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ABSTRACT

This report addresses the effect that the choice of die geometry has on shear viscosity and entrance pressure drop values determined by capillary extrusion rheometry. A number of thermoplastics have been tested including a fibre-filled material and conclusions are presented as to the acceptability of certain specified die geometries for shear viscosity and entrance pressure drop measurement. These results have been used as supporting evidence to enable recommendations to be made for the development of the capillary extrusion rheometry standard ISO 11443.

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on measurements related to the processability of materials

Approved on behalf of Managing Director, NPL
by Dr C Lea, Director, NPL Materials Centre

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1. INTRODUCTION

Capillary extrusion rheometers are widely used to measure shear viscosity for use in quality control or in flow simulation packages to predict, for example, the filling of a mould cavity in injection moulding or the flow through a die in extrusion. However, the capillary extrusion rheometry standard ISO 11443 [1] permits the use of a wide range of testing conditions for characterising molten plastics with the consequence that this introduces variability into the results and a lack of comparability of data. Recommendations have already been made to the relevant ISO committee [2] by the author to specify more tightly the range of testing conditions, specifically of shear rate and temperature, that should be used when testing in accordance with ISO 11443 [1] and ISO 11403-2 [3].

Of particular concern related to ISO 11443 is that for a two-die determination of shear viscosity a commonly used die length to diameter ratio of 16 is not permitted by the standard. The minimum length to diameter ratio of the long die is restricted to not less than 20. This report primarily addresses the effect that the choice of die geometry has on shear viscosity and entrance pressure drop values. The effect that other testing conditions may have on the measurement of shear viscosity and entrance pressure drop is also considered.

A specific objective is to provide the necessary supporting information such that recommendations can be made for the development of the capillary extrusion rheometry standard ISO 11443 to improve the method.

To address these issues testing has been performed on a number of thermoplastics including a range of polyethylenes that exhibit different degrees of chain branching and thus different extensional flow properties and entrance pressure drop behaviours.

2. CAPILLARY EXTRUSION RHEOMETRY

The capillary extrusion rheometry method is detailed extensively in ISO 11443 Plastics – Determination of the fluidity of plastics using capillary and slit die rheometers [1]. This standard covers the determination of both shear viscosity and entrance pressure drop values using capillary dies. Further details on the determination of entrance pressure drop values and their use to calculate extensional viscosities are presented by Rides and Allen [4]. Full details of the methods are not presented here, suffice to state that for any given instrument the test condition variables that are selected by the operator are:

- die geometries, i.e. length, diameter and entry geometry
- flow rates (and hence apparent shear rates)
- test temperatures

Shear viscosity is determined from the gradient of the Bagley plot of extrusion pressure versus die length to diameter ratio (L/D) where each line represents a single shear rate, for example as in Figure 1. The entrance pressure drop can be determined as the intercept of the line of constant shear rate with the y-axis, although it is also taken by some to be equivalent to the extrusion pressure obtained using a very short die. The intercept method is used herein.

For reasons of economy the use of two capillary dies, one short and one long but of the same diameter, are often used to obtain both shear viscosity and entrance pressure drop data – in effect a two-point Bagley plot (i.e two L/D values). Additional dies can be used to obtain an understanding of the non-linearity of the Bagley plots which can be an indication of, for example, pressure dependence of viscosity or viscous heating. However, at least 4 dies, or 3 dies with repeat testing, must be used to determine with any certainty whether the observed effect is due either to pressure dependence of viscosity or viscous heating, or to experimental scatter and material variability. The question remains as to how such behaviour should best be handled in terms of, for example, obtaining shear viscosity for use in flow simulation.

Due to the difficulties in manufacturing and calibrating very short dies and their increased susceptibility to damage, dies of length shorter than 0.2 mm have not been used in this investigation. Dies of 1 mm diameter were used except where otherwise stated.

The choice of flow rates and test temperatures are limited by the material behaviour and all materials must be assessed, on a case by case basis, as to whether they can meet the criteria specified in the capillary extrusion rheometry standard ISO 11443 and the acquisition and presentation of comparable multi-point data standard ISO 11403-2. The choice of flow rate will in particular be limited by the onset of melt distortion of the extrudate as the shear rate increases. This will limit the upper apparent shear rate at which data can be obtained (data obtained whilst melt distortion is occurring are invalid).

The effect of the choice of die geometry on the determined shear viscosity and entrance pressure drop values is now considered.

3. MATERIALS

Five materials have been investigated in this work. A high density polyethylene (HDPE) was provided by NIST [5]. Three further polyethylenes exhibiting significant differences in their extensional flow behaviour (high density polyethylene (HDPE), a low density polyethylene (LDPE) and a linear low density polyethylene (LLDPE)) and a glass-fibre filled polypropylene (GFPP) were also tested. The NPL references for these materials are:

HDPE (NIST)	HHC000
HDPE	HGH800
LDPE	HGE000
LLDPE	HGF000
GFPP	HFQ000

4. EFFECT OF DIE GEOMETRY ON SHEAR VISCOSITY AND ENTRANCE PRESSURE DROP

For the HDPE (HHC000) the Bagley plot was linear over the entire range from 0.25 to 30 of die length to diameter ratio (L/D), Figure 1. Even when considering only short dies of L/D less than 1 the best straight line fit to all the data (L/D from 0.25 to 30) gives an excellent fit, demonstrating that the data are an excellent linear fit over the entire range of die geometries. A number of different die geometries (Table 1) were used to determine shear viscosity values, Figure 3. Test reference 72 was for testing using 2 mm diameter dies, whereas other

tests were carried out using a 1 mm diameter die. Test reference 73 was for the long die having a L/D ratio of only 8, which is significantly shorter than that permitted by ISO 11443. Nevertheless the agreement with data obtained using longer dies was very good. Removing such test data, Figure 4, clearly reveals the small scatter in shear viscosity values obtained from many repeat measurements using the same conditions except for die length. The effect of the die geometry on entrance pressure drop values is presented for all tests in Figure 5. Again the removal of the 2 mm diameter die and the short L/D ratio results reveals the small scatter in results obtained from repeat measurements using the same conditions except for die length, Figure 6.

For the LDPE (HGE000) the onset of melt distortion limited the maximum shear rate that was achievable to $\approx 30 \text{ s}^{-1}$, Figure 7. The full Bagley plot shows reasonable linearity: the variations apparent from the best fit straight lines are considered to be due to scatter rather than a true effect such as pressure dependence of viscosity. The reduced Bagley plot shows that the scatter of data from the best fit lines to all the data is comparatively high, Figure 8.

For the HDPE (HGH800) again melt distortion was apparent at shear rates above 120 s^{-1} for the longer dies ($L/D \geq 8$), Figure 9. However, for the shorter dies melt distortion was not so prevalent although it is considered that the data were not reliable, Figure 10. From examination of the raw pressure traces it is speculated that the onset of distortion in one barrel with resultant pressure fluctuations therein may have caused transients to occur in the other barrel that affected measurements therein¹. For apparent shear rates $\leq 120 \text{ s}^{-1}$ the Bagley plot shows excellent linearity over the range of L/D, Figure 9. Again, scatter in values is more significant than any non-linearity of the data.

For the LLDPE (HGF000) reasonable linearity of the Bagley plot was obtained and again the plot to all the data fitted the restricted data for $L/D < 1$ reasonably well, Figures 11 and 12. The derived shear viscosity and entrance pressure drop data, Figures 13 and 14 respectively, indicate the low level of scatter in data obtained using the different combinations of die geometries.

For the glass-fibre filled material (HFQ000) the scatter in data appears to be larger although reasonably linear fits are still obtained, Figures 15 and 16. The extent of the scatter in shear viscosity and entrance pressure drop values is clearly shown in Figures 17 and 18. In particular the scatter is large in the entrance pressure drop values.

Analysis of the data for all the materials showed that the range² in shear viscosity values was less than $\pm 10\%$, the higher values occurring for the glass-fibre filled material (HFQ000), Figures 19 and 20. For the unfilled materials only the range in shear viscosity values was up to $\pm 6\%$. For entrance pressure drop values the range in measured values was less than $\pm 10\%$, except at low shear rates where the range increased up to $\pm 15\%$ for HGE000. Furthermore, the range for the glass-fibre filled material was generally higher than for the unfilled materials at equivalent strain rates.

¹ The capillary rheometer used was of the twin barrel configuration where both barrels are driven by the same cross-head.

² Range is taken herein to be $0.5 (\text{maximum value} - \text{the minimum value})/\text{average value}$, expressed as a percentage.

5. DISCUSSION

The main objective of this work was to identify whether the die geometry had a significant effect on the derived shear viscosity and entrance pressure drop values. It has been demonstrated that the use of different die geometries resulted in reasonable linear Bagley plot for the materials and testing conditions used. No obvious systematic curvature due to pressure dependence of shear viscosity or viscous heating has been identified. The more significant factors in affecting the accuracy of the determination of shear viscosity and entrance pressure drop were two-fold.

The most significant limitation to the measurement of shear viscosity and entrance pressure drop was the onset of melt distortion. For the material HGH800 melt distortion was worse for the long die than the short die at equivalent shear rates, thus suggesting that longer dies are not necessarily beneficial when melt distortion occurs. Viscosities may be obtainable at higher shear rates in the absence of melt distortion by using shorter dies.

Secondly, the material variability as indicated by the repeat measurements was potentially significant, particularly for the fibre-filled material. The inclusion of glass fibres in the material increased the scatter in results but did not appear to produce a non-linear Bagley plot.

If the percentage scatter in pressure values was independent of die length then the scatter in derived viscosity values would also be independent. It is noted however, although not conclusively, that the scatter for the $L/D = 30$ dies appeared larger than for shorter dies for the HDPE (HHC000) and LLDPE (HGF000), Figures 1 and 11. Thus any potential benefits of using the pressure transducer in the higher part of its range are not necessarily realised.

6. CONCLUSIONS

In conclusion:

- Based on the linearity of the Bagley plots, the use of a long die of length to diameter ratio of 16, rather than a value of 20 or greater, did not significantly affect the derived shear viscosity and entrance pressure drop values.
- The selection of a short die of length to diameter ratio from the range 0.21 to 1 did not significantly affect the derived shear viscosity and entrance pressure drop values, the latter being derived by extrapolation to a die length to diameter ratio of zero.
- The results indicate that selecting a short die from the range 0.21 to 1 and assuming that the extrusion pressure was equal to the entrance pressure drop would have a significant affect in degrading the reproducibility of entrance pressure drop measurement.
- The greatest factors influencing the measurement of shear viscosity and entrance pressure drop values were the onset of melt distortion and the repeatability of the individual extrusion pressure measurements.

7. ACKNOWLEDGEMENTS

The work was carried out as part of a project on viscoelastic measurement techniques for polymer melts (MPM 1.2, milestone 7). This project is part of a programme of underpinning research financed by the Engineering Industries Directorate (EID) of the Department of Trade and Industry on measurements related to the processability of materials.

8. REFERENCES

1. ISO 1133: Plastics - Determination of the fluidity of plastics using capillary and slit-die rheometers.
2. ISO/TC61/SC5/WG9: ISO Plastics / Physical-chemical properties / Melt Rheology Committee.
3. ISO 11403 Plastics - Acquisition and presentation of comparable multipoint data - Part 2: Thermal and processing properties.
4. Rides, M and Allen, CRG, Extensional flow properties of polymer melts using converging flow methods, Measurement Good Practice Guide No. 16, National Physical Laboratory, Teddington, TW111 0LW, UK.
5. NIST Standard Reference Material 1497. NIST, Gaithersburg, MD20899, USA.

TABLE 1: CAPILLARY DIE GEOMETRIES USED IN TESTING

Test reference	Material	Long die length, mm	Short die length, mm
ros127	HGE000	30	0.95
ros128	HGE000	20.004	0.322
ros129	HGE000	16.048	0.254
ros130	HGE000	n/a	8.023
ros131	HGE000	n/a	0.213
ros120	HFQ000	30	0.95
ros121	HFQ000	20.004	0.322
ros122	HFQ000	16.048	0.254
ros123	HFQ000	8.023	0.213
ros124	HFQ000	16.048	n/a
ros125	HFQ000	8.023	n/a
ros116	HGF000	30	0.95
ros117	HGF000	20.004	0.322
ros118	HGF000	16.048	0.254
ros119	HGF000	8.023	0.213
ros112	HGH800	30	0.95
ros113	HGH800	20.004	0.322
ros114	HGH800	16.048	0.254
ros115	HGH800	8.023	0.213
ros071	HHC000	16.048	0.213
ros072	HHC000	$32.01 \times 2^*$	$0.354 \times 2^*$
ros073	HHC000	8.023	0.213
ros105	HHC000	20.004	0.322
ros106	HHC000	20.004	0.322
ros107	HHC000	30	0.95
ros108	HHC000	30	0.95
ros110	HHC000	16.048	0.254
ros111	HHC000	8.023	0.254

*All dies 1 mm except for tests ros072 where 2 mm diameter dies were used (as indicated by x 2).

Note: Up to three determinations of shear viscosity and entrance pressure drop were obtained per test reference.

Ref: MATC(A)56_improved_capillary_1oct01_v#3.doc

FIGURES

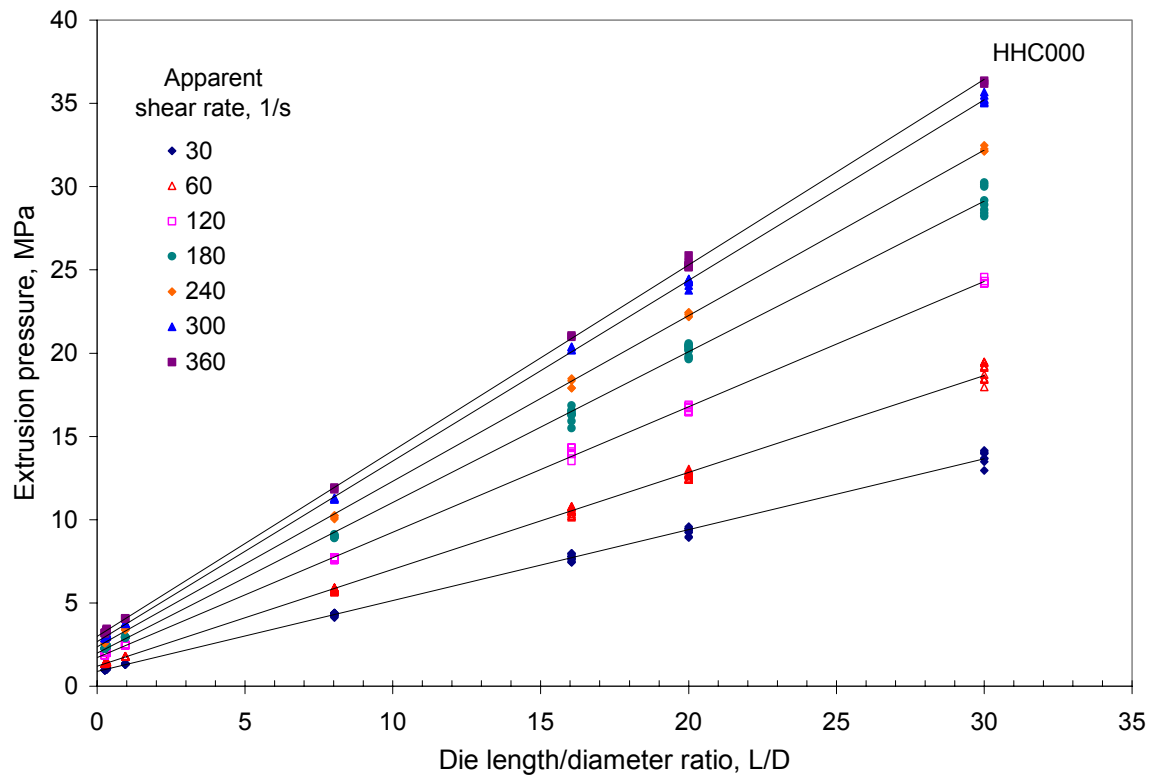


Figure 1: Bagley plot for a HDPE (HHC000) at 190 °C.

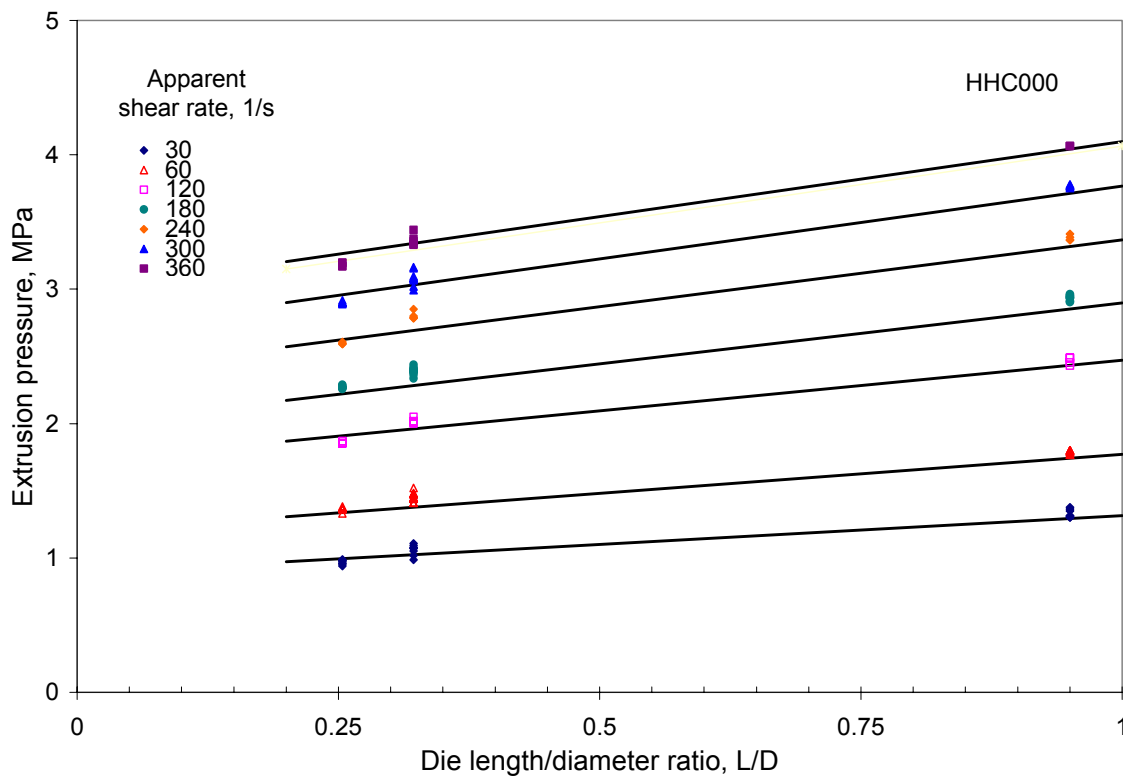


Figure 2: Restricted Bagley plot for a HDPE (HHC000) at 190 °C showing data for dies of L/D ratio up to 1 only, with best fit straight lines fitted to all L/D data (see Figure 1).

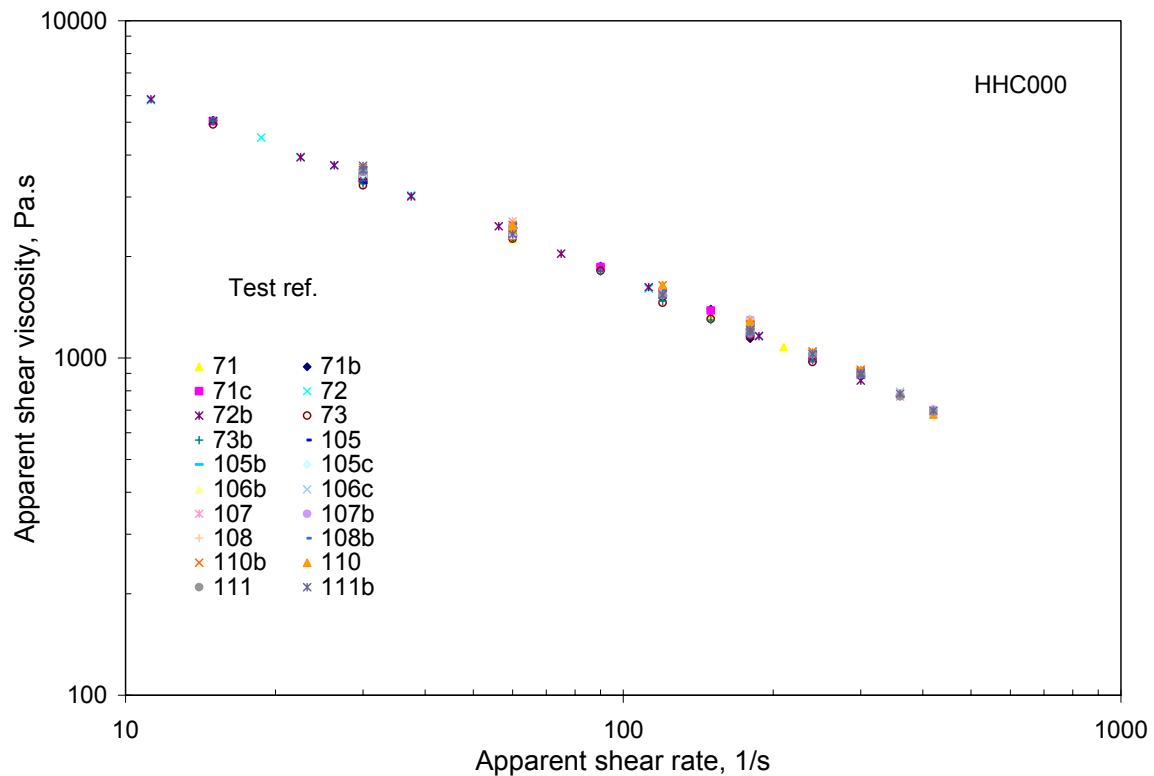


Figure 3: Shear viscosity data for a HDPE (HHC000) at 190 °C.
(Note: 71b and 71c in the test references indicate repeat tests using the same barrel charge as for 71)

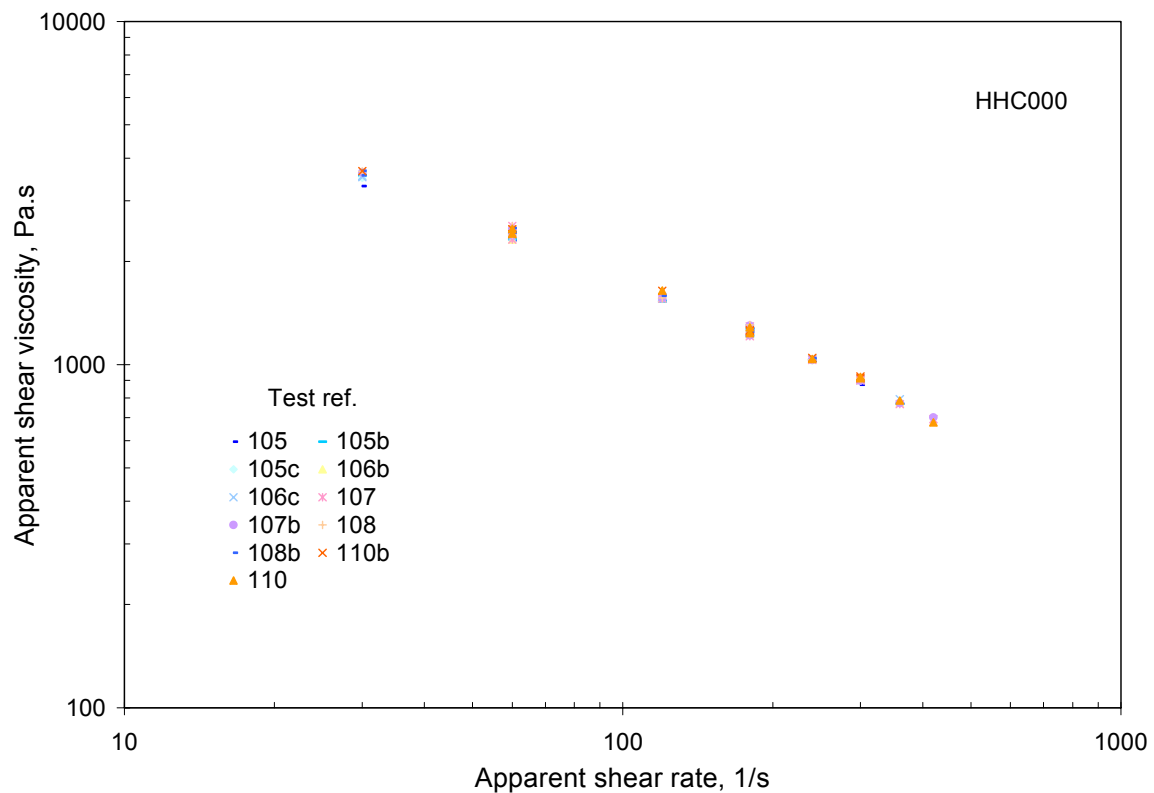


Figure 4: Shear viscosity data for a HDPE (HHC000) at 190 °C showing the repeatability of measurements using the same testing conditions except for die length.

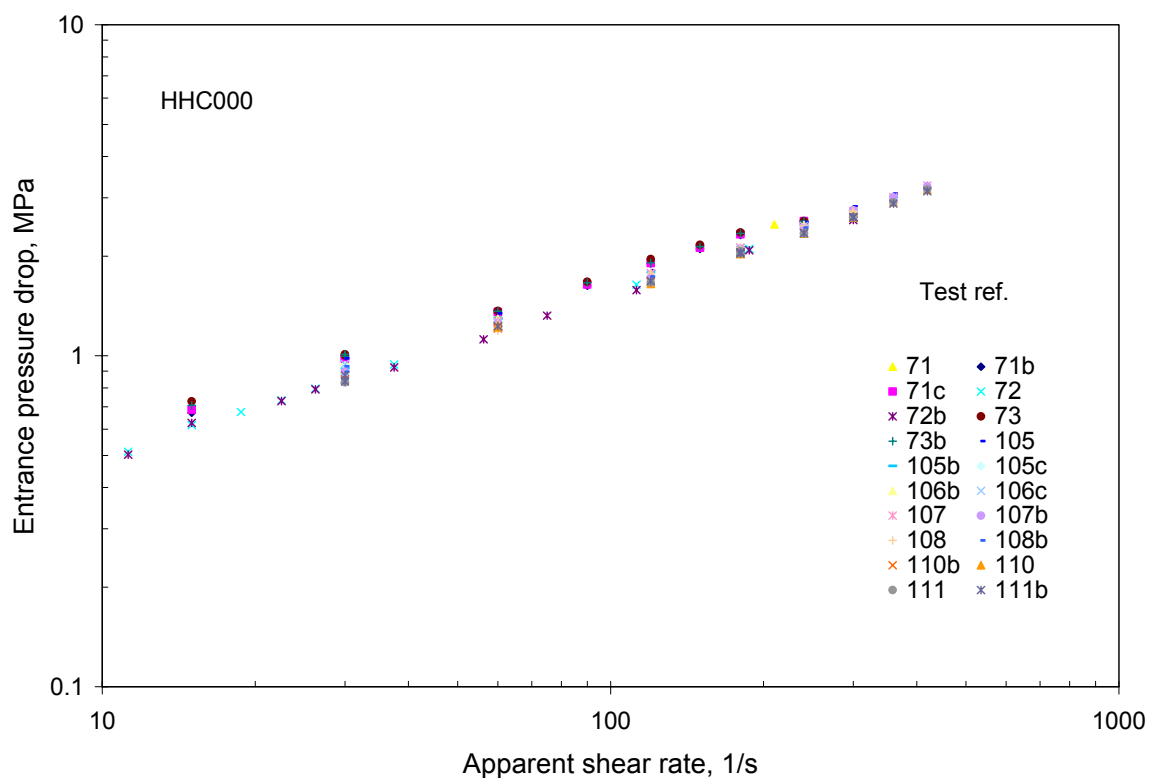


Figure 5: Entrance pressure drop data for a HDPE (HHC000) at 190 °C.

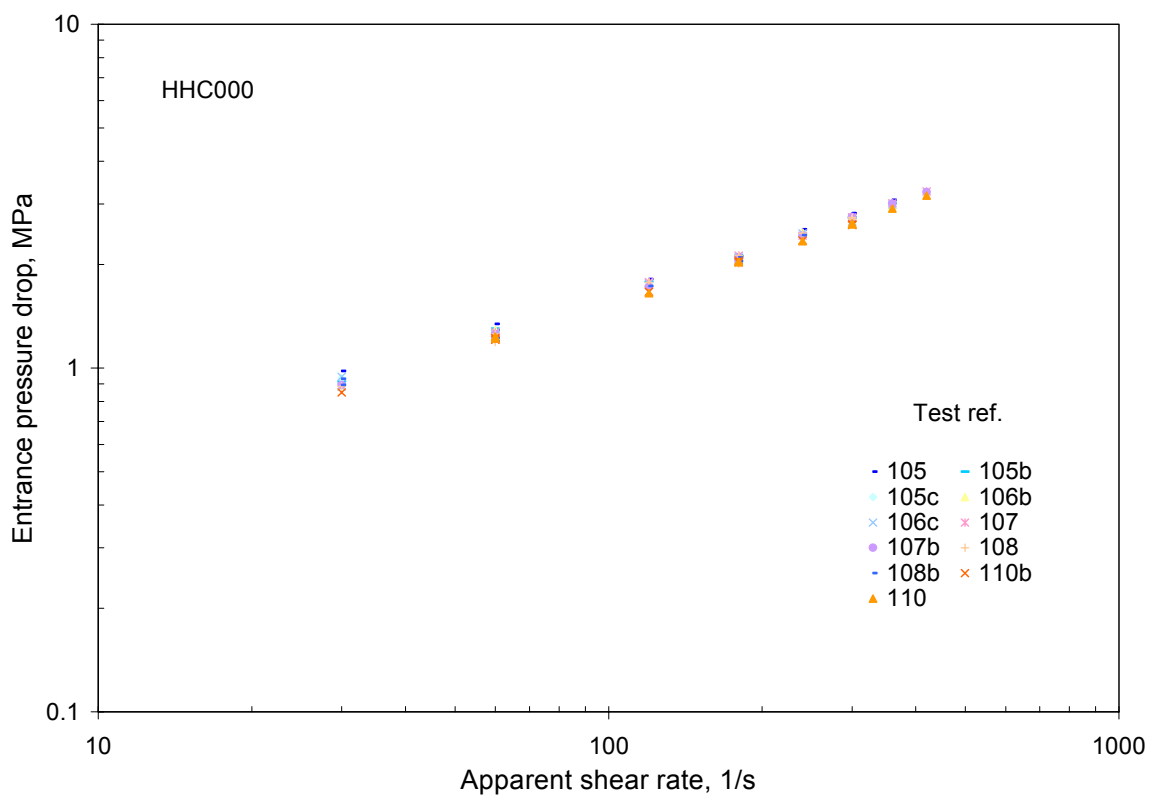


Figure 6: Entrance pressure drop data for a HDPE (HHC000) at 190 °C showing the repeatability of measurements using the same testing conditions except for die length.

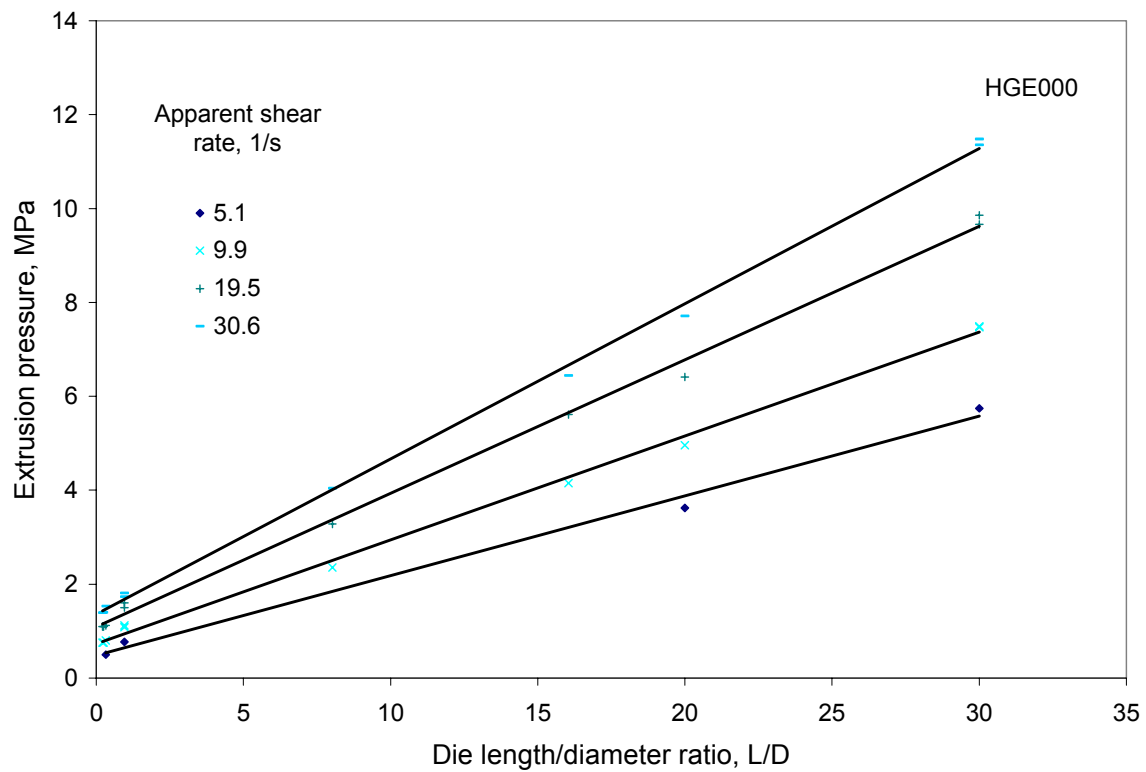


Figure 7: Bagley plot for a LDPE (HGE000) at 190 °C. Melt distortion occurred at an apparent shear rate above 30 s^{-1} .

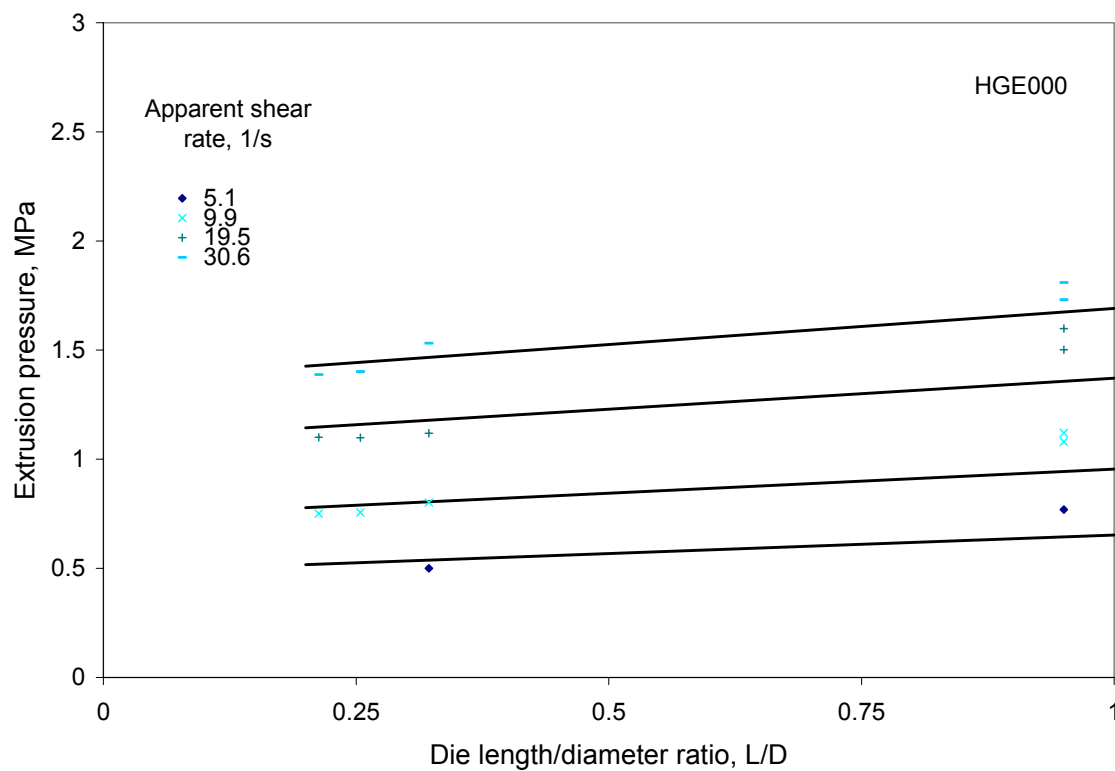


Figure 8: Restricted Bagley plot for a LDPE (HGE000) at 190 °C showing data for dies of L/D ratio up to 1 only, with best fit straight lines fitted to all L/D data (see Figure 7).

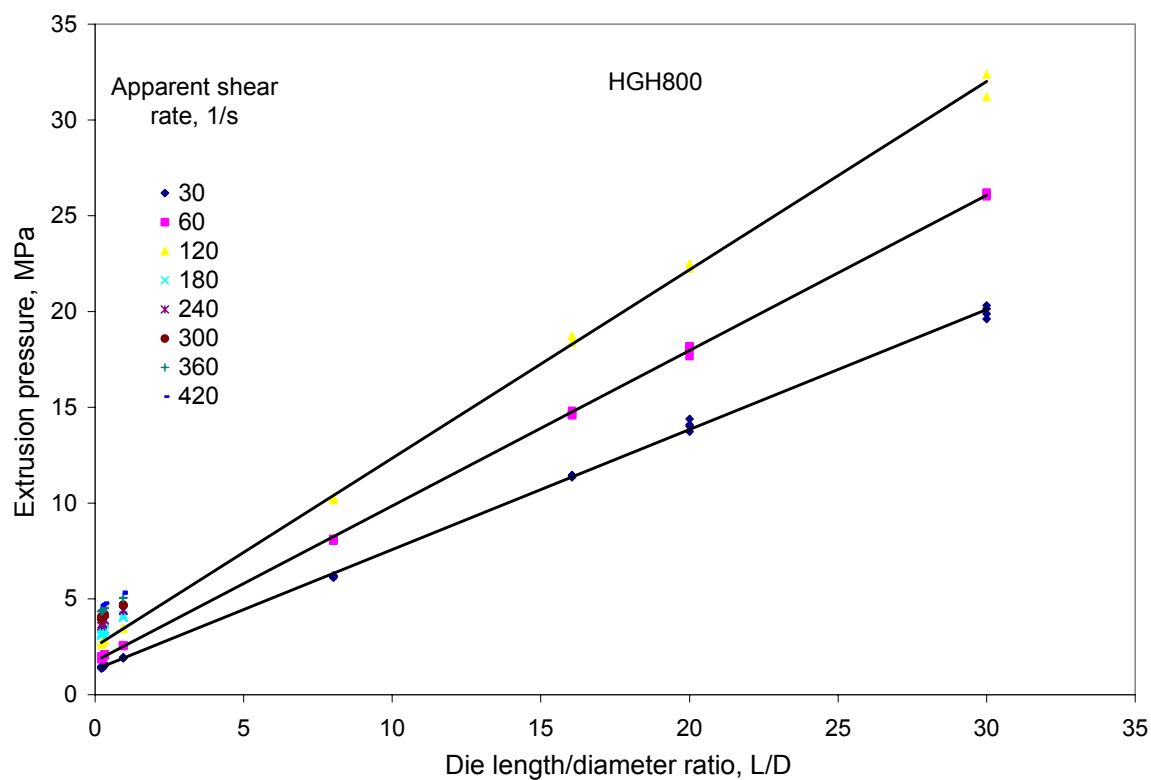


Figure 9: Bagley plot for a HDPE (HGH800) at 190 °C. Melt distortion at an apparent shear rates above 120 s⁻¹ for long dies.

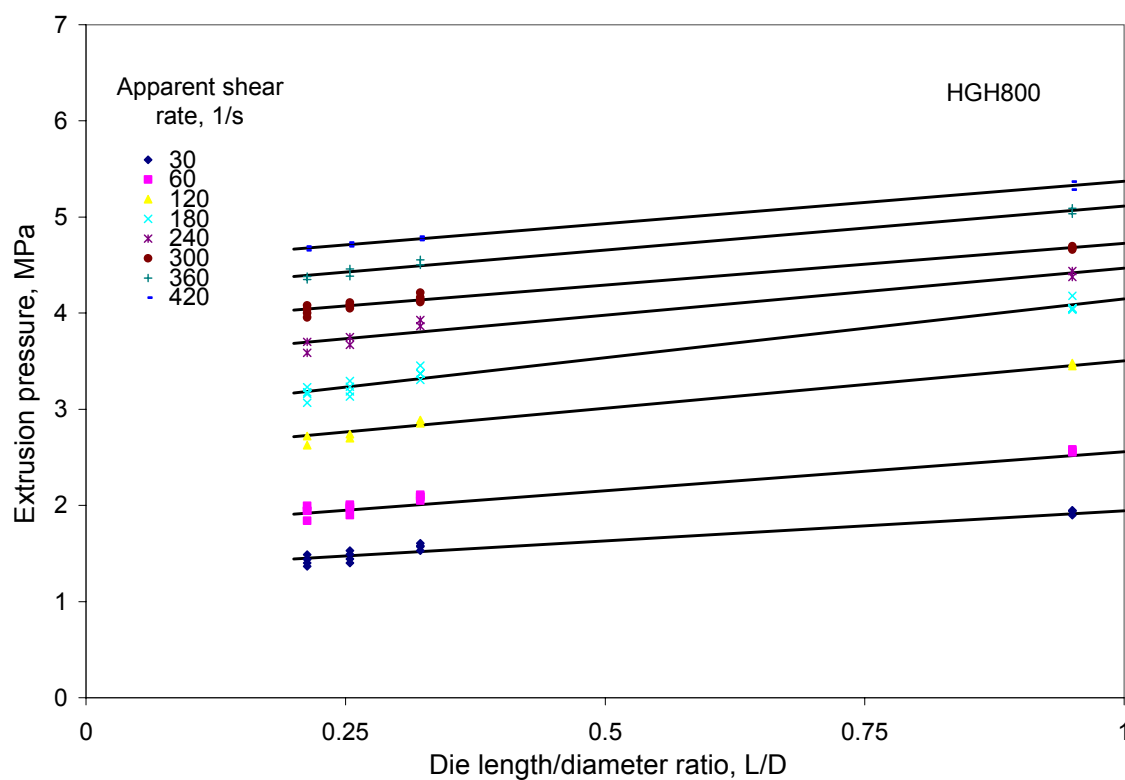


Figure 10: Restricted Bagley plot for a HDPE (HGH800) at 190 °C showing data for dies of L/D ratio up to 1 only, with best fit straight lines fitted to all L/D data (see Figure 9).

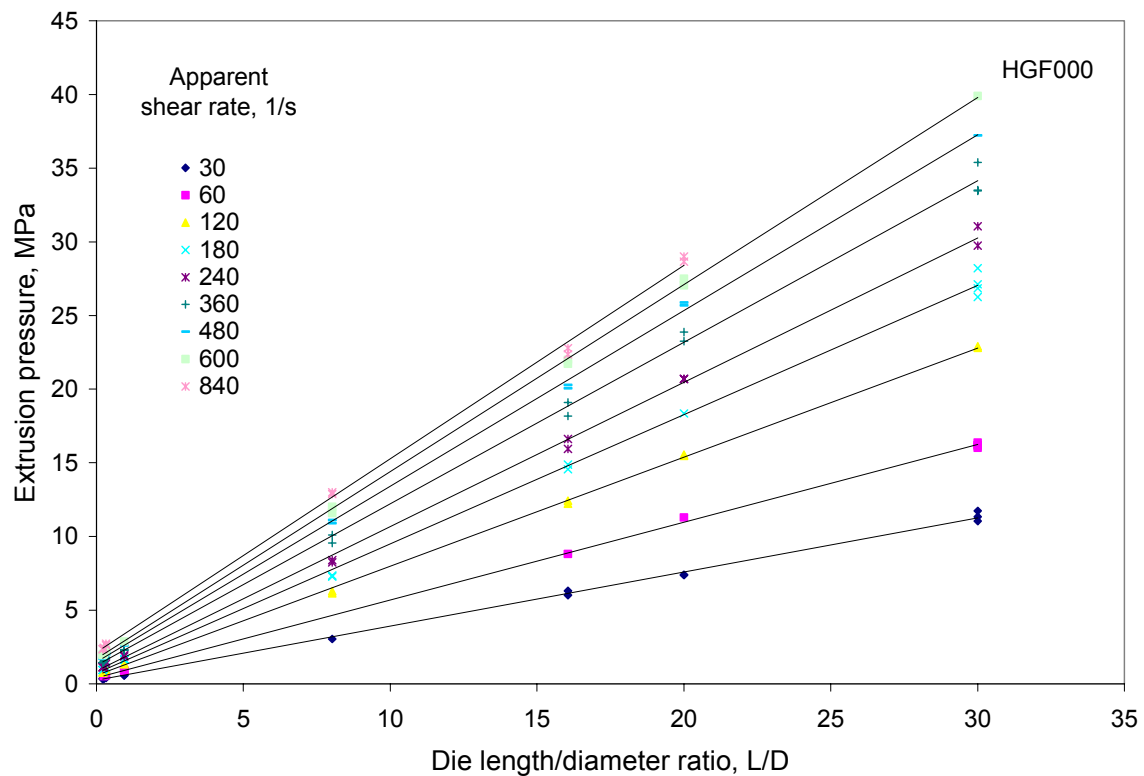


Figure 11: Bagley plot for a LLDPE (HGF000) at 190 °C.

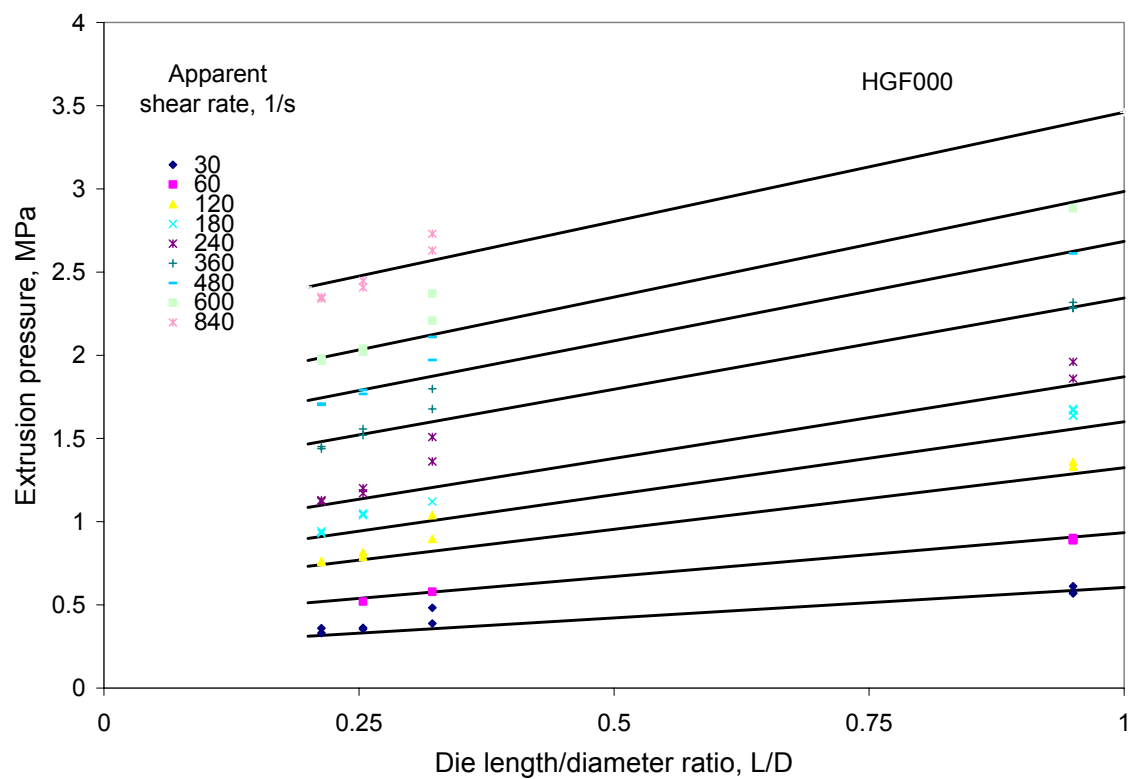


Figure 12: Restricted Bagley plot for a LLDPE (HGF000) at 190 °C showing data for dies of L/D ratio up to 1 only, with best fit straight lines fitted to all L/D data (see Figure 11).

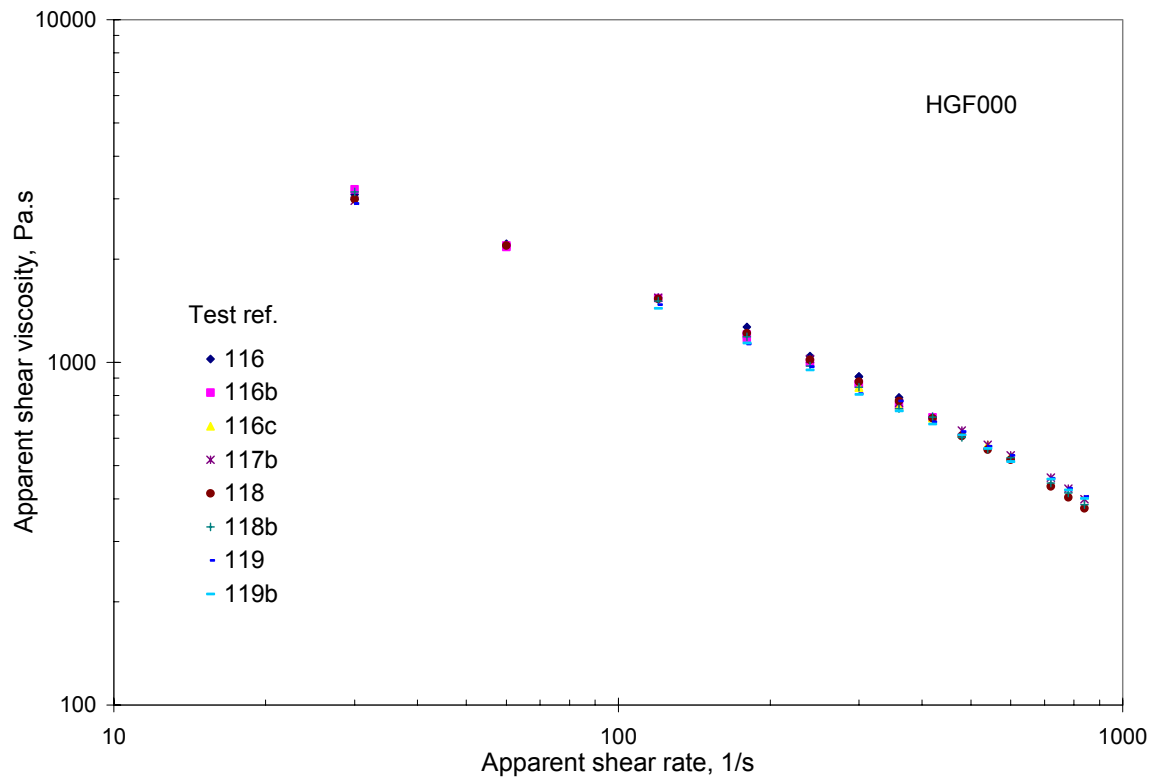


Figure 13: Shear viscosity plot for a LLDPE (HGF000) at 190 °C.

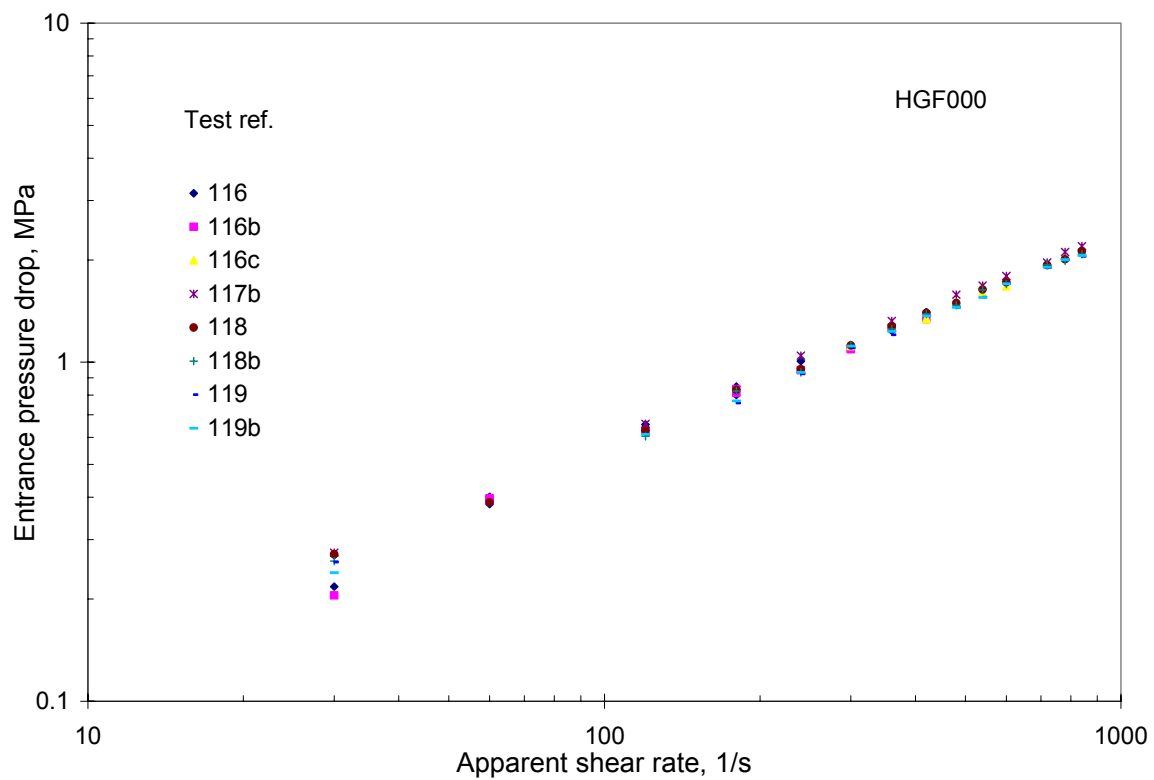


Figure 14: Entrance pressure drop plot for a LLDPE (HGF000) at 190 °C.

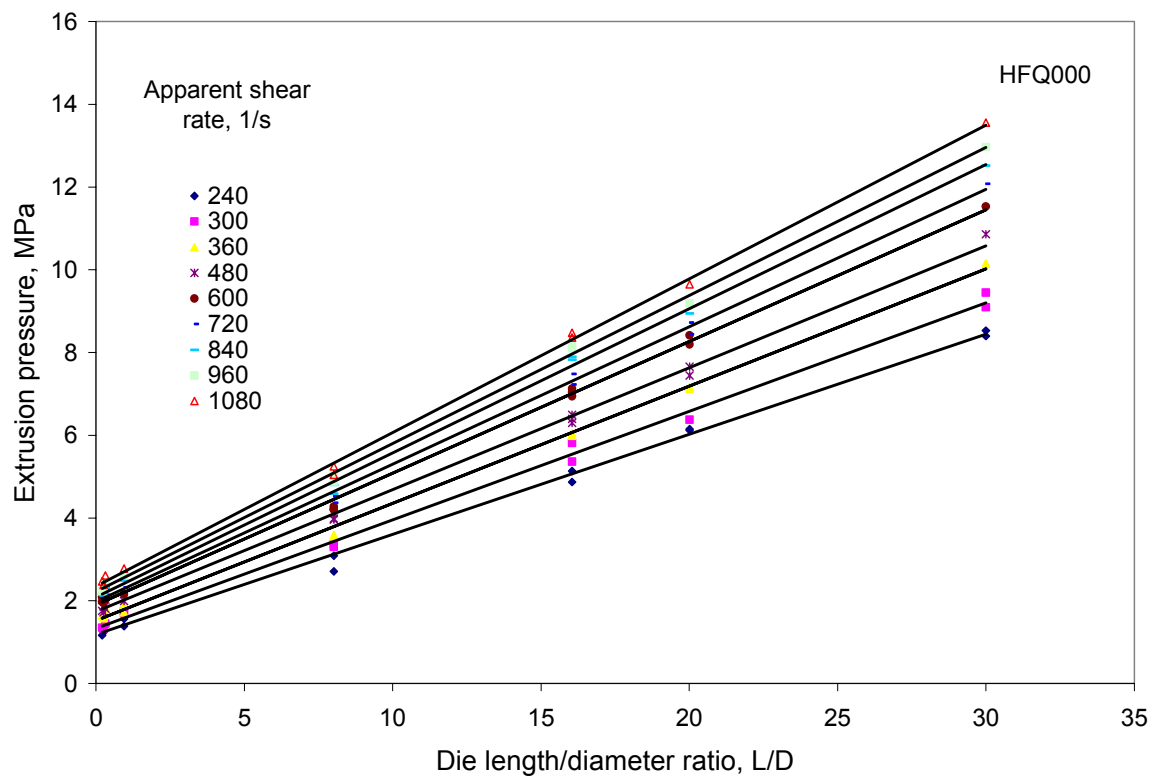


Figure 15: Bagley plot for a glass-fibre filled PP (HFQ000) at 200 °C.

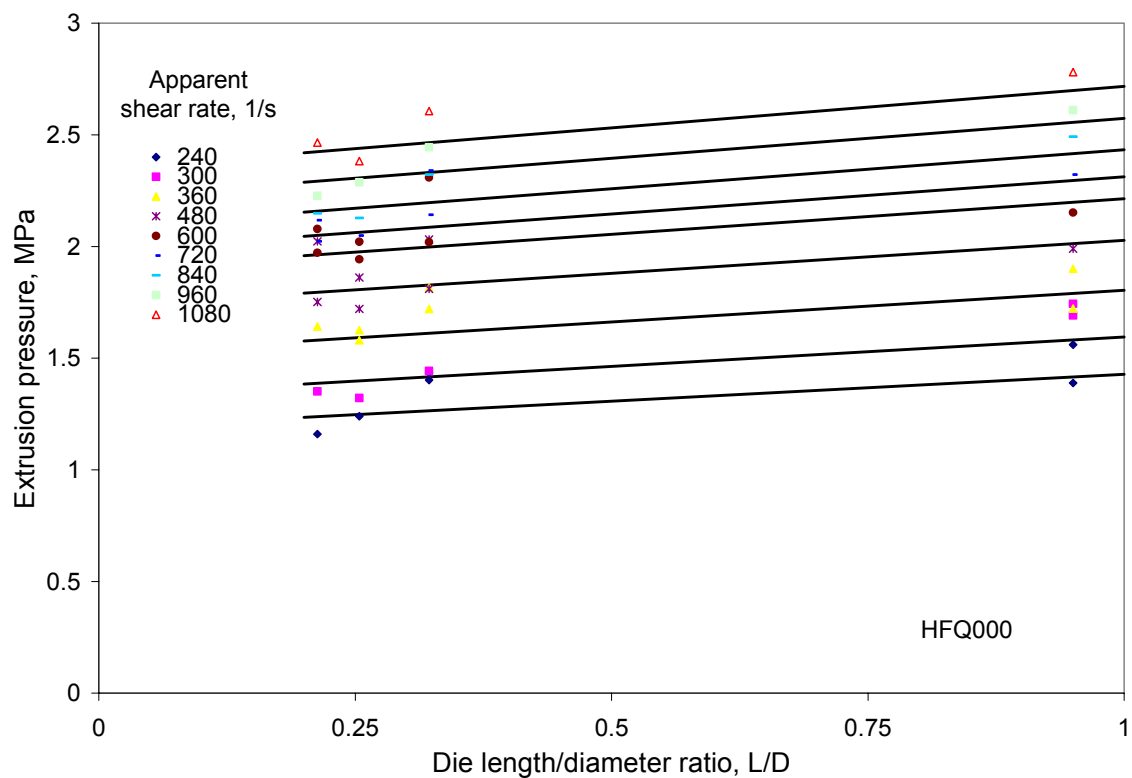


Figure 16: Restricted Bagley plot for a glass-fibre filled PP (HFQ000) at 200 °C showing data for dies of L/D ratio up to 1 only, with best fit straight lines fitted to all L/D data (see Figure 15).

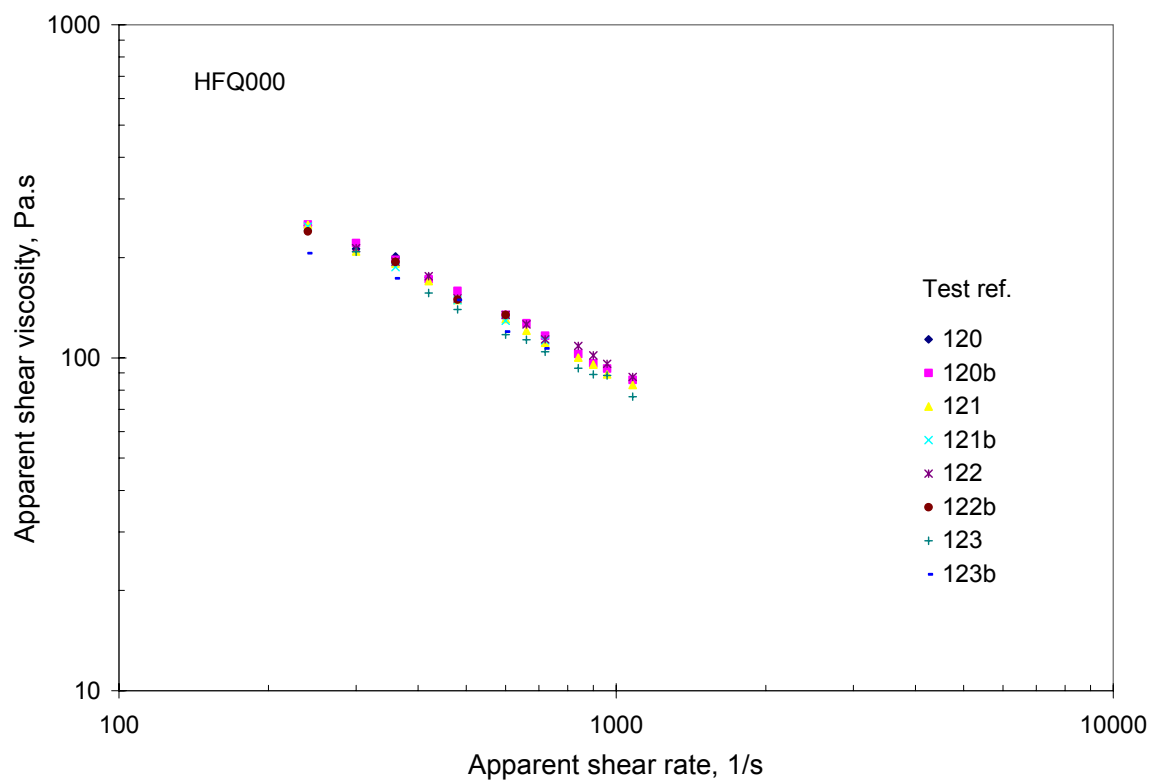


Figure 17: Shear viscosity plot for a glass-fibre filled PP (HFQ000) at 200 °C.

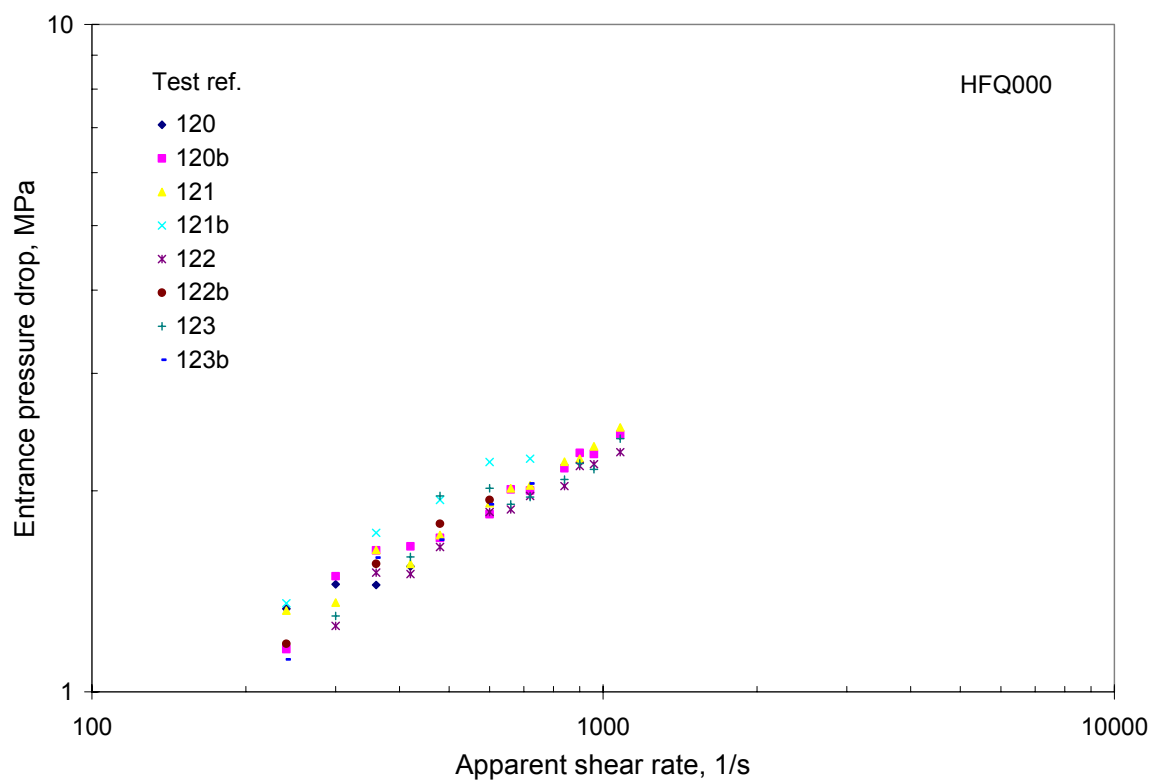


Figure 18: Entrance pressure drop data for a glass-fibre filled PP (HFQ000) at 200 °C.

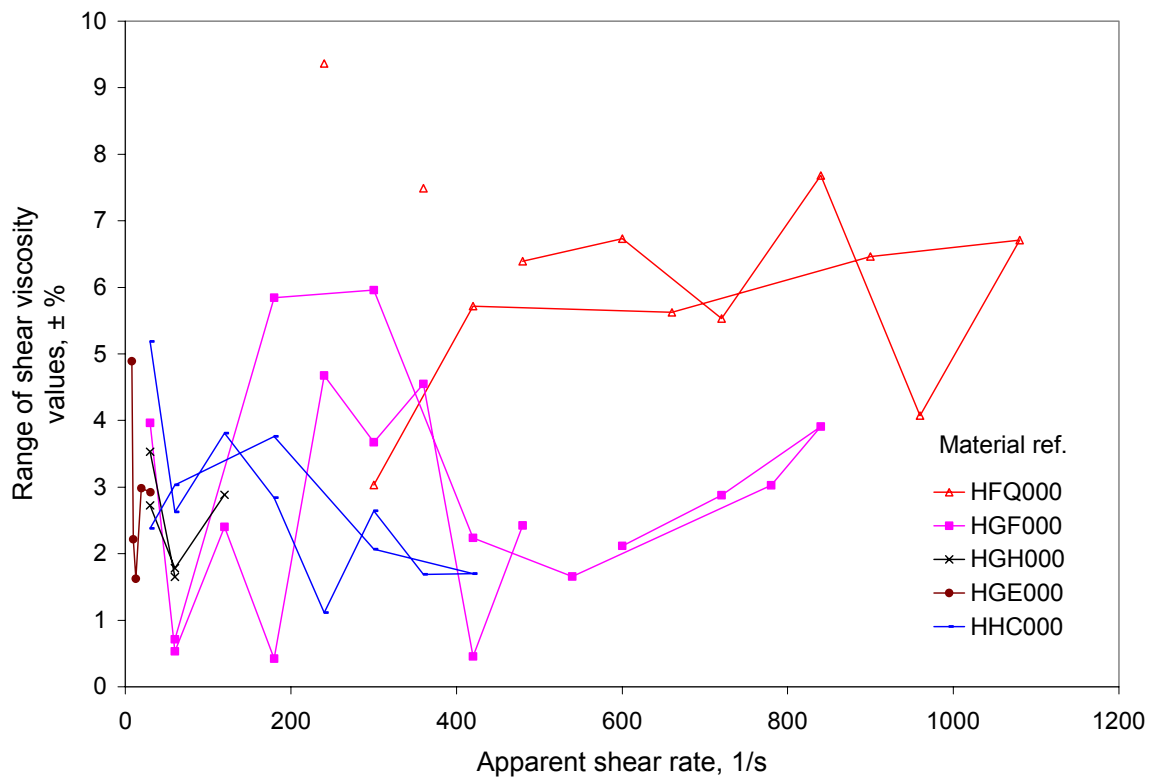


Figure 19: Effect of shear rate on the scatter in shear viscosity values for all materials.

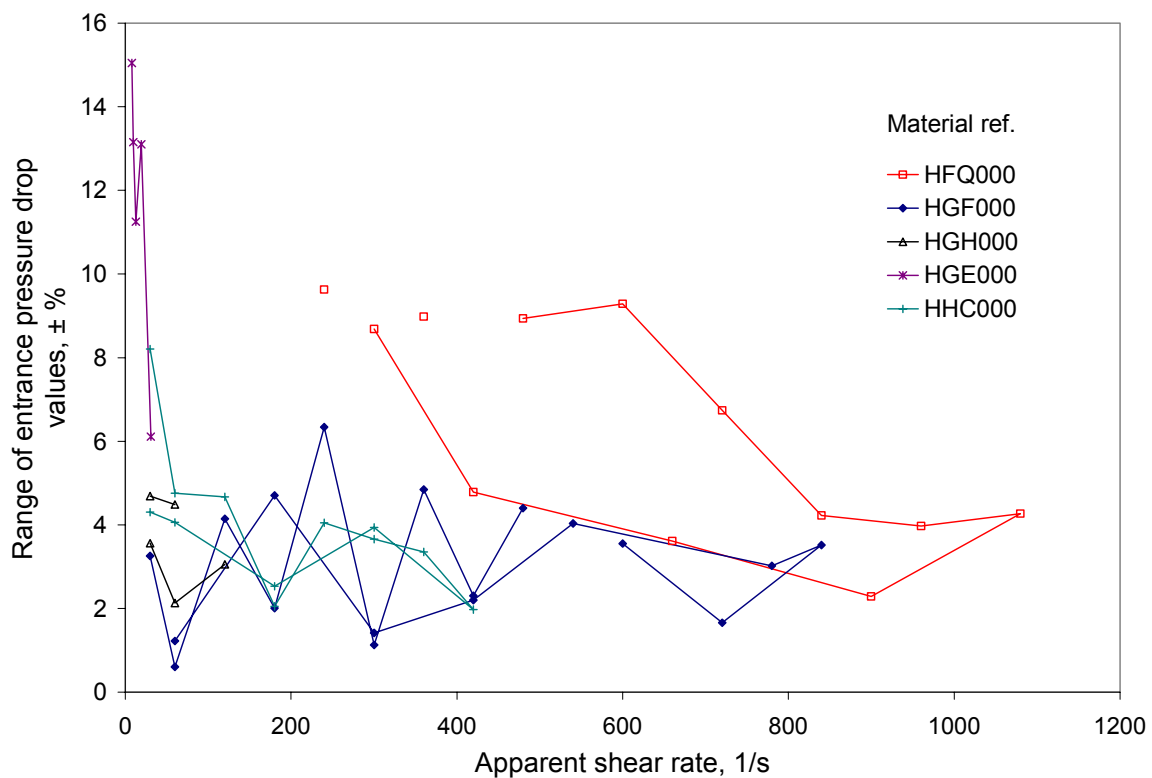


Figure 20: Effect of shear rate on the scatter in entrance pressure drop values for all materials.