



Rheological measurement methods for fibre filled plastics

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ABSTRACT

A review is presented of various rheological measurement methods that are suitable for characterising the flow behaviour of fibre filled plastics. The advantages and disadvantages of the various methods are discussed. Variation in the fibre content of a material has greatest effect on its extensional flow behaviour rather than on its shear flow behaviour. Methods for characterising extensional flow behaviour of fibre filled plastics are also examined. Applications for rheological data include the simulation of injection and compression moulding, resin transfer moulding and for cure monitoring.

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1 INTRODUCTION

An understanding of the rheological (flow) behaviour of plastics is necessary if one is to optimise the process of converting the raw material into the final moulded product. The ability to characterise reliably the rheological properties of plastics is essential to aid that understanding and thus to aid the manufacturer in processing. The rheological properties of materials are needed, for example, in predicting the flow behaviour of fibre filled thermoplastics or thermosetting materials into the mould cavity in injection moulding. Furthermore, characterisation of the rheological behaviour of thermosetting plastics as a function of the curing cycle will aid processors in injection, compression and resin transfer moulding.

The benefits to be obtained from an improved understanding of the flow properties of the materials are improved process and mould design, optimised materials selection, reduced set-up times, reduced cycle times, improved product properties and the use of more relevant quality control techniques.

This paper examines the measurement methods for characterising the flow behaviour of fibre filled plastics. These methods can be subdivided into three main categories:

- * shear flow, large amplitude displacement measurements;
- * extensional flow measurements;
- * shear flow, small amplitude displacement measurements.

The first two categories cover those techniques in which the material is subjected to very large strains that are typical of polymer processing. The last category covers those techniques that are suitable for characterising the rheological behaviour of curing systems.

All these measurement techniques can be further subdivided into two groups: those that provide fundamental properties data and those that provide a relative measure of some aspect, or aspects, of the processability of the material. For example, capillary extrusion and steady shear rotational rheometry yield shear viscosity values and thus fall into the former group. In the latter group are, for example, the cup flow (1) and spiral flow (2) methods which yield a measure of the combined flow, heat transfer and curing behaviour of the material within a relatively complex geometry. These comparative methods are suited for ranking or quality control type activities. Their value in determining fundamental properties data is rather limited. The remainder of this paper will be concerned with those methods that purport to yield quantitative values of fundamental rheological properties.

2 FUNDAMENTAL PROBLEMS ENCOUNTERED IN THE MEASUREMENT OF THE RHEOLOGICAL PROPERTIES OF FIBRE FILLED MATERIALS

The rheological characterisation of fibre filled materials is a difficult subject. The degree of difficulty typically increases with increasing fibre concentration and increasing fibre length. This difficulty primarily stems from problems associated with the physical size of the fibres relative to that of the measuring geometry that is used to characterise the material. In the rheological techniques that are well established the critical dimensions of the measuring geometry are of the order of a few millimetres at most. This is smaller than the length of the fibres in many reinforced plastics. Not surprisingly this results in experimental problems.

In all rheological techniques for fibre filled materials the sample is deformed in testing. Fibre degradation during testing is virtually inevitable as a result of the work done on the sample. Thus the measurements are of a material that is undergoing degradation and thus has an unknown fibre length distribution, even if the fibre length distribution was known prior to testing. To specify accurately the relationship between the fibre length distribution or concentration and the rheological properties of a material is therefore difficult, if not impossible to do (3,4). However, it is evident that the effect is greatest on the extensional flow behaviour of materials, rather than on the shear flow behaviour (5).

In addition to fibre degradation there is the complicating factor of the effect of fibre orientation on the rheological behaviour. The fibre orientation is difficult, if not impossible to control during rheological measurements. Thus measurements are typically of a material having an unspecified and probably non-homogeneous fibre orientation. The effect of fibre orientation on results is therefore difficult to determine.

In interpreting the raw experimental data in terms of viscosity values, as is necessary for flow modelling, there is the further complication that increasingly dubious theoretical assumptions need to be made in order to analyse that data. The analyses used in standard experimental techniques, for example capillary extrusion or rotational rheometry, are for fluids which can be described by a continuum model. These analyses do not take into account the peculiarities of particulate filled systems. In capillary rheometry, for example, the fluid is assumed to obey a Newtonian or generalised Newtonian shear viscosity versus shear rate relationship. This assumption implies that the velocity profile within the die is given by a simple analytical expression (6,7). No account is taken of the presence of fibres which would perturb this velocity profile. Furthermore, the analysis does not account for the effect of wall-fibre and fibre-fibre interactions. Analysis of the experimental data to determine wall slip can be performed (8) but again the theoretical basis for this is highly questionable.

Despite these complications valuable data on the flow behaviour of fibre filled materials can be obtained. This paper will clarify how such data can best be obtained and will highlight the potential hazards in the measurement and subsequent use of that data.

3 SHEAR FLOW, LARGE AMPLITUDE DISPLACEMENT MEASUREMENTS

3.1 GENERAL

Extrusion rheometry is the most suitable technique for determining quantitative rheological data for fibre filled materials. It can be carried out using either capillary or slit die geometries on laboratory equipment or using an extruder or a moulding machine. Rotational rheometry can also be used to determine quantitative data though, in comparison with extrusion rheometry, it is limited in what can be obtained for fibre filled materials. These various methods are discussed below.

3.2. EXTRUSION RHEOMETRY

3.2.1 Capillary die extrusion rheometry

Capillary extrusion rheometry is a well established technique (5,6,7). The basic principle of the method is that material is extruded through a capillary die at a given flow rate and temperature, Figure 1. The shear viscosity can then be determined from the measurement of the extrusion pressure. Test sample sizes range from approximately 50 - 100 cm³.

The standard ISO 11443: 1993 Plastics - Determination of the fluidity of plastics using capillary and slit die rheometers (9), specifies the procedure to be used for determining the shear viscosity of plastics melts. This standard does not exclude the measurement of fibre filled materials in its specification. In testing of fibre filled materials it is necessary to take into account the following aspects.

For fibre filled materials it is essential to make a correction for the pressure loss due to the flow of material into and out of the die (5). The contribution due to the flow out of the exit is small. Thus the correction, commonly referred to as the Bagley correction (10), is often considered to be an entrance pressure drop correction. It is carried out by performing measurements using dies of more than one length, but of the same diameter. A typical Bagley plot for an unsaturated polyester dough moulding compound (DMC) which contains

glass fibres of 6 mm in length and 12% by volume is presented in Figure 2. It shows that for a die of length 60 mm and radius 2 mm ($L/R = 30$) the correction, equal to the intercept value on the y-axis, can be as great as 60% of the total pressure drop. Thus shear viscosity values would be overestimated by 150% if this correction were not made. This error would be greater if a shorter die had been used. The entrance pressure drop can be interpreted, albeit rather crudely, as an extensional viscosity value. This will be considered in more detail in Section 5.

The extrusion pressure traces obtained for fibre filled materials show large variations in pressure, Figure 3. It has been shown (5) that these variations originate in the entry and exit regions of the die and are due to the fibres. By using an entry cone geometry to the die, Figure 4, the variations in the pressure trace can be reduced considerably thereby improving the accuracy of the determination of shear viscosity values, Figure 5. The use of different entry cone angles had no significant effect on the measured shear viscosity values, Figure 6.

Careful consideration of the dimensions of the capillary or slit die needs to be given to ensure that the measured viscosity values are independent of the test geometry used. For example, for an unsaturated polyester DMC containing fibres of 6 mm length, it was observed that higher shear viscosity values were measured when using capillary dies of 3 mm diameter compared with values that were obtained when using either 4 mm or 6 mm diameter dies (5), Figure 7. This observation is considered to be due to the effect of interactions between the fibres and the wall of the capillary die. Considering the relative size of the fibres it is not surprising that geometry dependent viscosity results are obtained.

To investigate the effect of the glass fibre content on the rheological behaviour of unsaturated polyester DMC three materials that differed only in their fibre reinforcement were tested (5), Figure 8. The volume percentage of fibres in each material was the same. The results show that the addition of the fibres to the matrix increased the viscosity by a factor of approximately x3, but that there was no significant difference in values between the three materials, Figure 9. However, the effect on the entrance pressure drop values was an order of magnitude greater, Figure 10. Entrance pressure drop values were a factor of x20-40 greater than that for the unfilled material and there were measurable differences between the three grades. Thus for the purpose of identifying differences in the behaviour of fibre filled materials it is clearly preferable to examine entrance pressure drop values rather than shear viscosity values.

The effect of capillary rheometry measurements on the fibre length distribution of a material can be seen in Figure 11. Increasing the extrusion rate caused a significant reduction in the

mean fibre length $\bar{L}$. Thus characterisation of what is nominally a material at different extrusion rates is in reality a complex characterisation of a material that changes with both flow rate and position within the test geometry.

Extrusion rheometry has its limitations in that curing systems cannot be characterised easily due to the problem of curing of the material within the rheometer. Materials which exhibit inhibition times, for example unsaturated polyester DMCs (11), can be characterised provided that the measurements are made within the period during which cure is inhibited. Measurements of a slow curing DMC have been made at temperatures up to 130 °C (12). However, the material is prone to styrene loss which must be taken into account when interpreting the results.

Laboratory based rheometers can be used to obtain viscosity values at shear rates up to approximately 500 s⁻¹ using a 4 mm diameter die. Numerical modelling of injection mould filling requires data at considerably higher shear rates (5). In order to obtain higher shear rate data injection moulding machines have been fitted with capillary die geometries by Crowson et al (13) to characterise short glass fibre reinforced (SGFR) thermoplastics, by Gibson and McClelland (14) for long glass fibre reinforced thermoplastics and by Gibson and Williamson (15,16) for DMCs. In these cases the machine was used as a high flow rate extruder thus giving higher shear rates than could be obtained using laboratory rheometers. In addition Isayev et al (17) have used a laboratory scale screw extruder for high rate measurements of SGFR-thermoplastics. Although higher shear rate data can be obtained there is a penalty to pay. As extrusion pressures increase then the effect of viscous dissipation on results will increase. This is discussed more fully in section 3.3.

3.2.2 Slit die extrusion rheometry

The difference between slit die extrusion rheometry and capillary die extrusion rheometry is that in the former a rectangular slit geometry is used in place of a capillary. The slit die can be either instrumented with pressure transducers along its length or it can be used in the same way as a capillary die with a pressure transducer upstream of the die entry. Both of these slit die methods are covered by the standard ISO 11443 (9).

Most of the comments made in the above section on capillary die extrusion rheometry apply also to slit die extrusion rheometry. The use of a slit die with a pressure transducer upstream of the die entry is of little advantage compared with using capillary dies. The use of an instrumented slit die with pressure transducers mounted along its length has certain

advantage and disadvantages over capillary dies for measuring fibre filled materials. The primary advantage in using instrumented slit dies is that it avoids the need to make entrance pressure drop corrections. Thus measurements can be made more rapidly. However, the maximum shear rate that can be obtained using a slit die is lower than that obtained using a capillary die due to geometry considerations. The width to thickness ratio of the slit die ideally needs to be not less than 10 to satisfy the assumptions made in the analysis of the experimental data. This results in a lower maximum shear rate when using a slit die (assuming the capillary die diameter and the slit die thickness are the same).

For an unsaturated polyester DMC the slit die method could be used to obtain shear viscosities at lower values of shear rate than could be obtained by capillary die rheometry. In using capillary dies at low flow rates the DMC would "log-jam" thereby preventing the measurement of shear viscosity (5). A comparison of results shows good agreement between the use of slit and capillary dies, Figure 12. Thus the use of both methods has extended the shear rate range over which the material has been characterised. This has benefits in that it will potentially improve the procedure of fitting a viscosity model to the data.

The use of slit dies for determining normal stress differences, following Han (18) and Boger (19), is not recommended for fibre filled materials due to the large variations in the measured exit pressures resulting from the presence of the fibres (5).

3.3 IN-LINE RHEOMETRY

As discussed in section 3.2.1 several researchers have developed techniques to measure the flow behaviour of polymers using either injection moulding machines or extruders fitted with capillary rheometer geometries. However comparatively little effort has been made to develop on-line or in-line rheological techniques (20,21,22). On-line methods are defined as those that take a sample out of the process line and then test it. In-line methods are defined as those that test the material without taking a sample out of the process line. Hence in-line measurements are preferable in that they characterise material that is used in the process rather than material that has been extracted from it. With on-line methods there is the possibility that the material is degraded or modified in extracting it from the process and thus subsequent measurement of the sample will give misleading information on the state of the material in the process. Furthermore any possibility of degradation of the material makes it inadvisable to return that material to the process, thereby increasing wastage rates.

An in-line technique has been developed by the National Physical Laboratory (22). The technique can be used to characterise the flow behaviour of fibre filled plastics during the injection moulding process. It is based on the use of an extended nozzle on an injection moulding machine, Figures 13 and 14. The nozzle, which was commercially available, was instrumented with two pressure transducers positioned along its length. The shear viscosity of the material flowing in the nozzle can be determined from measurement of the pressure drop along the nozzle (Figures 15 and 16), the material temperature, the flow rate and the nozzle geometry. The technique has been demonstrated while moulding a centre-gated tray using an unsaturated polyester DMC, Figure 17. The test sample size was approximately 250 cm³.

A principle benefit of this method is that shear viscosity data can be obtained at higher shear rates than could otherwise be obtained using a capillary extrusion rheometer. This is due to the higher flow rates that can be delivered by the injection moulding machine. Furthermore, the modification to the injection moulding machine is relatively simple and inexpensive. It has the added advantages that the technique does not interfere with the production process, that it characterises the material immediately prior to its entry into the injection mould cavity and that it can provide real-time data for process monitoring and control. However, the technique does have its disadvantages and these are discussed below.

The results obtained using the in-line method have been compared with shear viscosity values obtained using a laboratory based capillary extrusion rheometer, Figure 18. Reasonable agreement of the results of the two methods has been obtained. The discrepancies observed can be accounted for due to the effects of viscous heating, the pressure-dependence of viscosity and the contribution due to the converging flow in the nozzle.

The nozzle channel is tapered with an angle of 1° over the majority of its length, but of 3° over the final few millimetres. The effect of this contraction flow is estimated, using the theories due to Gibson (23), Binding (24) and Cogswell (25), to be in the region of 10-20% of the total pressure drop. Thus it is estimated that the shear viscosities determined using this technique are 10-20% higher than actual due to the use of the converging geometry.

A major problem in measuring the flow behaviour of materials at higher flow rates is that the effect of viscous dissipation becomes increasingly significant. For these in-line DMC measurements an average temperature rise of approximately 30 °C was measured at the highest flow rate, Figure 19. For this material a 1 °C increase in temperature equates to a

3% decrease in the viscosity. Thus it can be seen that shear heating will have a significant effect on the measured viscosity of the material.

To evaluate the effects of viscous dissipation, the pressure dependence of viscosity and the converging flow on the measurement of viscosity requires the use of numerical modelling techniques. These, including the generation of pressure dependent viscosity data, are topics of current work of the National Physical Laboratory.

3.4 ROTATIONAL RHEOMETRY - STEADY SHEAR

Rotational rheometry is a well established technique for unfilled materials (6,7, 26). The principle of the technique is to subject a sample that is held between two plates to a shearing stress. The resultant rotational displacement of one of the plates relative to the other is then measured (alternatively one plate is rotated relative to the other and the required torque is measured). Viscosity functions can then be determined, given the geometry dimensions. Various geometries can be used.

The technique of rotational rheometry is generally not recommended for characterisation of fibre filled materials. The sample size is small, typically of the order of 2-4 cm³ with a maximum sample thickness (gap between the plates) of a few millimetres. The technique is also limited to relatively low shear rates. This is particularly the case where a large gap size is required to accommodate the large particle size of the fibres in the material.

For fibre filled materials the most suitable standard geometry is that of parallel plates, where the sample is held between two circular, flat parallel plates. However, analysis of the results for this geometry is not straight forward (6,7,26). Cone and plate geometries are not recommended for fibre filled materials due to the relatively small gap at the centre of the plates (normally a truncated cone is used). Cross and Kaye (27) have examined the use of the displaced cone and plate and angular cone and plate geometries to overcome the problems that are associated with the measurement of fibre filled systems, as are illustrated for example by Mutel and Kamal (28). However, steady shear rotational rheometry testing is generally not recommended for characterising fibre filled materials.

4 SHEAR FLOW, SMALL AMPLITUDE DISPLACEMENT MEASUREMENTS

Techniques for characterising the rheological behaviour of materials using small amplitudes of displacement are well established for unfilled materials (6,7,26). They are suited for characterising the behaviour of materials which undergo changes, for example the curing of thermosetting plastics and adhesives and the degradation of thermoplastics. Parallel plate oscillatory rheometry is probably the most widely used technique and can be used for characterising fibre filled materials.

Many of the complications that were described for steady shear rotational rheometry also apply to oscillatory rotational rheometry. For measuring curing systems an additional complication is that the compliance of the machine is often too large. This results in the imposed deformation being accommodated mainly by the deformation of the machine rather than by the sample being tested (29). This effect can be corrected for, but the correction is sensitive to the value used for the compliance of the machine which needs to be determined experimentally.

Dynamic viscosity values have been related to steady shear viscosity values for unfilled polymer melts, following the Cox-Merz rule (30,31). However, evidence indicates that this rule does not in general apply for filled systems (32). Correlation of dynamic with steady shear viscosity data on a quantitative basis is not recommended for fibre filled materials.

5 EXTENSIONAL FLOW MEASUREMENTS

Extensional flows occur in injection moulding, for example in the nozzle of the injection moulding machine, in passing into the gate of the moulding and where radial flow occurs as is found when a moulding is filled from a small gate. Extensional flows also occur in compression moulding where the flow is of a radial spreading nature. Thus extensional viscosity data are important for accurate flow modelling of polymer processes.

In comparison with the measurement of shear viscosity, the measurement of the extensional viscosity of materials is less well developed. For a polymeric solution (polyisobutylene in decalin) differences of up to two decades in extensional viscosity values were obtained depending on the method used (33). One would anticipate that the extensional flow behaviour of polymeric solutions is considerably simpler than that of fibre filled polymer melts. Thus it would be expected that the scatter of results obtained for fibre filled materials would also be considerable. To make matters worse many of the extensional viscosity

techniques that were used in reference 33 are not suited for testing of fibre filled materials. The technique that is perhaps most suitable is the determination of extensional viscosity values from the measurement of the entrance pressure drop in capillary extrusion rheometry. This method is discussed latter.

The parallel plate squeeze flow technique (34) is also potentially suitable for measuring fibre filled materials. However, the technique has also been used to characterise the shear flow behaviour of sheet moulding compounds (35). The validity of the analysis for materials having a high Trouton ratio, that is the ratio of extensional viscosity to shear viscosity (36), is questioned. The squeeze flow technique with lubricated plates has been used by Lee et al (37) to measure the biaxial extensional viscosity of SMCs. Lubrication was achieved by applying silicon grease to Teflon plates so that slip of the SMC occurred at the sample/plate interface thereby yielding a known velocity field. Values for shear and biaxial extensional viscosities were presented. Results indicated that the addition of fibres increased the shear viscosity by a factor of $\times 1.6 - 2.3$ and the biaxial extensional viscosity by $\times 60 - 200$, depending on fibre length. An increase in fibre length resulted in an increase in both shear viscosity and biaxial extensional viscosity. However, a small maximum value of 0.12 s^{-1} for the average strain rate was reported for this technique (37). This value is considerably lower than the strain rates that occur in processing polymers.

Converging flow is a form of extensional flow. It would be expected, therefore, that extensional viscosity values might be obtained from measurements of the pressure drop and flow rate in converging flows. This approach is appealing as experimental data is either readily available or is easily obtained from standard capillary rheometry testing provided that either a Bagley correction is made, Figure 2, or a zero length die is used to obtain entrance pressure drop values. Thus both shear and extensional viscosity values can, in principle, be obtained from the same capillary rheometry test. This technique also has advantages over the squeeze flow method described above, in particular higher strain rates can be obtained.

Numerous models for determining extensional viscosity from entrance pressure drop measurements have been presented in the literature (15, 16, 38-53) and have been reviewed in reference 5. A detailed comparison of the predictions of the models due to Gibson (23), Binding (4) and Cogswell (25) has been presented by Rides (5). For the materials studied in that work the model due to Gibson (23) is preferred. The use of converging dies to obtain extensional viscosity data has also been reviewed by Gibson (54).

The ratio of extensional viscosity to shear viscosity values obtained using the Gibson model for an unsaturated polyester DMC was in the region of 30-40, Figure 20. In comparison the

use of the Cogswell and Binding models predicted values in the ranges 70-80 and 35-40 respectively, Figure 20. This figure shows the level of disagreement between these models. However, this is currently the best data that can be obtained for fibre filled materials.

Evaluation of the Binding model (24) was undertaken by the University of Wales, Aberystwyth and the National Physical Laboratory. This work has been reported by Davies et al (55) and by Rides et al (56). This evaluation has raised questions over the validity of the Binding model for the determination of extensional viscosity values for materials which exhibit certain types of behaviour. A particular problem encountered in using the Binding model was the high sensitivity of the calculated extensional viscosity parameters to the selection of the flow rate range of pressure drop values.

To provide an alternative contraction flow analysis, a numerical contraction flow model was developed and was used to establish an algorithm (57). This algorithm can be used to determine extensional viscosity parameters, given entrance pressure drop versus flow rate data and shear viscosity data for the material. The algorithm has been applied to results for an unsaturated polyester slurry and has been shown to give a good fit to experimental entrance pressure drop data to within 5% (56). Trouton ratios of the order of 3 were obtained at low flow rates - the value expected for a Newtonian fluid. However, the determination of extensional viscosity values of a long glass fibre filled unsaturated polyester DMC proved less satisfactory. In checking the calculations of extensional viscosity parameters by recalculating the entrance pressure drop data, poor agreement was obtained with experimental results. This was due to the high sensitivity of the measured pressure drop data to flow rate at low flow rates. To improve the capability of the model to yield extensional viscosity values for these fibre filled materials an improved form of the constitutive equation has been proposed (56).

Theories to predict the extensional viscosity of fully aligned fibre filled materials have been developed by Batchelor (58), Goddard (59) and more recently by Toll (60). The suitability of the equations due to Batchelor (58) and Goddard (59) have been investigated by Rides (5). For the highly concentrated fibre suspensions typical of unsaturated polyester DMCs the predictions of these models do not compare favourably with values determined using the entrance pressure drop techniques proposed by Gibson (23), Binding (24) and Cogswell (25).

6 DISCUSSION

There are major practical and theoretical problems associated with the rheological measurement of fibre filled materials. The practical problems restrict the range of conditions, primarily that of shear rate, that can be obtained in testing. These can be overcome to some degree by the use of alternative methods, for example the use of the in-line technique to obtain higher shear rate data. However, this has the consequence that the errors in the data due to viscous heating and pressure dependence of viscosity increase. The error due to the converging flow can be avoided by using a non-converging channel although this is not desirable from a practical point of view. Further problems in characterising fibre filled materials are related to fibre degradation and orientation effects.

The theoretical problem of how to interpret experimental data in terms of rheological behaviour remains. Current analysis of experimental data obtained using the established rheological techniques is based on the use of continuum assumptions. These are obviously approximate assumptions but they permit simple representation of the bulk flow behaviour of fibre filled materials. The application of such data to the modelling of flow in different situations, eg different geometries, must therefore be carried out with caution. This is particularly the case where testing has indicated that the viscosity values are geometry dependent. This implies that the original interpretation of the raw data in terms of viscosity was inadequate.

Various methods exist for characterising the bulk flow, large amplitude displacement behaviour of fibre filled materials. For the above reasons the quantitative accuracy of these techniques is questioned. Nevertheless, valuable information can be obtained using capillary extrusion rheometry for characterising and predicting the flow behaviour of fibre filled materials. Steady shear rotational techniques are not favoured for measurement of fibre filled material due, in part, to the small sample size. However, oscillatory techniques employing small displacement amplitudes can be used to characterise curing systems which cannot be adequately characterised otherwise.

An in-line technique has been demonstrated and can provide viscosity data at higher shear rates than is obtainable using laboratory based extrusion rheometers. Measurement of viscosity of fibre filled materials at high shear rates is a particular problem as the critical dimensions of the test geometry need to be larger than for unfilled materials. Such high rate data are desirable for flow modelling as it reduces the need to extrapolate data to the high rates that occur in processing.

Extensional flows are an important component of many polymer processing methods. It has been shown that for fibre filled materials the extensional flow component can be a significant part of the overall filling behaviour. This is indicated by the large entrance pressure drop values obtained in the Bagley plot, Figure 2. Methods for determining the extensional viscosity using converging flow models has been reviewed elsewhere (5). The errors associated with these models are considered to be potentially significant. However, there is currently no suitable alternative method. However, errors made in determining extensional viscosity values from pressure drop data will be, to some extent, corrected for in applying the values to predict flow behaviour in similar geometries.

7 CONCLUSIONS

In carrying out rheological measurements of fibre filled materials it is important to consider the following aspects.

- * Contraction flow analysis of entrance pressure drop data obtained from capillary rheometry is currently the most suitable method for determining extensional viscosity data for fibre filled materials.
- * Due to the many complications in testing of fibre filled materials it is preferable to use a method that mimics the process for which the data is required.
- * Fibre-fibre and fibre-wall interactions, fibre breakage and fibre orientation may affect results.
- * Results can show a test geometry dependence, particularly when the fibre size is large relative to the critical dimensions of the test geometry.
- * In capillary extrusion rheometry testing of fibre filled materials, more accurate viscosity data can be obtained by using a conical entry region to the die.
- * In carrying out capillary extrusion rheometry it is essential to make a Bagley correction to account for the entrance pressure drop.
- * Differences in the rheological behaviour of fibre filled materials are more likely to be identified through the measurement of entrance pressure drop values rather than shear viscosity values.

- * Capillary and slit die extrusion rheometry can provide complementary data to expand the range of shear rates beyond that which either technique could obtain individually. Slit dies can be used to obtain low shear rate data for which log-jamming is a problem in using capillary dies.
- * High shear rate data can be obtained using in-line techniques. However, errors due to viscous heating become increasingly significant.
- * Reasonable agreement of shear viscosity values was obtained between capillary extrusion and in-line testing.

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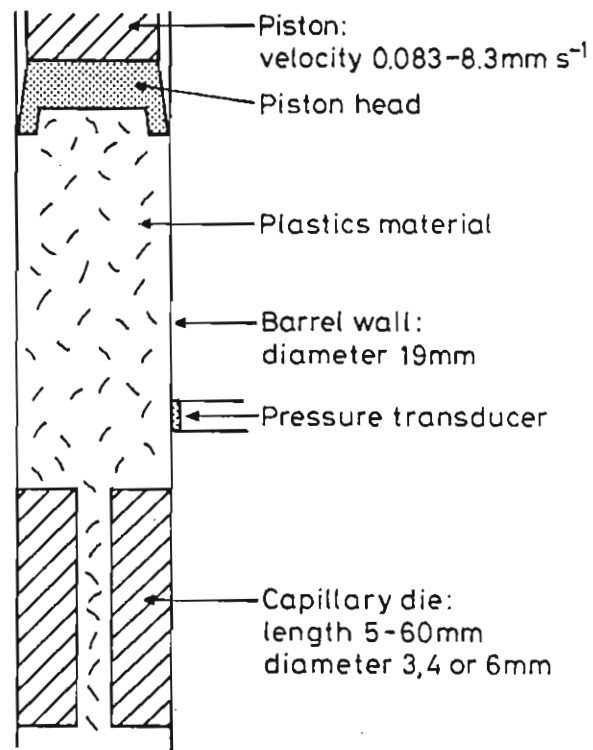


Figure 1 Capillary die extrusion rheometer.

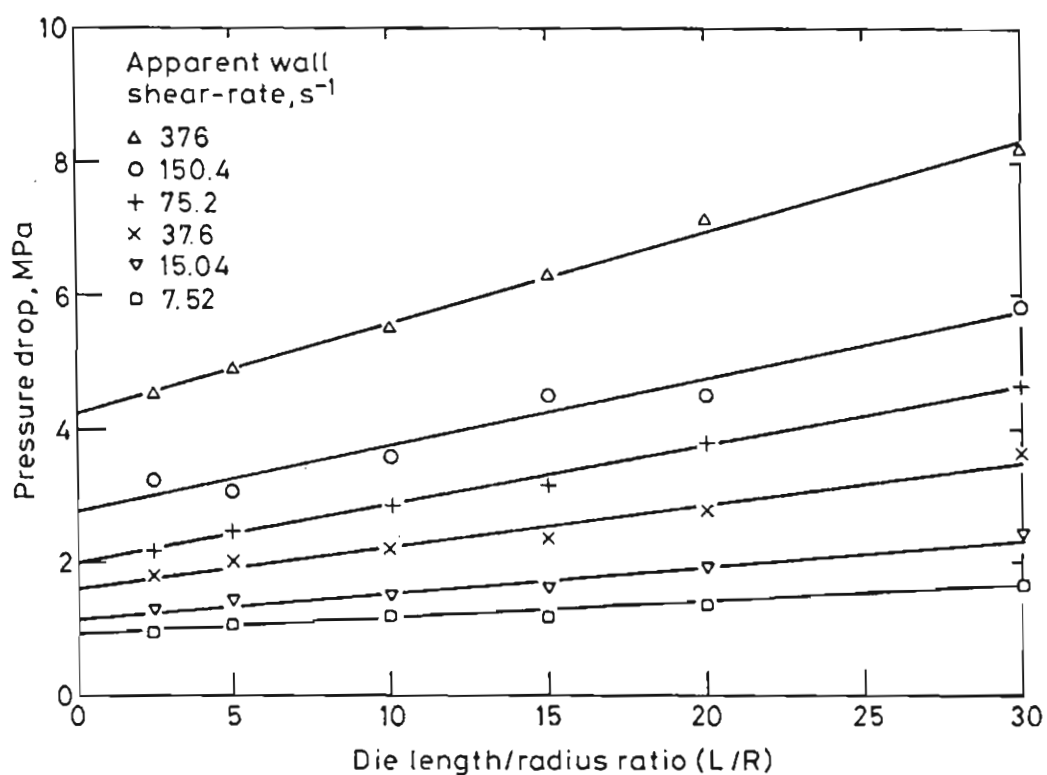


Figure 2 Bagley plot for an unsaturated polyester dough moulding compound (DMC) exhibiting high entrance pressure drop values.

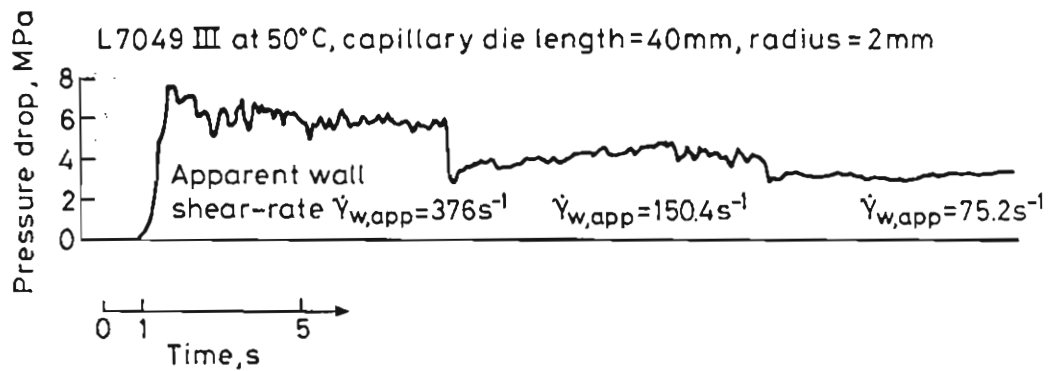


Figure 3 Capillary die extrusion pressure trace for an unsaturated polyester DMC.

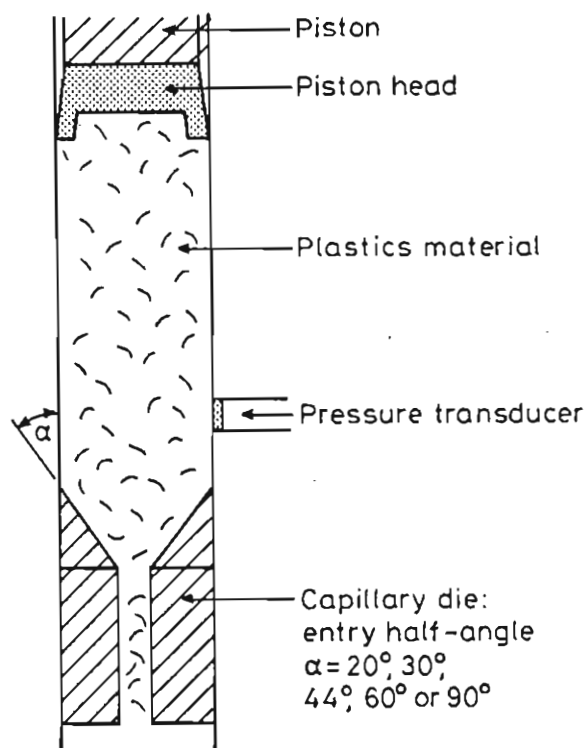


Figure 4 Capillary die extrusion rheometer with a conical entry geometry.

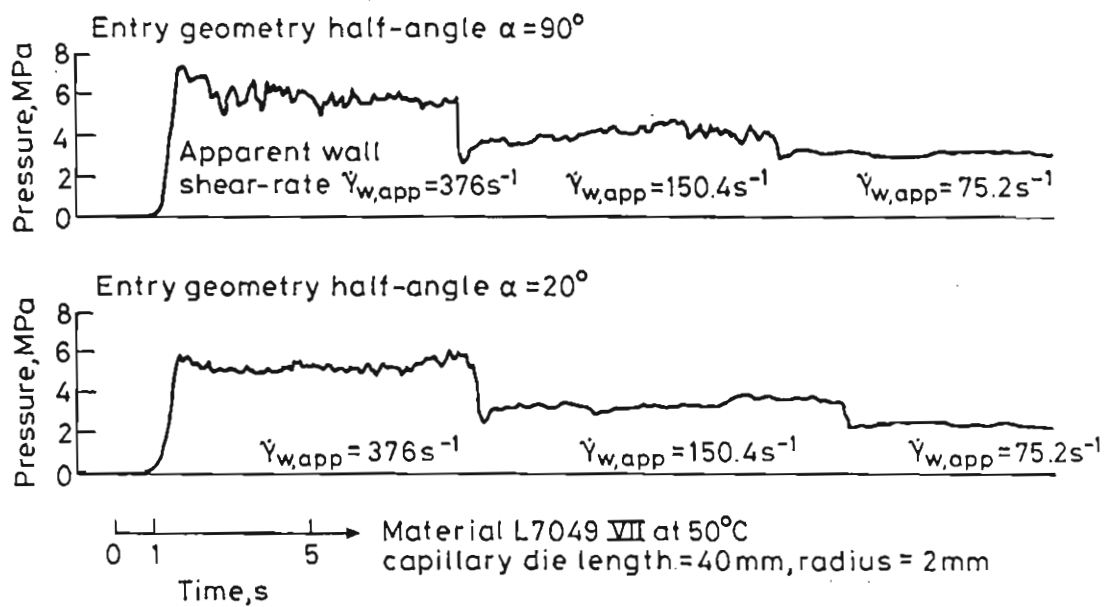


Figure 5 Effect of capillary entry geometry on extrusion pressure for an unsaturated polyester DMC.

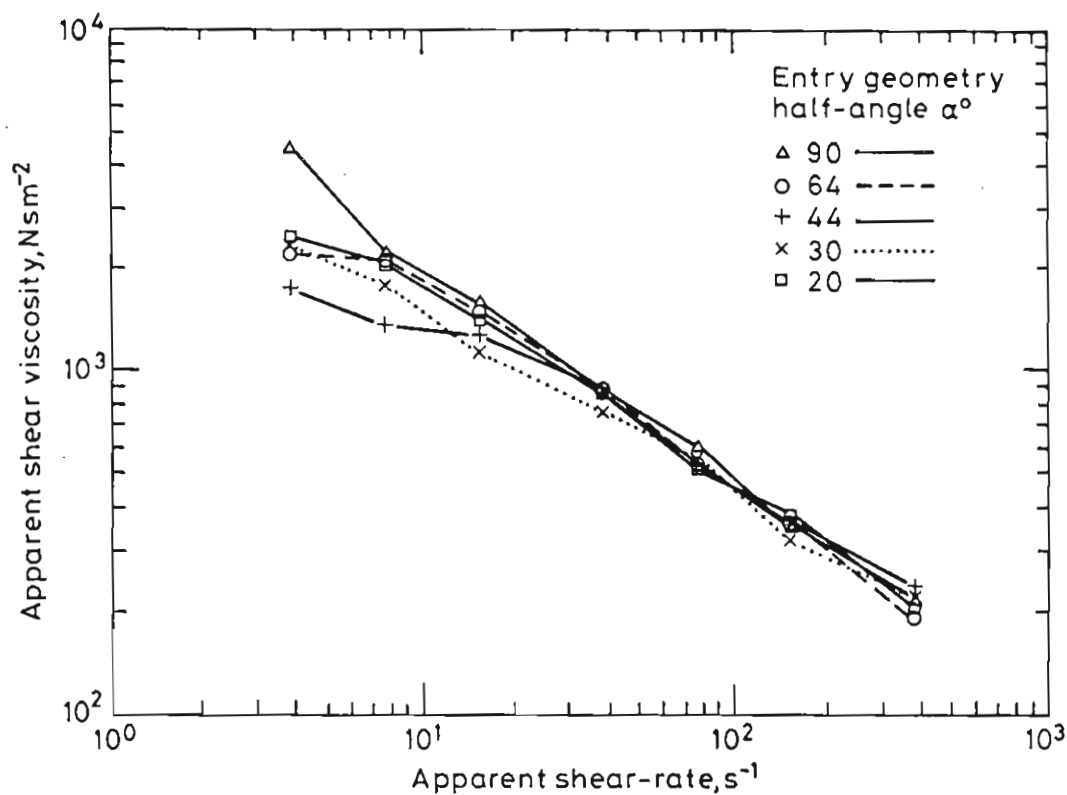


Figure 6 Effect of capillary entry geometry on the measurement of shear viscosity of an unsaturated polyester DMC.

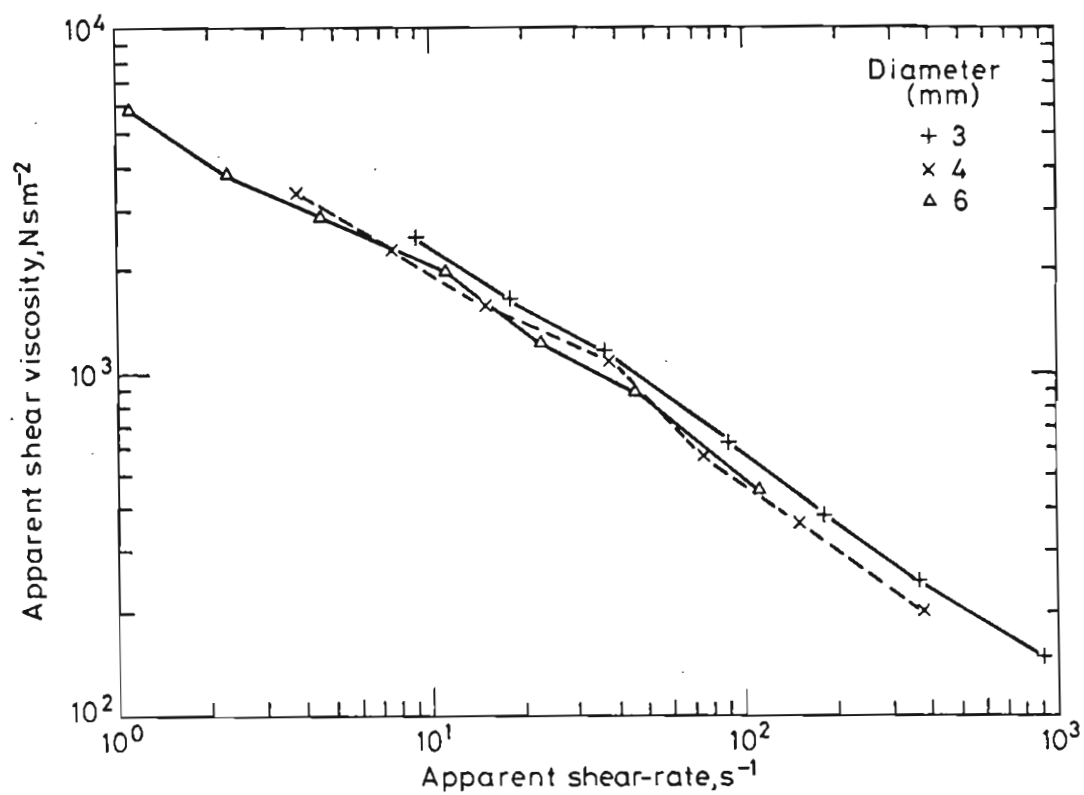
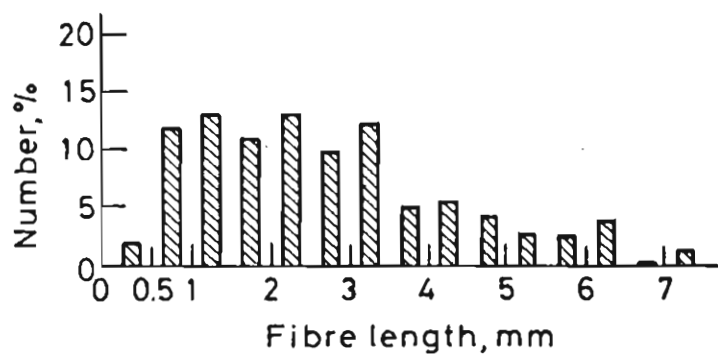


Figure 7 Effect of capillary die diameter on the measurement of shear viscosity of an unsaturated polyester DMC.

PPG 6mm glass-fibres



Sample size

$n = 228$

Number mean length

$\bar{L} = 2.78 \text{ mm}$

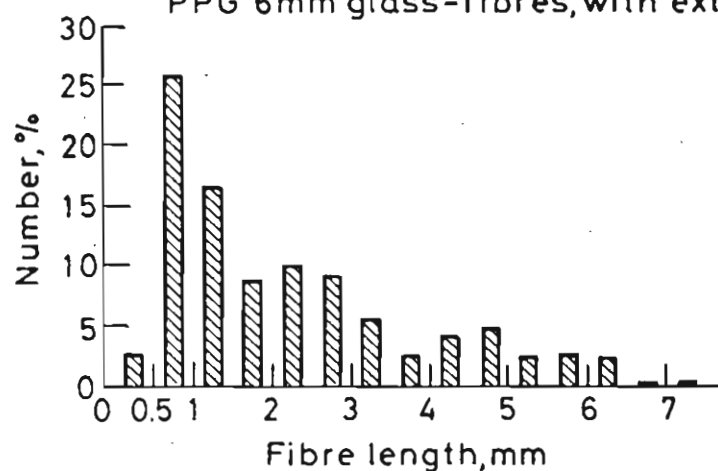
Volume mean length

$\bar{V} = 3.82 \text{ mm}$

Number % < 1mm

$N = 13.9\%$

PPG 6mm glass-fibres, with extra mixing



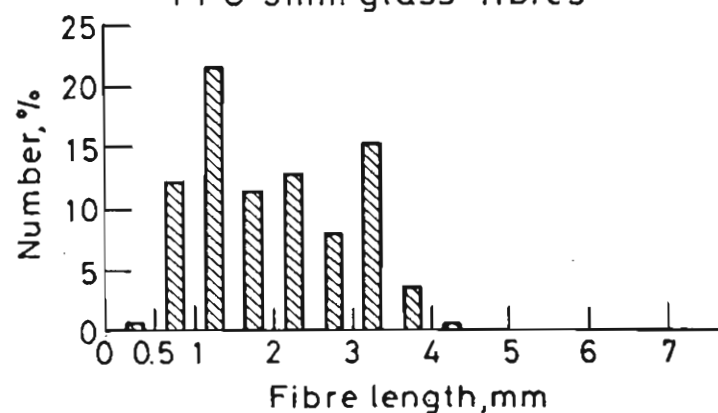
$n = 252$

$\bar{L} = 2.27 \text{ mm}$

$\bar{V} = 3.53 \text{ mm}$

$N = 28.5\%$

PPG 3mm glass-fibres



$n = 247$

$\bar{L} = 2.25 \text{ mm}$

$\bar{V} = 2.82 \text{ mm}$

$N = 12.9\%$

Figure 8

Length distributions of glass fibres extracted from three grades of unsaturated polyester DMCs.

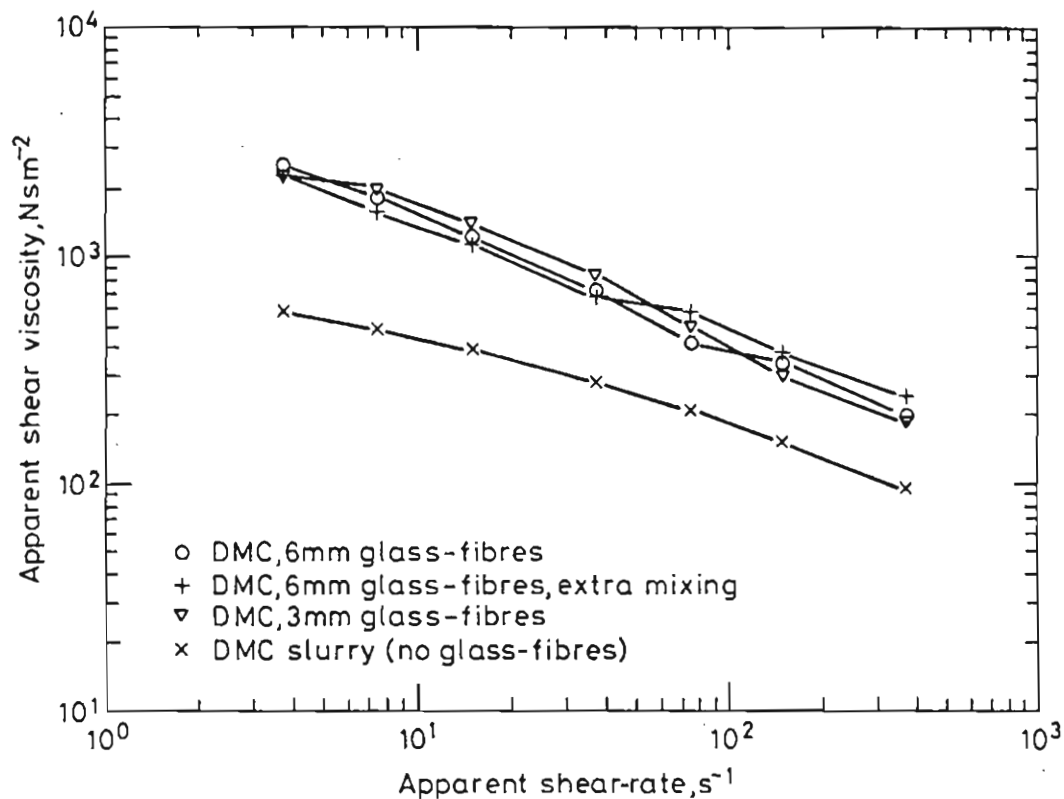


Figure 9 Effect of differences in the glass fibre content on the shear viscosity of an unsaturated polyester DMC.

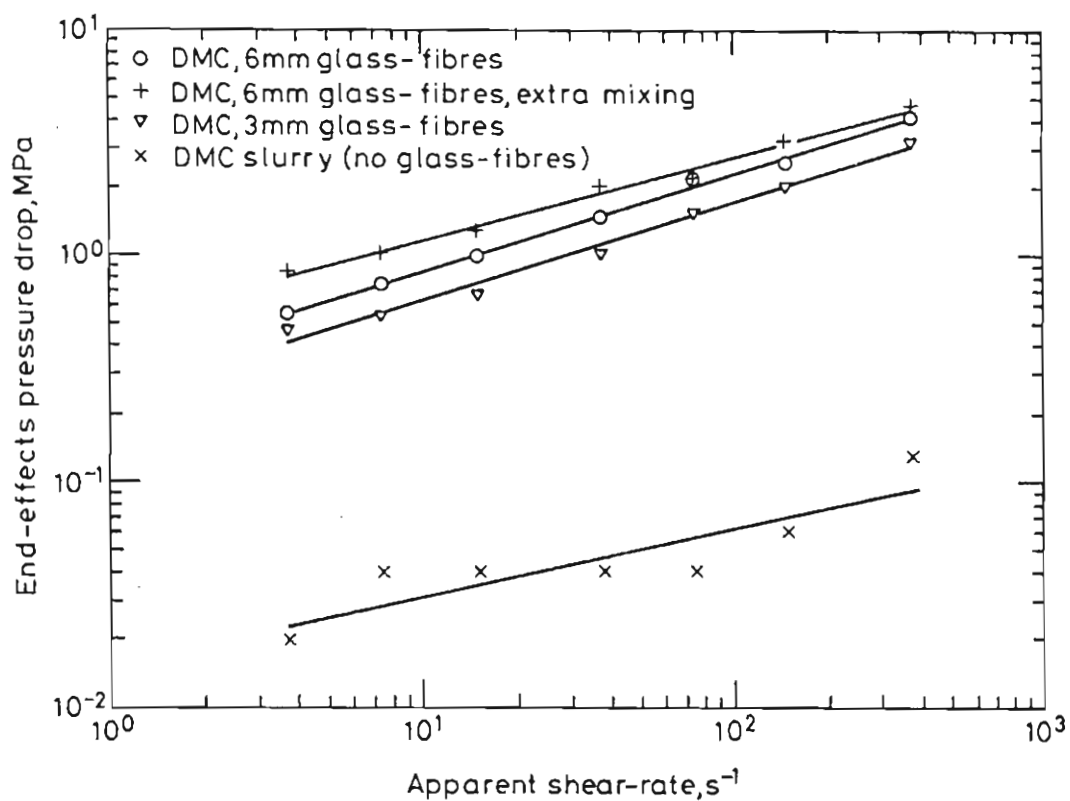


Figure 10 Effect of differences in the glass fibre content on the entrance pressure drop of an unsaturated polyester DMC.

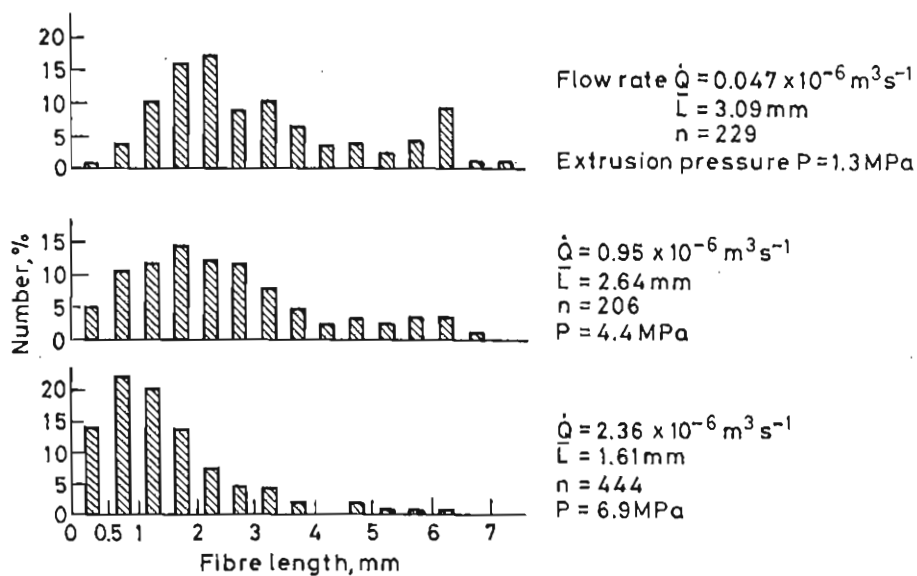


Figure 11 Length distributions of glass fibres extracted from capillary die DMC extrudates obtained at various flow rates, using a die of length 30 mm and radius 2 mm ($\bar{L}$ = number mean fibre length; n = sample size).

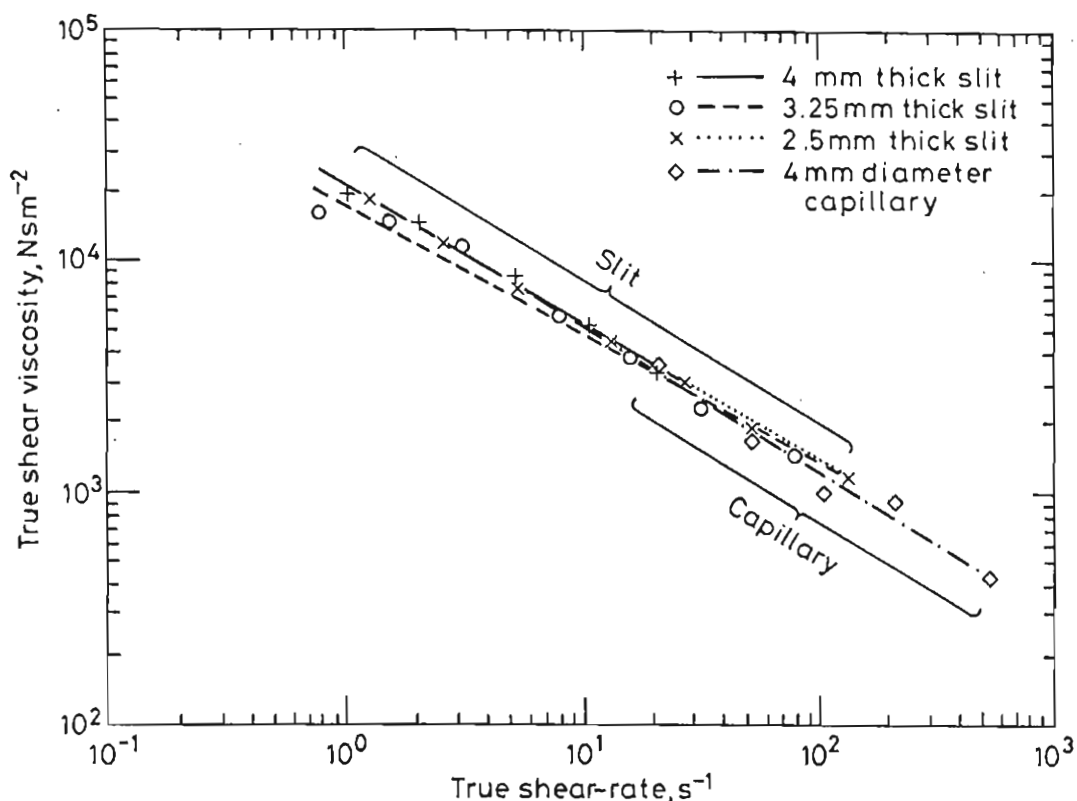


Figure 12 Comparison of slit die and capillary die shear viscosity measurements of an unsaturated polyester DMC.

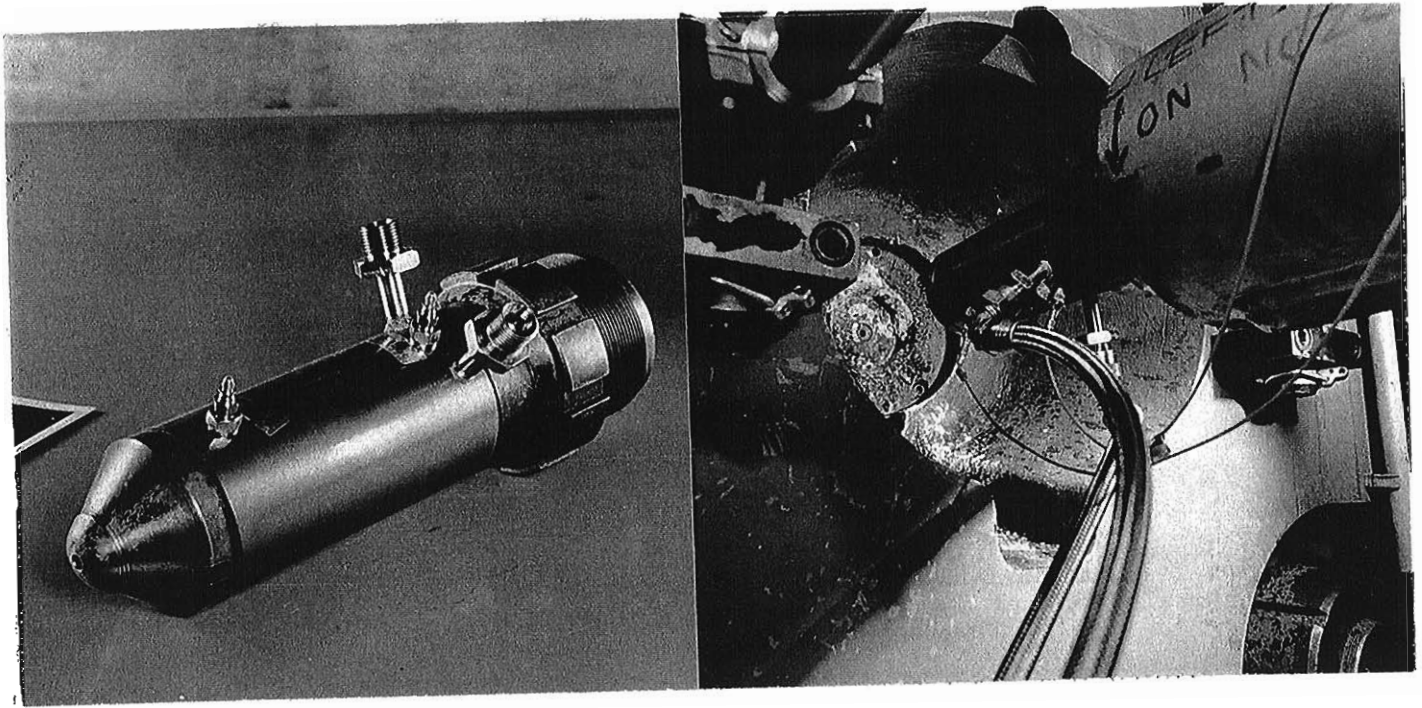


Figure 13 Example of a nozzle used for in-line measurement of viscosity.

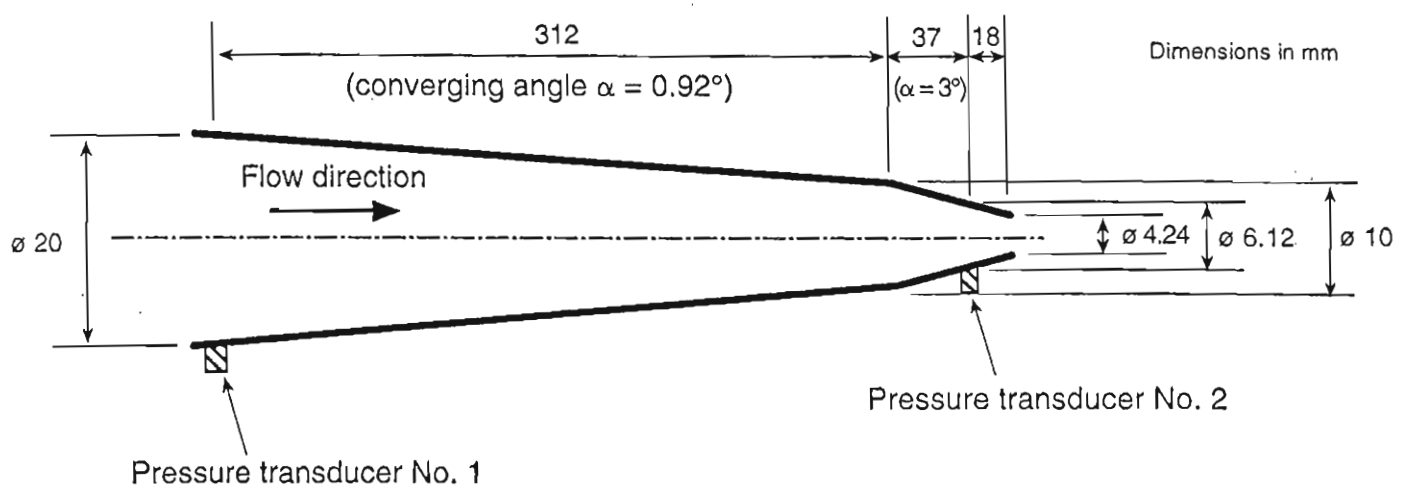


Figure 14 Instrumented injection moulding machine nozzle geometry.

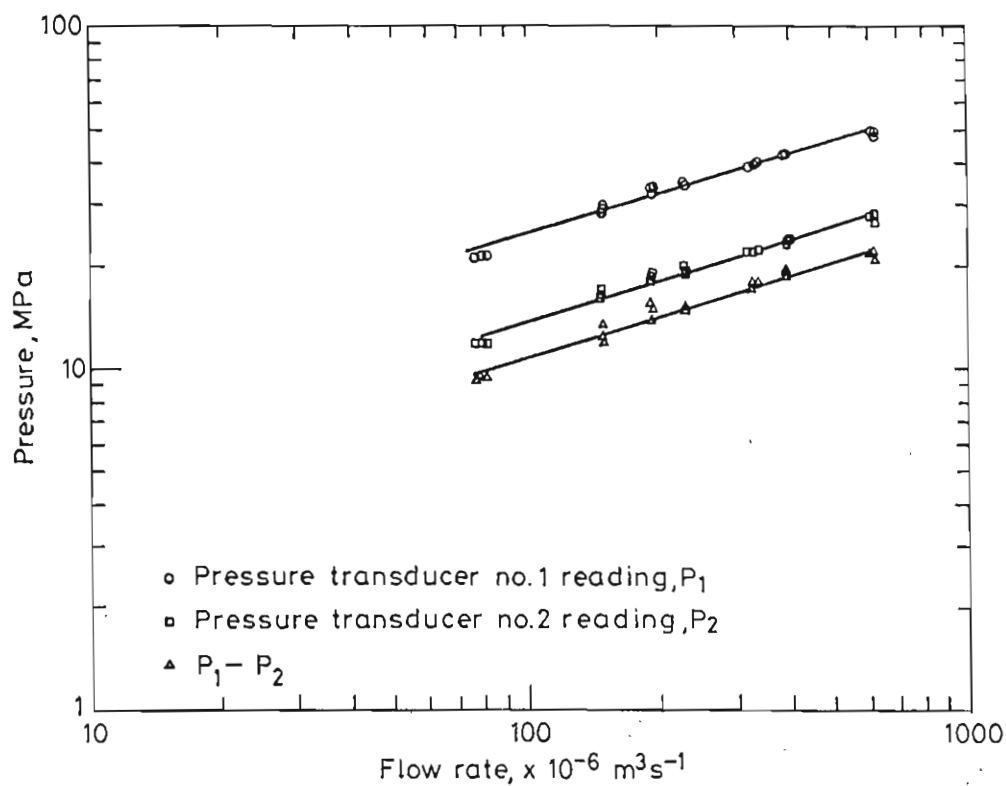


Figure 15 Pressures measured in the nozzle while extruding an unsaturated polyester DMC to air.

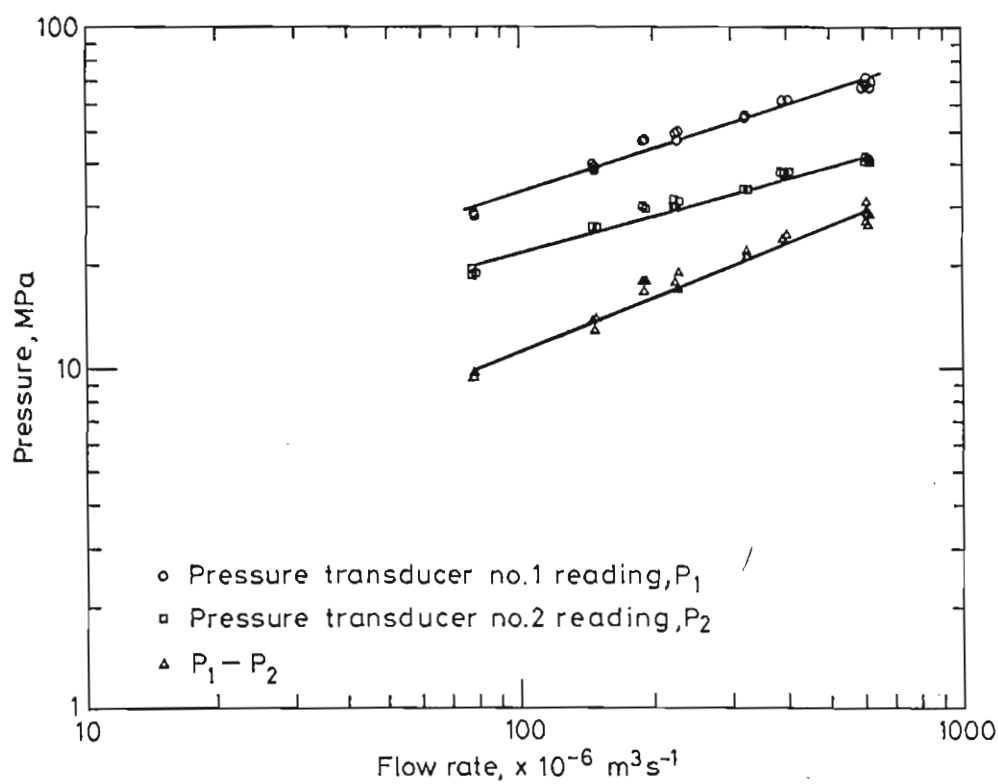


Figure 16 Pressures measured in the nozzle during injection moulding of a tray using an unsaturated polyester DMC.

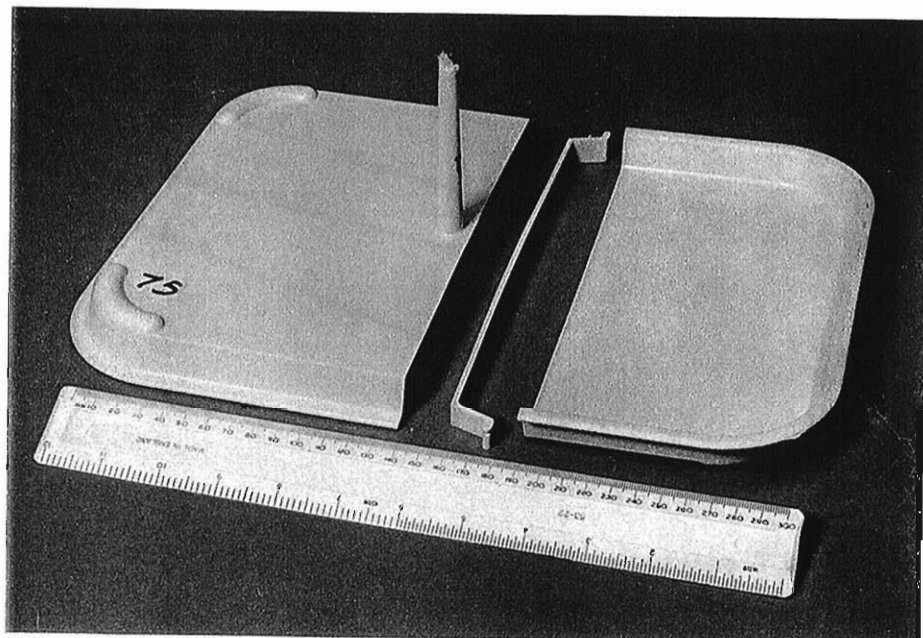
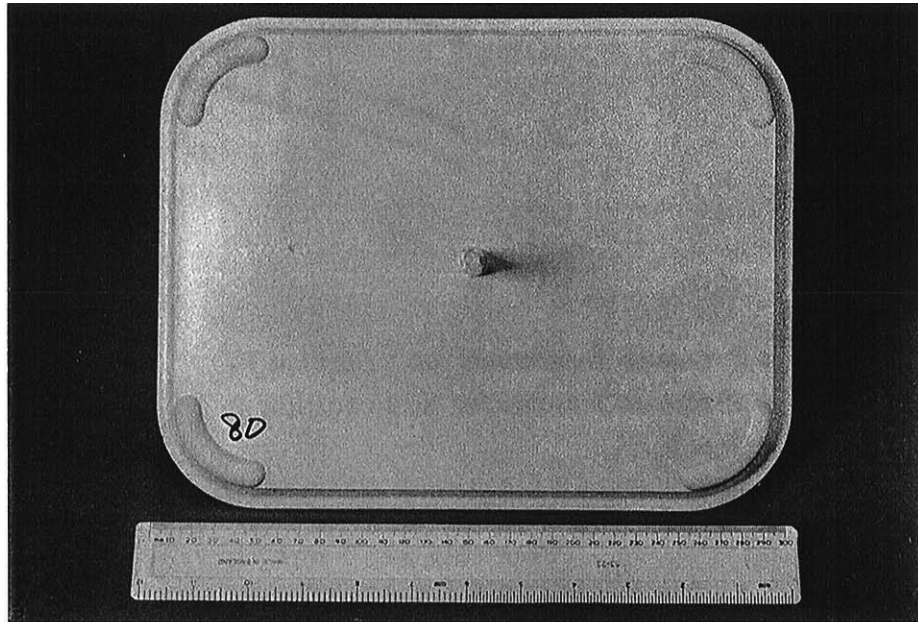


Figure 17 Centre-sprue gated injection moulded tray produced during the in-line viscosity measurement trials.

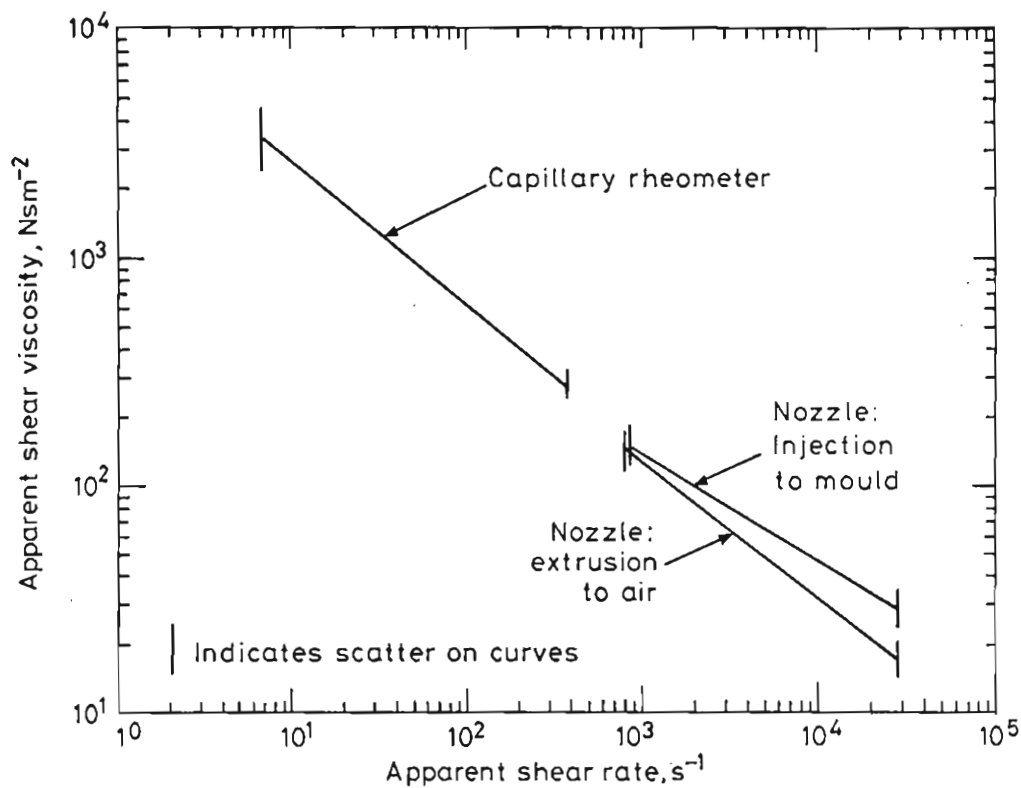


Figure 18 Comparison of shear viscosities measured using the nozzle with capillary rheometer measurements for an unsaturated polyester DMC.

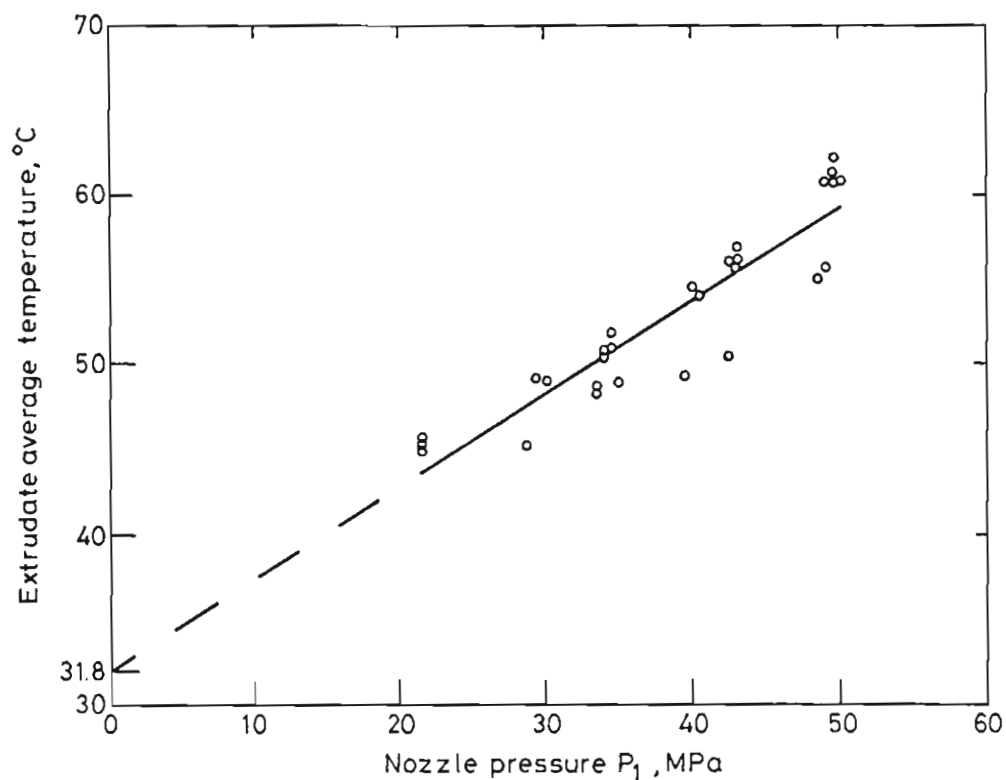
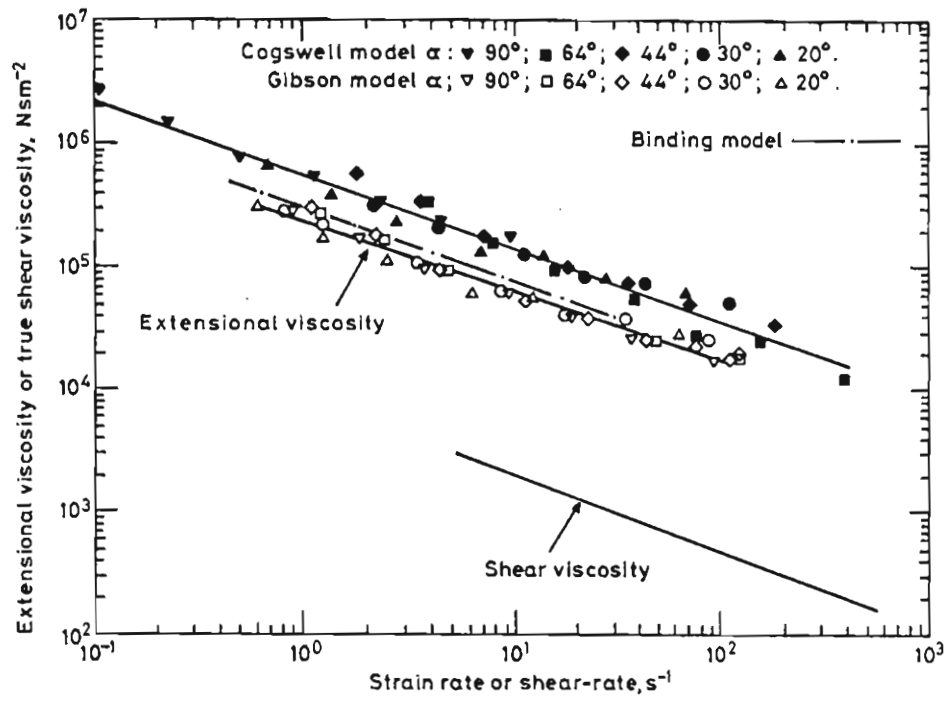


Figure 19 Relationship between the pressure drop P_1 measured near the upstream end of the nozzle and the average temperature of the extrudate.



Notes on best straight line fits to data: Cogswell $\lambda'_0 = 0.57 \times 10^6 \text{ Nsm}^{-2}$, $m = 0.4$;
 Binding $\lambda'_0 = 0.30 \times 10^6 \text{ Nsm}^{-2}$, $m = 0.40$; Gibson $\lambda'_0 = 0.24 \times 10^6 \text{ Nsm}^{-2}$, $m = 0.43$;
 Shear viscosity $\eta'_0 = 8300 \text{ Nsm}^{-2}$, $n = 0.37$

Figure 20 Comparison of predicted extensional viscosity values for an unsaturated polyester DMC using the Gibson, Binding and Cogswell converging flow models.