Measuring The Extensional Flow Properties Of Polymer Melts

Summary

In many polymer-forming processes the polymer undergoes significant extensional or stretching flow, for example in blow moulding, vacuum forming and film extrusion. The extensional flow behaviour of materials will therefore have a significant effect on their processability and also on the properties of the moulded product. However, established rheological techniques predominantly characterise the behaviour of polymers in shear and it can prove difficult to relate the behaviour measured in shear with that observed in processing. An ability to characterise their behaviour more appropriately will have benefits in materials development and in process and product design.

The techniques most suitable for characterising the extensional flow behaviour of polymer melts are converging flow methods that use capillary extrusion rheometry data and stretching flow methods. These techniques have been the focus of recent research at NPL. The methods were critically reviewed [1, 2]. These reviews also identify sources of information on testing of a range of materials. Evaluation of the converging flow approach to determine extensional viscosity has been carried out and is reported in detail elsewhere [3]. To address measurement issues concerning the stretching flow methods an extensional rheometer was developed at the NPL. This note briefly describes that instrument for measuring the extensional viscoelasticity of molten polymers. It also illustrates the use of that instrument in characterising the flow properties of three quite different polymers: a high density polyethylene (HDPE), a low density polyethylene (LDPE) and a linear low density polyethylene (LLDPE), and also in an industrial case study of three similar high density polyethylenes.

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A Stretching Business: The Method

The NPL has developed an instrument for measuring the extensional viscoelasticity of polymer melts (see photograph). It has been used up to 230°C and has a maximum operating temperature of 250°C, limited by the silicone oil into which the specimen is immersed. The operating principle of the instrument is to stretch a molten specimen between a rotating clamp and a fixed clamp. The fixed clamp is used to measure the tensile force developed in the specimen during stretching. The method is described in further detail elsewhere [4].
Measurement Of Three Different Polymers

Three grades of polyethylene, a linear low density polyethylene (LLDPE), a low density polyethylene (LDPE) and a high density polyethylene (HDPE) were investigated using this instrument, Figures 1 and 2. These materials exhibited significant differences in their extensional strain hardening behaviour. The LDPE exhibited greatest strain hardening with extensional viscosity values increasing significantly with increasing strain. In comparison, the LLDPE exhibited no further strain hardening beyond a strain of ≈ 0.5. High strain rates up to 10 s⁻¹ have been achieved using this instrument.
Figure 1: Significantly different strain hardening behaviour of three polyethylenes tested at 150°C and at a strain rate of 0.7 s\(^{-1}\).

Of significance was that the shear viscosities of these three materials in order of decreasing magnitude were LLDPE, LDPE and then HDPE, although HDPE and LDPE had similar values at a shear rate of 100 s\(^{-1}\). The shear viscosity of the LLDPE was \(\approx 40\%\) higher than that of the HDPE. This ranking differs from that of the extensional viscosity, Figure 1, indicating that the reliance on shear flow data alone can be misleading, particularly when using that information to predict how a material will perform in a predominantly extensional flow process.

Figure 2: Effect of strain rate on the extensional flow behaviour of a high density polyethylene.

Measurement Of Three Similar Polymers

Obviously, the results presented above clearly demonstrate the importance of extensional flow measurements. However, it does not demonstrate the capability of the method for discriminating between similar materials. After all, the designer or processor usually has to make a choice between similar materials rather than different material types. Durapipe S&LP were particularly interested in three HDPEs that have obtained PE100 rating, referred to as HGZ000, HHA000 and HHB000. Results of testing of these materials, supplied by Durapipe S&LP, showed that at low shear rates there was no significant difference in their shear viscosities. At shear rates greater than 100 s\(^{-1}\) melt distortion appears to have occurred, Figure 3. These data thus provide a limited basis on which to compare the materials. Analysis of entrance pressure drop data at the lowest rates indicated that HHB000 had the highest values, by \(\approx 50\%\), and HGZ000 and HHA000 had similar values. The difference between HGZ000 and HHA000 was not clear, possibly due to experimental scatter. At the lowest rates HGZ000 had slightly higher values of entrance pressure drop but at higher rates its values were lower.
Figure 3: Comparison of shear viscosities of three polyethylene materials at 200°C, suggesting melt fracture at shear rates above 100 s\(^{-1}\).

The extensional rheometer developed at NPL was used to test these materials. Testing was carried out at three strain rates 0.1, 1 and 7.9 s\(^{-1}\) at 200°C, Figure 4. At each of these three rates the trend in extensional viscosity values was, in order of decreasing value, HHB000, HGZ000 and HHA000. The difference between HHB000 and HGZ000 was \(\approx 30\%\), and the difference between HGZ000 and HHA000 was \(\approx 10\%\). The scatter in the data obtained using the lowest strain rate at high strain was due to the forces measured at these conditions approaching the resolution of the instrument. Under such conditions the forces are very low due to the significant reduction in the cross sectional area of the specimen.

Figure 4: Comparison of the extensional stress growth coefficient behaviour of three high density polyethylenes at three different strain rates at 200°C.

One of the reasons for wanting to characterise these materials was to understand how they compared, and to
relate their measured behaviour to their performance both in extrusion and injection moulding. The differences apparent in the extensional flow behaviour suggest that the material HHB000, having the highest extensional viscosity, may be more suitable for thick-walled pipe extrusion and less prone to wall thickness variations in the pipe. Because of pipe specifications, it is necessary to use the same quality of material for the pipe fittings as is used for the pipe. Thus the suitability of these materials for injection moulding is also important. The results also indicate that HHB000, having the highest extensional viscosity, would be the most problematic of the materials for injection moulding, requiring higher pressures to force material through the gate region for example.

**Conclusions**

In conclusion, the use of the new rheometer in characterising the extensional flow behaviour of polymer melts has been demonstrated.

The comparison of extensional viscoelasticity data with shear viscosity data clearly demonstrates that materials characterisation solely on the basis of shear flow measurements is inadequate, particularly when the data are required for predicting the performance of materials in processes that are predominantly extensional flows.

**Footnotes**

1. "Extensional viscosity" is used as a colloquialism herein for the term "tensile stress growth coefficient", equivalent to the ratio of tensile stress to strain rate.

2. Hencky strain and strain rates are used herein.

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**References**


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