to the plates. Insulation discs and supports were designed to mount the whole assembly on a Hounsfield tensile testing machine. A load cell of 5kN was used in most of the tests. The assembly of the rig including a detail of the plates used in the test is shown in Fig. 1.

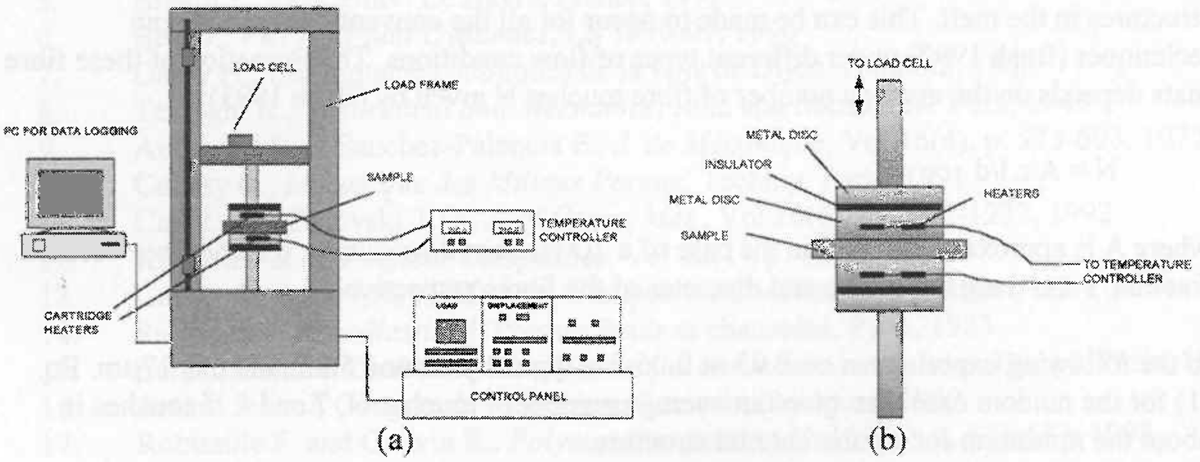


Fig. 1. Squeeze load test. (a) General arrangement. (b)Detail of the plates

The tests have been carried out at three temperatures : 180C, 200C, and 230C spanning the typical PP processing range. The materials were polypropylene homo- and copolymer at two different long fibre concentrations (3% , 6% v/v) which are known to provide very significant increases in mechanical properties (Bush et al 1995). For each temperature and fibre concentration (including the zero concentration case) four plate-closing velocities (u) were set on the crosshead (Fig. 1(a)). For each velocity (u), the squeezing force (F) was recorded for a range of plate separations between 6 and 3mm (Fig. 1(b)).

2.3 Results and Discussion

The results have been arranged to display an apparent squeeze viscosity $[\eta]_s$ *defined* by

$$[\eta]_s \equiv (F / \pi R^2) / \varepsilon_s \quad (2)$$

where R is the radius of the plates (Fig 1(b)), and the squeeze strain rate is

$$\varepsilon_s \equiv u/h \quad (2a)$$

The results at 230° C and a plate separation (h) of 4.8mm are shown in Fig. 2.

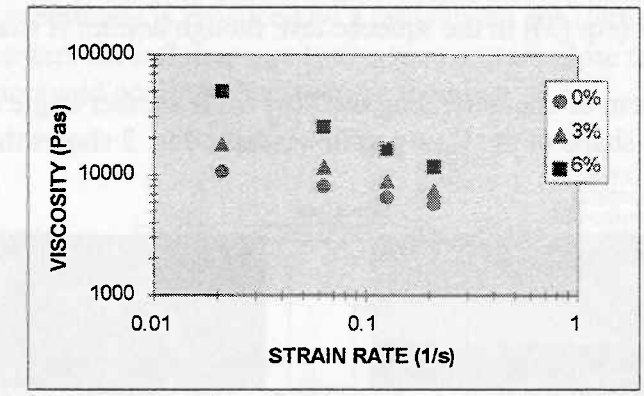


Fig. 2. Experimental results of viscosity vs strain rate for LGF-PP (1100L) at 230C

Results at the other plate separations (h) show a sharp increase in $[\eta]_s$ as h decreases, and consistent increases in $[\eta]_s$ of up to 3 times relative to the unreinforced polymer can be observed at 6% v/v, for a given h and T.

If it is assumed that shear stresses dominate the squeezing process, then $[\eta]_s$ can be found theoretically for a power law fluid from the equation of Scott (1935) as

$$[\eta]_{SS} = \frac{A_{SS}}{(m+3)}\left(\frac{2m+1}{m}\right)^m \left(\frac{R}{h}\right)^{m+1} \left(\frac{u}{h}\right)^{(m-1)} \quad (3)$$

where

$$\tau_{rx} = A_{SS} (\gamma_{rx})^m \quad (4)$$

is the power law relating τ_{rx} and γ_{rx}, the shear stress and strain rate on the planes normal to a radius (r) (Fig. 1(b)).

From Fig. 2 it may be calculated that on this basis m changes from 0.7 at 0% fibre to 0.35 at 6% v/v, indicating a very marked change in rheology over this concentration range. This reduction in m is found consistently for all h and T values.

It is found from eq. (3) and Fig. 2 that $[\eta]_{SS}$ depends on $(R/h)^{1.7}$ to $(R/h)^{1.35}$. The measured squeeze viscosity $[\eta]_s$ defined by eq. (2) was found to increase as the gap h decreased although the exponent of (R/h) could not be reliably determined. However, A_{SS} in eq. (4) *can be evaluated* from Fig.2 and gives a value of 150 at 0% and 200 at 6% v/v. If it is assumed that extensional flow dominates the squeezing process, then it is easily shown that for the power law fluid (eq. (4)), the squeezing viscosity is given by

$$[\eta]_{se} = 3A_{se}\,(u/h)^{m-1} \tag{5}$$

where $$\tau_{rx} = A_{se}\,(\gamma_{rx})^{m} \tag{6}$$

Then A_{se} in eq. (6) can be evaluated from Fig.2 and gives a value of 1000 at 0% and 1400 at 6%. Clearly the two models eq(3) and eq(5) give widely different results.

In section 4 below it is shown that extensional flow (eq. (5)) is a much closer model of the results than shear flow (eq. (3)) in the squeeze test, though neither is exact.

To approach the problem of characterising the flow from another angle different colours have been used to show the shape of the flow profile visually. Fig. 3 shows the colour interfaces in two cases.

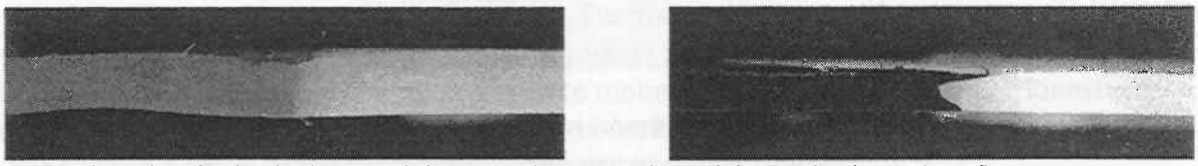


Fig.3. Flow visualisation in the SLT, left: predominant plug flow, right: predominant shear flow

In both cases the flow direction is from right to left. The figure on the left shows a typical plug profile, as obtained at a compression of 25%. The figure on the right shows a typical parabolic profile as obtained at a higher compression ratio (75%). This suggests that the flow varies from extensional flow (eq. (5)) to shear flow (eq. (3)) as the compression ratio increases. The results in Fig. 2 were obtained at h = 4.8 mm – a 25% compression ratio i.e. where plug flow is predominant.

3. BUBBLE INFLATION TEST (BIT)

3.1 Background

The inflation of membranes has been used in the past for comparative and quantitative measurement of the biaxial elongational viscosity of polymers. Denson and Gallo (1971) and Joye et al.(1972a,b) presented some of the earlier results on elongational viscosity of polymers using this method. Earlier still Treloar (1944) used the bubble inflation method to study the biaxial extension of elastic rubber sheets. More recently Schmidt and Carley (1975) used the inflation method on heat softened plastic sheets made from amorphous polymers, and they include a comparison between experimental results and numerical calculations. Tonkin (1998) used the bubble inflation test to characterise the blow moulding behaviour of LGF polymer composites at processing temperatures. This latter work was aimed at obtaining a qualitative understanding of the blowing characteristics of LGF polymer composites for practical blow moulding processes but no quantitative data were obtained.

One of the main advantages of the BIT is its direct applicability in the simulation of free surface processes such as extrusion film blowing, blow moulding and thermoforming. In the case of thermoforming of LGF polymer composites, it is necessary to characterise the elongational behaviour of the materials in conditions similar to the actual processing conditions, i.e. 2D extensional flows in the vicinity of the polymer melting point. Viscosity measurements obtained with capillary rheometers are useless for this purpose because they represent one dimensional shear flows.

3.2 Experimental Set Up

Fig. 4 shows the experimental set up used for the inflation of the LGF composites. Heating is achieved in the experiments presented here by putting the rig in a temperature controlled oven. Inflation of the membrane is produced by a pressurised gas (Nitrogen). It is desirable that the inflating gas enters the inflation cavity at a temperature close to the testing temperature in order to avoid cooling effects on the membrane.

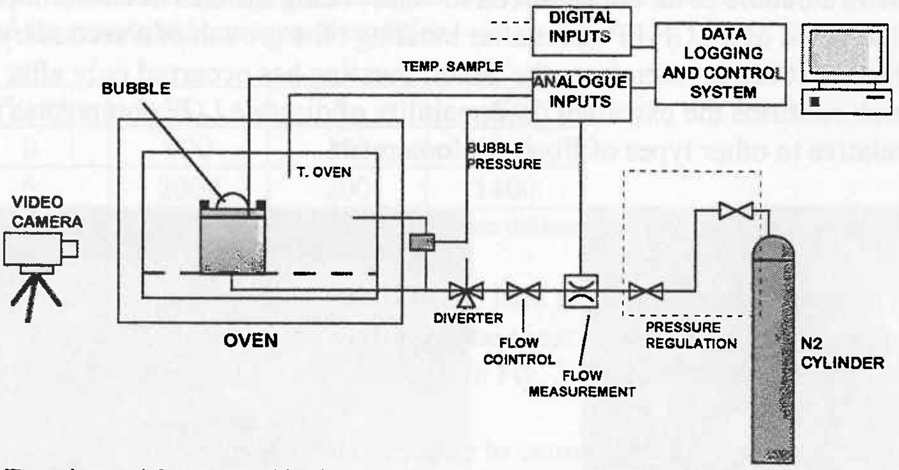


Fig. 4. Experimental Set up used in the Bubble Inflation Test (BIT) (adapted from Neilson, 1999)

In principle experiments may be carried out at (a)constant stress in the membrane, (b) constant strain rate, (c) constant inflation pressure and (d) constant gas flow rate. In practice, constant inflation pressure is by far the easiest to manage experimentally. Two types of samples were used in the experiments : (i) samples cut from extruded sheets with a thickness t of approx. 1.5mm, and (ii) injection moulded samples (t = 3mm).The materials used in the experiments were the same as for the SLT. (section 2.2)

3.3 Results of the BIT experiments

The variations of pressure and flowrate with time during a bursting test have been plotted in Fig.5. In order to carry out the experiment at constant pressure, it can be seen that it is necessary to reach a pressure plateau. From the same trace the bursting time can be determined. It can be observed that the burst time (t_b) for the 6% LGF reinforced material is around five times that for the 3% case. At constant pressure inflation, the time to a given strain is proportional to the extensional viscosity. The sharp increase in t_b, with fibre concentration seen in Fig. 5 is thus consistent with increases in the squeeze viscosity found at low strain rates (< .02 sec^{-1}) in Fig 2.

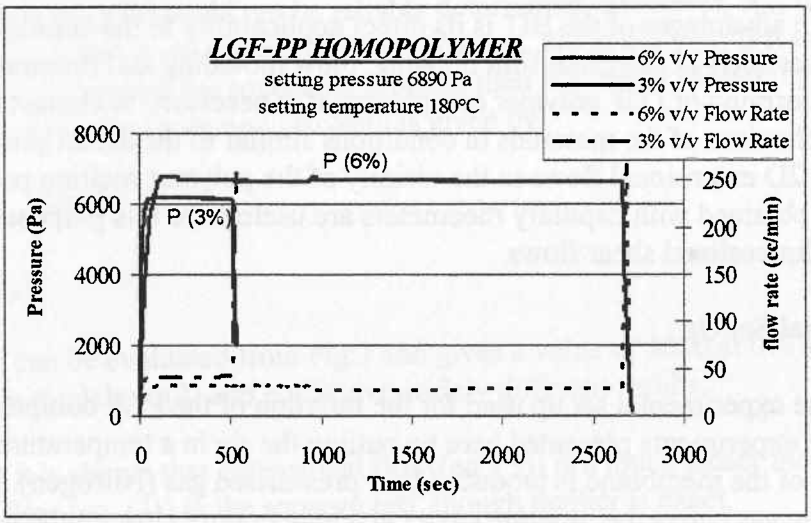


Fig. 5. Pressure, flowrate and temperature plots in the BIT in a constant pressure experiment

Fig. 6. a) shows a bubble of an unreinforced PP sheet being inflated at constant pressure. Fig. 6.b) shows the detail of a LGF-PP sheet after bursting : the growth of a secondary bubble is clearly visible. As it can be seen from the detail, bursting has occurred only after significant drawing which confirms the excellent deformability of discrete LGF composites in stretching processes, relative to other types of fibre reinforcement.

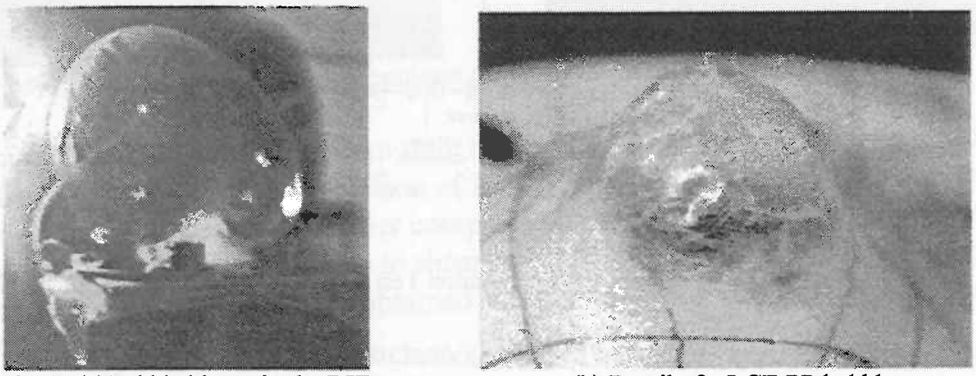
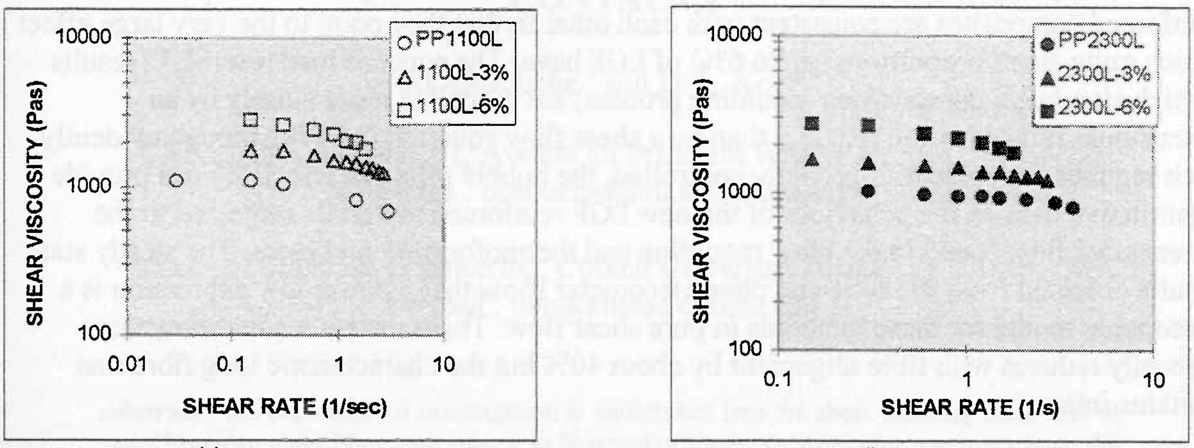


Fig. 6. (a)Bubble blown in the BIT (b) Detail of a LGF-PP bubble

4. PURE SHEAR EXPERIMENTS

Both transient and steady state shear viscosity measurements have been carried out using a cone and plate Weissenberg Rheogoniometer Carri Med R20, the cone tip being truncated. Discs of approximately 45mm were cut from extruded LGF reinforced sheets and placed on the plate. The temperature was raised to 230C. The materials used were the same compositions as those used for the SLT and BIT reported above.

The results for the different materials tested are presented in Figs. 7. a) and b). The viscosities found for the unreinforced polymers are in agreement with standard viscosity data for the two grades of PP. The increases in shear viscosity due to the presence of long glass fibres is approximately 60% at 3%(v/v) and 150%-180% at 6%(v/v) relative to the unreinforced polymer.



(a) (b)

Fig. 7. Shear viscosity curves for PP (a) homo- and (b) copolymer with 3% and 6% glass fibres by volume

From Fig. 7(a) we obtain values A_s in the viscosity power law (eq. (4)). These are compared in Table 1 with the values A_{ss} and A_{se} obtained above from the two different models (eq.(3) and eq.(5)) of the measured squeeze viscosities.

c % v/v	A_s	A_{ss}	A_{se}
0	900	150	1000
6	2000	200	1400

Table 1. Comparison of power law constants A obtained from different models and experiments

Clearly the extensional flow model of the squeeze load test at 25% compression (Fig. 2) indicated by A_{se} is much closer to the truth than the shear flow model indicated by A_{ss}. This result conforms well with the flow visualisation in Fig. 3 (left).

The rotational motion may be expected over time to cause some preferential alignment of the fibres in the circumferential direction, thus reducing the shear viscosity (Torres, 1997). Fig. 8 shows that this is indeed the case. However Fig. 9. shows that despite this deformation, the number of touches N (eq. (1)) is adequate to maintain the characteristic mat structure.

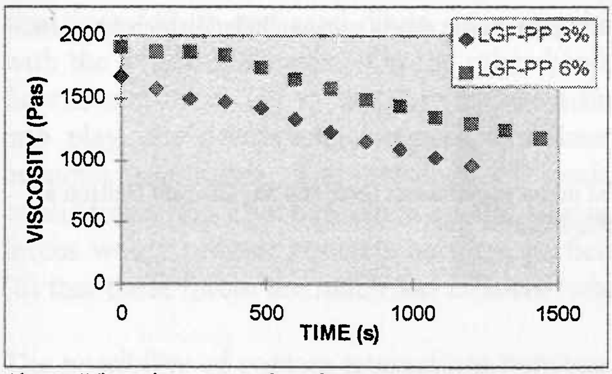


Fig. 8. Viscosity versus time for LGF-PP at 3% and 6%

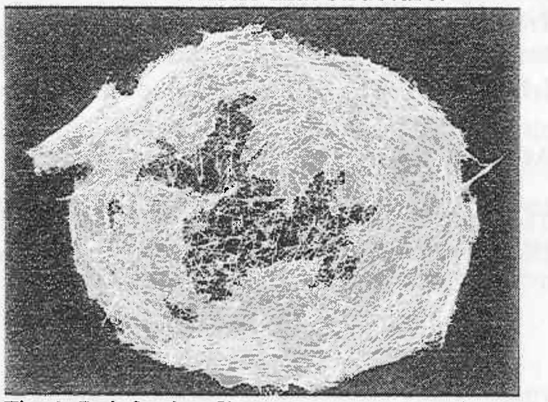


Fig. 9. Reinforcing fibre mat structure obtained after burn-off from a rotational viscometry sample

5. CONCLUSIONS

The three experimental techniques used to characterise the rheological behaviour of LGF reinforced composites are consistent with each other in that they point to the very large effect which quite small proportions (up to 6%) of LGF have. The squeeze load test (SLT) results (which simulate a compression moulding process) are modelled more closely by an extensional flow equation (eq. (5)) than by a shear flow equation (eq. (3)), though evidently both regimes are present. If properly controlled, the bubble inflation test (BIT) can provide quantitative data on the behaviour of the new LGF reinforced materials subjected to the extensional flow found in the blow moulding and thermoforming processes. The steady state results obtained from the cone and plate viscometer show that a power law expression is a reasonable model for these materials in pure shear flow. The transient results show that viscosity reduces with fibre alignment by about 40% but the characteristic long fibre mat remains intact.

REFERENCES

Bush, S.F, Fibre reinforced polymer compositions and process for the production thereof. US Patent 5 264 261 (1993)

Bush, S.F.: Long glass fibre reinforcement of thermoplastics : Results from injection and blow moulding, sheet and pipe extrusion. Poly. Proc. Soc. Euromeeting, Gothenburg, Sweden, 19-21 Aug. 1997

Bush, S.F. Yilmaz, F., Zhang, P.F. Impact Strengths of injection moulded polypropylene long glass fibre composites. Plast. Rubb. Comp. Proc. App. **24** (1995) 139-147

Denson, C.D., Gallo, R.J.: Measurements on the biaxial extension viscosity of bulk polymers : The inflation of a thin polymer sheet. Polym. Eng. Sci. 1971;**11**(2) : 174-176

Joye, D.D., Poehlin, G.W., Denson, C.D., A bubble inflation technique for the measurement of viscoelastic properties in equal biaxial extensional flow. II. Trans. Soc. Rheol. 1973 ; **17**(2) : 287-302

Kotsikos, G., Bland, J.H., Gibson, A.G., and Chandler, H.W., Squeeze flow testing of glass mat thermoplastic material, Composites Part A, **27A** (1996), 1195-1200

Michaeli, W. and Starke, J., Measurement of the viscosity of Glass Mat or long fibre reinforced thermoplastics by compression molding.SPE Antec Tech. Papers, 1993, **39**, p.3125

Neilson, G., Polymer Engineering Research Laboratory Report 99/3. UMIST, 1999

Oelgarth, A.: Analyse und Charakterisierung des Fliessverhaltens langfaserverstaerkter Pressmassen. Dissertation RWTH Aachen, 1997

Scott, J.R., Trans. Inst. Rubb. Ind. **10** (1935) 481-93

Tonkin, J.D., Recycling of thermoplastics. EngD Thesis, UMIST, Manchester, UK, 1998

Torres, F.G.: M.Phil. Thesis, Polymer and polymer composite flows : Modelling of molecular chain conformations and reinforcing fibre orientations. UMIST, Manchester, UK, 1997

Treloar, L.R.G., Trans. Inst. Rubb. Ind., **19**, (1944), p.201

Schmidt, L.R. and Carley, J.F., Biaxial stretching of heat-softened plastic sheets using an inflation technique. Int. J. Engng. Sci. 1975; **13** : 563-578

ACKNOWLEDGEMENTS

The authors want to thank Targor™ for the PP grades used in the experiments here, and Mr Graham Neilson at UMIST PERL for his help in the development of the Bubble Inflation Rig.