Measurement Good Practice Guide No. 63

Extensional Flow Properties of Polymers using Stretching Flow Methods

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Abstract:

In many forming processes the polymer melt undergoes predominantly extensional deformation, for example in blow moulding and film blowing. Furthermore, the extensional flow behaviour of melts differ significantly from their shear flow behaviour. Thus extensional flow measurement methods are necessary to appropriately characterise the melt behaviour. Such data can be used for the development and selection of materials, and for improving processing.

This Guide discusses various issues related to the need for, and measurement of the extensional flow properties of polymer melts using stretching methods. It describes practical considerations for measuring extensional flow behaviour, including measurements at very high and low strain rates, and includes details on the analysis of the raw data and assessment of the effect of uncertainties on the measured values. To illustrate the measurement technique, typical data obtained using the NPL extensional rheometer are presented along with results obtained from an international intercomparison.

This Guide is aimed at those carrying out, or intending to carry out, such testing or are building/modifying test equipment. It complements the Standard that is being developed in ISO (ISO/CD 20965 Plastics – Determination of the transient extensional viscosity of polymer melts) and provides additional information to the Standard that will assist in making and analysing such measurements, and in understanding the results..

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Foreword

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This Guide describes methods for determining the extensional flow behaviour of plastics melts using tensile stretching methods. It builds on and supercedes the earlier Measurement Good Practice Guide No.18 - Extensional flow properties of polymer melts using stretching flow methods (May 1999). In particular, it provides guidance on testing at very high strain rates and very low strain rates; contains an improved analysis of the uncertainties in testing; and presents results of an intercomparison of extensional flow testing. Since the original publication of the Measurement Good Practice Guide No. 18, a draft ISO standard has been prepared and is currently being developed in ISO (ISO/CD 20965 Plastics – Determination of the transient extensional viscosity of polymer melts). Persons carrying out such measurements must also refer to the latest version of that Standard. This Guide provides additional information to the Standard that will assist in making and analysing such measurements, and in understanding the results.

In the drafting of this Guide, it has been assumed that the collection and analysis of data will be carried out by suitably qualified and experienced personnel.

Users of this Guide are invited to give feedback to the authors on any issues related to its use or content.

1. Scope

This Guide:

- summarises the key issues in measuring the extensional flow behaviour of polymer melts

- gives practical advice and defines good practice on how to obtain extensional viscosity values of polymer melts using stretching methods, including at high and low strain rates

- presents an assessment of the effect of uncertainties on the measured extensional viscosity values

It illustrates the measurement of extensional flow properties with reference to results obtained using an instrument developed by NPL, and also by reference to results obtained in an international intercomparison of extensional flow properties.

2. Introduction

In many plastics forming processes the melt undergoes predominantly extensional deformation, for example in blow moulding, fibre spinning, film blowing and vacuum forming [1-3]. Extensional flow also occurs in profile extrusion and injection moulding, for example in the nozzle of the injection moulding machine and in passing from the runner to the gate of the moulding. The extensional flow behaviour of polymer melts can differ significantly from their shear flow behaviour. For example, the ranking of materials in order of increasing extensional viscosity may not be the same as their ranking in order of increasing shear viscosity. Polymers may also exhibit additional trends in extensional flow behaviour not normally exhibited in shear, for example strain hardening. Thus extensional flow measurements are necessary to appropriately characterise the melt behaviour. Such data can be used for the development and selection of materials, quality control and for process design and optimisation.

There are two basic approaches to determining extensional flow properties of polymer melts: either tensile testing or converging flow methods. Converging flow methods were critically reviewed and guidance on their use is presented elsewhere [4,5]. Stretching flow methods were also critically reviewed elsewhere [6]. These reviews also identify sources of information on testing of a range of materials.

The converging flow approach for determining the extensional flow behaviour of polymer melts is based on the measurement of the resistance to flow in a contraction region. A converging flow is, by definition, a type of extensional flow. It is expected, therefore, that extensional viscosity might be obtained from the measurement of pressure drop and flow rate in a converging flow. The approach of using converging flow methods is appealing as experimental data are easily obtained from capillary extrusion rheometry testing, normally carried out to determine the shear viscosity of materials. Thus extensional viscosity values can be obtained from the same capillary rheometry tests as already carried out to determine shear viscosity at little or no extra experimental cost. However, in using this approach it is assumed that extensional viscosity is strain independent, and other significant assumptions are necessarily made in the analysis. Consequently, there is debate about the ability of converging flow methods to generate quantitatively accurate data, although such methods are considered to be suitable for qualitative characterisation [5].

In comparison, stretching flow methods can be used to generate quantitatively accurate data on the extensional viscoelasticity of polymer melts. These methods are the subject of this Guide.

In summary, this Guide:

- discusses various issues related to the need for, and measurement of the extensional flow properties of polymer melts (Section 3)

- defines approved terminology and presents definitions (Section 4)

- describes important considerations for stretching flow measurements (Section 5)

- describes practical considerations for measuring extensional flow behaviour, including testing at high and low strain rates, analysis of the raw data and assessment of the effect of uncertainties on the measured values (Sections 6 and 8)

- presents typical data obtained using the NPL extensional rheometer and from an international intercomparison (Section 7).

Details of the analysis of experimental data and of the uncertainty analysis are presented in the Appendices.

3. General measurement considerations

3.1 Extensional flow behaviour of polymer melts

In contrast to shear flows where reference is normally made only to steady shear flow behaviour, extensional flow behaviour is best described as being transient. The extensional viscosity may vary as a function of both strain and strain rate, particularly at low values of strain. This transient behaviour is due to the continued development of the molecular orientation caused by the extensional flow field over the range of strains typically encountered in testing and processing. In comparison, in processes that are predominantly shearing flow the shear strain values tend to be significantly higher and the process is dominated by the material's steady state rather than transient shear flow behaviour.

In describing the transient behaviour of materials in extension they may exhibit either an unbounded stress growth[1] behaviour [for example reference 7, 8] or an equilibrium extensional viscosity value typically at large strains [for example references 9-11] - see also Figure 15. An equilibrium extensional viscosity is dependent on strain rate but not on strain or time. A maximum in the tensile stress growth coefficient value was observed [8] although this was possibly due to large experimental errors occurring at large strains, rather than to a true material response. This topic is discussed in more detail elsewhere [6, 12].

In conclusion, to fully understand the flow behaviour of polymer melts in extension it is necessary to characterise their transient flow behaviour.

3.2 Extensional flow stretching methods

In carrying out extensional flow measurements there are four types of measurement that are normally made: constant strain rate, constant stress, constant force or constant speed. Extensional techniques can be described, following Nazem *et al* [13], as ‘controllable’ or ‘non-controllable’ [14, 15]. ‘Controllable’ implies that the instantaneous values of strain rate and stress are uniform throughout the specimen and that either the strain rate or stress is held constant with time.

The results of non-controllable methods, for example fibre spinning and converging flow analysis [16-18] tend to be difficult to interpret [4-6, 19] as they are neither constant stress nor constant strain rate [14]. This is emphasised by Gupta *et al* [16] who commented that uniaxial stretching methods are the preferred methods for obtaining fundamental data using constant strain rate or constant stress, but not constant force or speed methods. Cogswell [20]

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[1] For definitions see Section 4.

also concluded that constant speed or constant force methods provide no information that cannot be provided better by the ‘direct measurement’ constant stress or constant strain methods.

3.3 Industrial needs

The industrial needs for extensional viscoelasticity testing, specifically in terms of the strains, strain rates and temperatures were assessed by Rides [21]. In summary, it was concluded that there was a need for measurements typically up to Hencky strains of ≈ 2 and strain rates of up to $\approx 5\ \text{s}^{-1}$ at temperatures of up to $\approx 260\ ^{\circ}\text{C}$, although higher values in both strain and strain rate will sometimes be required, for example higher strain rates for wire coating. A Hencky strain of 2 corresponds to a change in length by a factor of ≈ ×8.

In carrying out extensional flow measurements there are four basic categories of measurement that can be made: constant strain rate, constant stress, constant force or constant speed. However, industrial processes can rarely, if at all, be described by a single one of these categories. For example although film blowing may be carried out at constant bubble pressure and constant nip (or wind-up) tension, modelling of the process is complex [3] and it may not be possible to approximate it as a constant force process. Thus, in characterising the material’s flow behaviour the measurements should preferably be of true material properties, i.e. using constant strain rate or constant stress, rather than yielding results that are dependent on the measurement method, e.g. constant force or constant speed measurements. However, constant stress or constant strain rate measurements tend to be more difficult and expensive to make, and measurements that more closely mimic the process, for example those that are constant force or speed, may be considered to be more appropriate.

In conclusion, there is clear need to characterise the transient extensional flow behaviour of polymer melts if one is to obtain a full understanding of their behaviour, for example for modelling purposes. However, certain requirements, for example for quality control and materials comparison, may be met by qualitative ‘non-controlled’ measurement methods, rather than quantitatively accurate ‘controlled’ methods, and this may be the preferred route.

3.4 Advantages and disadvantages of stretching flow methods

A summary of the main advantages and disadvantages of stretching flow methods is presented. This represents, in part, a comparison with converging flow methods that can be used to generate qualitative data.

- Transient behaviour

  Stretching techniques can be used to characterise the transient extensional viscoelastic behaviour of polymer melts. In comparison, converging flow methods typically determine apparent equilibrium behaviour.

- True properties

  Stretching instruments can be developed to operate in either constant strain rate or constant stress modes thereby yielding true material properties rather than data that are dependent on the measurement method.

- Experimental difficulty

  The concept of tensile testing is simple. However, the implementation of the method is not so simple as experimental difficulties can be considerable. The analysis of the raw experimental data obtained using constant strain rate methods is however relatively straight forward. In comparison, non-controlled methods, e.g. converging flow, may be easier to implement and may be appropriate for quality control type applications but are more difficult, if not impossible, to interpret in terms of true materials properties data.

- Strain and strain rate range

  The maximum strain that is achievable need not be limited by the design of the instrument. Maximum achievable strain rates are typically up to $\approx 1\ s^{-1}$ although significantly higher values have been achieved using the NPL instrument. The strain rates obtained using tensile methods are not as high as can be achieved using converging flow methods, but the latter do not yield a strain dependence of properties.

- Closely mimics many processing methods

  It is preferable to use techniques that mimic the processes for which the data are being sought. In so doing the testing conditions more closely match the conditions experienced in processing, and thus the data are likely to be more appropriate. The stretching flow method is widely applicable in the polymer processing sector as many processes are based on stretching the melt.

In selecting the appropriate technique one obviously has to consider the above advantages and disadvantages in the light of the industrial need for the data. Where qualitative data are required then experimentally simpler techniques such as converging flow or fibre drawing [spinning] may be more suitable. However, where the need is for quantitatively accurate transient extensional stress growth data then one has to resort to 'controlled' methods, either constant strain rate or constant stress stretching methods. Constant strain rate measurements tend to be more commonly carried out than constant stress measurements. This reflects their relative ease of execution, that they are more appropriate for determining the transient nature of the flow, and that they are more representative of deformations occurring in processing (see Section 5.2).

This Guide specifically addresses constant strain rate methods ('controlled') although many issues addressed are relevant to other stretching methods.

4. Definitions

4.1 General

The following definitions are given by Whorlow [14] for strains and strain rates, and by the Nomenclature Committee of the Society of Rheology for start-up flow in tensile (simple) extension at constant (Hencky) strain rate $\dot{\varepsilon}$ [Dealy 22]. Additional detail is presented in Appendix A1. Further descriptions are given by, for example, Gupta *et al* [16] and Dealy [19, 23].

In describing and modelling plastics processing the Hencky strain is preferred as the rate of strain of an element of fluid within a flow is then independent of its original length and is determined only from the velocity field of that element. **Strain or strain rate is taken by default herein to imply Hencky values.**

4.2 Elongation ratio

Elongation ratio, ER (dimensionless): the ratio of the current length ℓ to the initial length ℓ_o of the specimen:

$$\mathrm{ER} = \ell / \ell_o \qquad (1)$$

4.3 Hencky strain

The Hencky strain ε (dimensionless) is also referred to as the natural or true strain. It is given by the natural logarithm of the elongation ratio: where ℓ is the specimen length (m) and ℓ_o is the original specimen length (m):

$$\varepsilon = \ln(\ell / \ell_o) \tag{2}$$

4.4 Hencky strain rates

Hencky strain rate $\dot{\varepsilon}$ (s^{-1}): is given by

$$\dot{\varepsilon} = 1/\ell \times \partial\ell/\partial t. \tag{3}$$

where t is time. It is independent of the original length ℓ_o.

4.5 Net tensile stress

The net tensile stress σ_E (Pa): in tensile (simple) extension it is defined by

$$\sigma_E = \sigma_{11} - \sigma_{22} = \sigma_{11} - \sigma_{33} = \sigma_{zz} - \sigma_{rr} \tag{4}$$

where σ_{ii} is a stress tensor in rectangular or axisymmetric coordinates.

4.6 Tensile stress growth coefficient

The tensile stress growth coefficient η_E^+ (Pa.s): the ratio of the net tensile stress to strain rate

$$\eta_E^+(t, \dot{\varepsilon}) = \sigma_E / \dot{\varepsilon} \tag{5}$$

where t is time, and + indicates start-up of flow. The above term is a transient term and is used to present the results of testing herein.

4.7 Tensile viscosity

The tensile viscosity η_E (Pa.s) is defined by

$$\eta_E(t,\dot{\varepsilon}) = \lim_{t\to\infty}[\eta_E^+(t,\dot{\varepsilon})] \tag{6}$$

It is the limiting tensile stress growth coefficient value and represents an equilibrium extensional viscosity if a steady value is achieved. However for materials that do not exhibit a steady state behaviour the use of an 'equilibrium extensional viscosity' such as this is obviously not appropriate.

5. Stretching flow methods

5.1 General

This Section discusses some of the more important considerations for stretching flow measurements of polymer melts based principally on a review of the literature [6]. It is presented as it provides the background information for understanding the important aspects of extensional testing, which may differ from one material to another, and thus the basis for good measurement practice.

The methods used for tensile testing of polymer melts are described (Section 5.2) and their limitations and the assumptions made in their use are discussed (Section 5.3). This is then followed by a more detailed consideration of the measurement issues (Section 5.4), specifically: preparation of test specimens (Section 5.4.1); methods used for clamping (Section 5.4.2); methods used to control the temperature of the test specimen (Section 5.4.3); the use and effect of the supporting medium (typically silicone oil) on the polymer (Sections 5.4.4 and 5.4.5); the control and/or measurement of strain, strain rate and stress (Sections 5.4.6 and 5.4.7); and the calibration of instruments (Section 5.4.8). The critical features of such instruments are then discussed in a broader context and concluding remarks are drawn (Section 5.4.9).

5.2 Description of stretching flow methods

The basic principle behind stretching flow measurements is to subject a specimen to a tensile stretching deformation, and through the measurement of the force and deformation the stresses and strains and hence strain rate can be determined. The restriction to 'controlled' methods implies that either the strain rate or stress is kept constant during the test. The use of haul-off or fibre spinning device on an extruder [11, 24, 25] are typically 'uncontrolled' measurements [13] and are not considered any further here. However, they are reviewed elsewhere [6].

Cogswell [20] commented that constant stress measurements are preferred if characterisation of the steady state behaviour is required as steady state conditions are more readily achieved than when using constant strain rate measurements. Conversely, the constant strain rate mode is more suited to providing information about the transient behaviour of the material. Axtell *et al* [26] similarly commented that in using the controlled stress mode of operation the strain rate and recoverable deformation attained their equilibrium values relatively quickly. However, Axtell *et al* [26] added that transient data from constant strain rate tests are often more representative of the deformations occurring in polymer processing and are therefore more desirable. It is clear that if constant strain rate methods are to be used, then strains greater than 4 are required to characterise fully the material including its steady state behaviour, if it exists.

The majority of methods reviewed [6] operate in a constant strain rate mode. In these instruments the specimen was stretched typically between either two rotating clamps [11, 27-30], or a single rotating clamp and a fixed clamp [31-34], or translating clamps (or a single translating clamp and a fixed clamp) [11, 24, 26, 35-39, 66], Figure 1. A translating clamp is one that moves in the direction of the axis of the specimen thereby stretching the specimen.

The following *Types* are defined for convenience to describe the various techniques, Figure 1.

Type A:	Single rotating clamp and a fixed clamp. (The rotating clamp consists of either a single or a pair of rotating elements, either smooth or textured. When used singly the clamp is fitted with a specimen locking mechanism.)
Type B:	Two rotating clamps. (Each clamp consists of either a single or pair of rotating elements. Clamps may be smooth or textured.)
Type C:	Ones moving (i.e. translating, non-rotating) and one fixed clamp.
Type D:	Two moving (i.e. translating, non-rotating) clamps.

In summarising the current state of measurement capability, extensional flow measurements at strains up to 7, strain rates up to 30 s^{-1} and temperatures up to 350 ^{o}C were reported although these conditions were not attained simultaneously [6]. The availability of equipment to operate at these conditions is very limited. More typically, the upper limits of testing were of strains up to ≈ 4, strain rates up to ≈ 1 s^{-1} and temperatures up to ≈ 200 °C but such equipment is still of limited availability.

5.3 Limitations in using stretching flow methods

The requirement for extensional measurements is for methods that provide true material properties data at testing conditions up to high values of strain, strain rate and temperature. The difficulties that this requirement pose and the limitations of current methods are now addressed.

Dealy [19] commented that limitations on the maximum strain, end-effects, non-uniformities in temperature and deformation, and the measurement of very small forces are the key experimental problems facing the measurement of extensional properties. Similarly, Gupta *et al* [16] commented that the major difficulties in using instruments for constant strain rate measurements are due to temperature and strain rate uniformity and also gravitational effects (for low viscosity melts). The maximum strain issue can be easily overcome by instrument design (i.e. by use of types A and B, Figure 1), and the end-effects can be easily assessed. However, the other factors remain key difficulties in reliable measurement.

The application of stretching techniques to the characterisation of materials is limited by the viscosity of those materials. The lower limit of viscosity is due to the resolution of the force measuring device, instrument friction and inertia, and the effects on the specimen of its inertia, fluid drag, gravity and surface tension. Cogswell [35] commented that the constant stress method is suited to melts with viscosities greater than 10^5 Pa.s. Rhi-Sausi *et al* [40] reported that improvements to this type of instrument were made but did not quantify the magnitude of those improvements. Gupta *et al* [16] commented that constant stretch rate methods are limited to polymers having shear viscosities at the test temperature in excess of approximately 10^4 Pa.s [16]. Petrie [18] commented that in making constant stress measurements of melts having viscosities less than 10^4 Pa.s the difficulties associated with instrument friction and fluid drag become significant [19]. Also, the support of the specimen against the effect of gravity is more critical. Gupta *et al* [16] commented that such techniques may not be suitable for use with nylons and polyesters that may have shear viscosities as low as 100 Pa.s. Furthermore they [16] commented that other non-controllable techniques, for example spinning or converging flow analysis may be more suitable for such lower viscosity materials. In using the instrument by Meissner *et al* [27] extensional viscosities as low as 3×10^3 Pa.s were reported for HDPE at 150 °C, 8×10^3 Pa.s for nylons at 250 °C and 7×10^3 Pa.s for poly(ethersulfone) at 350 °C. The instrument used by Meissner *et al* [27] was a Type B instrument in which the specimens were supported on a nitrogen cushion and tank-tracks, rather than circular clamps, were used to impose the strain. Special fittings were available for testing low viscosity materials. Thus the lower limit for testing using existing stretching techniques was as low as 3×10^3 Pa.s although more commonly the value is of the order of at least 10^4 Pa.s.

The most fundamental difference between Types A and B and Types C and D is that the maximum strains achievable using Types C and D instruments are limited by the physical sizes of the instrument and the original specimen length [19]. For Types A and B the gauge length is constant and the maximum strain is unlimited. Furthermore, the strain rate is proportional to the take-up speed of the clamp(s), whereas for Type C and D instruments clamps of exponentially increasing velocity are required to produce constant strain rates in the specimen. This also has consequences for other aspects of the measurement capability of the instrument, in particular the maximum strain rates that can be achieved and also the level of temperature uniformity of the specimen.

Cogswell [20] commented that translating clamp techniques (Types C and D) are limited by the design of the instrument, typically to maximum strains of approximately 2. However, since then strains up to a value of 4 have been achieved using such methods [30, 41]. Higher strain values cannot simply be obtained using Type C and D devices by employing shorter initial specimen lengths as end-effects may become more significant. This factor is addressed in detail in Sections 5.4.2 and 7.2. Furthermore, if the length to diameter aspect ratio of the specimen is kept the same then the magnitude of the tensile force supported by the specimen will be reduced for shorter specimens (proportional to the cross-sectional area of the specimen) thereby increasing errors in the measurement. To obtain higher strain values, up to a maximum value of 7, Type A and B instruments with rotating clamps have been successfully used [10, 27, 30,42, 43].

The limitation in the maximum strain imposed by equipment design has important implications in terms of characterising the equilibrium behaviour of the materials, particularly when using controlled strain rate instruments. Petrie [18], commenting on the work by Laun *et al* [10], stated that the equilibrium extensional viscosity value was reached at strains greater than 4 or if the time of elongation exceeded the greatest relaxation time of the material. However, Meissner *et al* [42] observed that the specimens did not attain steady state or equilibrium conditions even up to strains of 7 in controlled strain measurements. A maximum in the tensile stress was observed for LDPE at a strain of approximately 5 to 6, beyond which the stress decreased. Münstedt *et al* [30] commented that for constant strain rate experiments a broad maximum was observed in stress values indicating that there was no steady state behaviour in elongation. However, they commented that the fall in values after the maximum may be due to inhomogeneous deformation of the specimen at the very large strains.

At large strains the accuracy of measurement of the force supported by the specimen will also be a limiting factor in the determination of extensional flow properties. The limit of any measurement will be a combination of the force resolution of the instrument combined with the extensional stress growth behaviour of the material. For materials that exhibit little strain hardening the errors will be greater at relatively lower strains than for those materials that exhibit a large strain hardening.

5.4 Measurement considerations

The following Sections contain detailed discussions of various aspects of extensional properties measurements, which are summarised in Section 5.4.9. It is suggested that the reader first consults the summary and then visits each of the Sections 5.4.1 to 5.4.7 as necessary. Section 5.4.8 discusses the important issue of calibration and checking of the instrument. These Sections provided the background to establishing good measurement practice (Section 6).

5.4.1 Specimen preparation

It is accepted that the preparation of specimens is of critical importance for extensional viscoelasticity measurements [8]. Problems related to specimen preparation include: non-uniformity of the specimen dimensions, degradation of the material during specimen preparation, stress relaxation of the specimen (due to internal stresses locked in the specimen during its preparation) prior to and during testing, and impurities including trapped air causing stress concentrations and possible degradation. The criticality of these factors is dependent, however, on the material and its extensional flow behaviour.

Methods for preparing specimens were principally extrusion [9-11, 26, 28, 31, 32, 34, 37, 44-48] or transfer moulding that was predominantly carried out under vacuum [38, 49-53, 66]. Injection moulding [24, 39], compression moulding [27,54] and machining from larger samples [26, 55] were also used. The references listed here are indicative rather than exhaustive as details presented in the references were often insufficient. A large number of the papers reported annealing of the specimen prior to testing to remove residual stresses. However, Vinogradov [56] reported that annealing PS specimens made by extruding from a capillary rheometer into water had no effect on their stress growth behaviour. Ballman [55] used birefringence to identify whether specimens were free of stress.

Hingmann *et al* [37], for example, commented that specimen preparation was critical to extensional measurements: a perfectly cylindrical shape is required for homogenous uniaxial stretching. They prepared specimens by extruding melt into an oil bath, but reported that voids in the specimen can cause premature failure. They commented that voids can be minimised by controlling extrusion temperature and cooling rate as voidage is related to crystallisation. They also commented that although voidage problems could be avoided by using injection moulded specimens they are not suitable as they contain highly orientated layers. These layers will relax on re-melting and may cause excessive distortion of the specimen.

Schlund *et al* [50] indicated that specimen preparation, specifically the degree of chain entanglement, can have a significant effect on the measured properties: the moulding time (transfer moulding under vacuum and then annealed) affected their stress growth behaviour. Similarly, Laun *et al* [11] identified that for melt spinning experiments there was a significant effect of the geometry of the contraction region, and hence of the orientation of the extrudate on the measured extensional properties of LDPE. Thus molecular orientation clearly has an effect on the stress growth behaviour but is not expected to affect equilibrium extensional viscosity values (if they exist) as they should be independent of the initial orientation.

On the subject of impurities in specimens, for example, Meissner [8] reported that dissolved air in PS will result in the formation of bubbles in the specimens on re-melting. These would then cause premature failure of the specimens due to their stress concentration effect. The specimens were therefore produced under vacuum [8]. Vacuum was used in conjunction with transfer moulding by a number of other researchers to prepare specimens.

For methods of specimen preparation that do not cause significant flow to the polymer, e.g. compression moulding, there is the risk that, when using granules, the structure of the granules is not “destroyed” in the process and that premature failure will occur in the specimen at the site of a granule boundary.

The homogeneity of deformation that is necessary, particularly for obtaining data at high strains, is critically dependent on specimen homogeneity. Meissner *et al* [42] commented that further increases in the strain to failure above a value of 7 are probably limited by inhomogeneity of the specimen. It is noted, however, that this strain is significantly outside the range encountered in most industrial practices.

Meissner *et al* [42] commented that a disadvantage of Type A compared with Type C and D instruments is that larger specimen sizes are typically required. In testing materials that are under development this may be an important consideration. However, the difference in specimen size may not be significant compared with the sample size required for capillary extrusion measurements to determine shear viscosity. It is also noted that larger specimens sizes are potentially more susceptible to impurities and non-uniformity.

5.4.2 Clamping

5.4.2.1 General

Clamping the specimens is obviously essential but presents problems due to the relatively low viscosity of the material that is being clamped (the term ‘clamped’ is used here to indicate any method by which a force can be transmitted to the specimen). Two main approaches are used, either fixing the specimen to the translating or rotating clamp using either an adhesive or mechanical means (termed herein as ‘fixed clamp’, e.g. Type A, C or D), or by drawing the specimen between two counter-rotating clamps that may be textured to improve the friction between the specimen and the rotors (Type B). The use of either approach has advantages and disadvantages.

5.4.2.2 Translating or single rotating clamps (Types A, C and D)

In using fixed clamps (e.g. Type A, C and D) the specimen is either mechanically clamped or stuck to the clamp using adhesive. The use of mechanical clamping may require that the clamp is cooled to avoid necking due to stress concentrations near the clamp [35]. However, this has the disadvantage that it may also affect the temperature uniformity of the specimen that is important for uniformity of the deformation of the specimen, see Section 5.4.3. The alternative and more commonly adopted approach is to use an adhesive to stick etched specimens to the clamps [for example reference 9]. This approach avoids the need to use clamp cooling. Dealy [23] commented that the use of adhesives seems to produce minimum end-effects and that epoxy adhesives appear to be suitable for polyolefins [23]. Alternative approaches include the use of clamps similar to artist's pencil grips (i.e. collet-type clamps) as used by Vinogradov *et al* [56], self-locking clamps [57, 58], and also the use of a hoop-shaped specimen mounted over two PTFE posts so that gripping of the specimen is not required [26]. The latter method would however generate stress concentrations at the ends of the specimen that would result in non-uniformity of deformation. For higher viscosity materials traditional clamping methods for solids were also used [59-61].

Meissner [62] commented that Type C and D instruments have the advantage that very small specimens, stuck to clamps using adhesive, can be used but the deformation is inhomogeneous and the maximum strain is limited. The use of fixed clamps (Type C and D) introduces end-errors [10] that are more significant for specimens of small length to diameter aspect ratio. It is also noted that the non-uniformity of the cross-sectional area of the specimen is considered to be a major limiting factor in obtaining high strains and that larger specimens are more likely to be non-uniform, particularly if produced by extrusion.

The effect of end-errors when using fixed clamps, i.e. the inhomogeneous deformation due to the constraint of the end-plates, was studied both experimentally and theoretically for polyisobutylene solutions by Spiegelberg *et al* [74]. They concluded that the end-effects are apparent for Newtonian liquids for strains up to 2, and for viscoelastic fluids up to strains of 4. Vinogradov *et al* [56] observed that for PS specimens of length to diameter ratio greater than approximately 10 there was no effect of the aspect ratio on the measured stress growth behaviour. However, there was a significant effect when shorter specimens were used. In comparison Cogswell [20] stated that for a length to diameter ratio of at least 5 the end-effects will be comparable to the scatter in results, thus contradicting the observed results of Vinogradov et al [56]. A ratio for the length to diameter of the specimen of at least 10 is thus preferred.

Everage et al [60] used a modified Rheometrics Mechanical Spectrometer (rotational rheometer) using its off-set drive and torque measuring system to perform extensional measurements. This approach is appealing as it employs an existing instrument with limited modification for extensional testing. Forced air heating was used as slumping of the specimen was not found to be a problem, thus restricting it high viscosity materials, although a silicone oil bath was reported to be available. However, a major difficulty with this instrument was that a maximum specimen length of approximately 20 mm only could be used, which could result in significant end-effect errors. Dealy [19], referring to Garfield *et al* [63], reported that the end-effects resulted in non-uniform strain rates, particularly at high strains and for low viscosity materials (see also Connelly *et al* [61]). Connelly *et al* [61] related the degree of non-uniformity of the stretching process to the Weissenberg number characterising the flow. Petrie [18] commented that according to Pearson (personal communication referenced in [18]) the non-uniformity of the stretching of PS was most significant at low strain rates and high temperatures.

Also see Section 5.4.6 for discussion on the uniformity of deformation of specimens.

5.4.2.3 Counter-rotating clamps (Type B)

The use of a counter-rotating clamp systems (for example Meissner [7]) has the advantages that constant Hencky strain rates are achieved using constant rotational speeds of the rotors, the total elongation of the specimen does not depend on the length of the rheometer, and the clamps may be used for polymer melts of relatively low viscosity. The use of counter-rotating clamps also avoids any necking of the polymer or end-effects as polymer is not allowed to build up in the vicinity of the clamps. The deformation of the specimen is therefore homogenous along the specimen length [20, 42]. However, the use of heavy profiling of the counter-rotating clamps may result in differences between ‘circumferential’ and take-up speed due to yielding of the specimen between the clamps (i.e. calendering). In one case the effect was reported to be up to 10% [42]. The counter-rotating clamp arrangement of Meissner [7] was further developed [27] in which two pairs of tank tracks are used to stretch the specimen.

Laun *et al* [43] commented that difficulties in using two pairs of counter-rotating clamps, due to the need to both drive and measure the tension through at least one of the pairs, can be avoided by using a single pair of counter-rotating clamps and a fixed clamp. Thus this arrangement separates the drive and force measuring components of the instrument. The experimental problems associated with the use of two pairs of counter-rotating clamps is particularly great at low strain rates ($<10^{-4}$ s^{-1}), i.e. low tensile forces.

Münstedt *et al* [30] made steady state measurements of extensional viscosity of LDPE and HDPE using both Type B, C and D instruments and observed that there was good agreement of results between the methods. This indicates that the magnitude of the effects due to the difficulties described above in using either fixed or counter-rotating clamps can at best be negligible.

5.4.3 Temperature control

With the exception of four papers, all the work referenced herein using ‘controllable’ methods were at temperatures equal to or less than 200 ^{o}C. The four exceptions to this were for testing up to: 250 ^{o}C for PA and 350 ^{o}C for PES [43], 210 ^{o}C for PP [36], 220 ^{o}C for LLDPE and HDPE/LDPE blends [53], and 220 ^{o}C for PP [31]. All of these papers reported the use of silicone oils as mediums except for Meissner *et al* [27] in which a cushion of nitrogen was used to support the specimen. With reference to the use of oils, Muller [57] commented that the oil must have relatively low viscosity at the test temperature to avoid fluid resistance to the movement of the clamps and viscous drag on the specimen.

Muller [57] commented that the viscosity of polymers in the molten state (particularly for amorphous polymers near T_g) is particularly sensitive to temperature variations. This is particularly the case in making extensional measurements as, compared with injection moulding, processing temperatures are typically lower and testing is therefore carried out nearer the solid-melt transition temperature range over which the temperature sensitivity may be particularly high. Good temperature control is therefore necessary to ensure homogeneous deformation over the length of the specimen: localised hot-spots will result in excessive localised deformation of the specimen and eventual failure at that position with consequent errors in the measured values for the specimen. The magnitude of the effect of temperature non-uniformity on the measured behaviour will be dependent on the temperature dependence of the material and also its strain hardening characteristics.

The importance of good temperature control has been acknowledged by various researchers. For example, to achieve good temperature control Muller *et al* [57] used a double oil bath arrangement. The clamp in the bath was also separately heated to control effects due to heat loss from the bath. The authors claimed to obtain temperature control to within ± 0.2 °C. In using this vertical arrangement with an unstirred inner bath there may be problems due to convection within the inner bath causing a temperature gradient along the length of the specimen. Sebastian *et al* [64] also used a double-layer heating arrangement. Axtell *et al* [26] commented that the use of an oil bath has the advantage over hot-air in that better stability is obtainable.

Meissner *et al* [42] commented that temperature uniformity is essential, particularly for determining the failure behaviour, as it influences the homogeneity of the deformation of the specimen. They considered it to be a critical controlling parameter in testing that limits the maximum strains that can be achieved. In earlier work a glass cover was used to improve temperature uniformity within the oil bath. In subsequent developments a copper trough and cover were used to further improve the temperature uniformity. The motor was also placed in the silicone oil to further improve on the temperature homogeneity that was reported to be better than ± 0.1 °C at 150 °C [42]. With these improvements strains of up to 7 were obtained. Meissner *et al* [42] commented that further increases in the strain to failure (> 7) were probably limited due to inhomogeneity in the specimen dimensions.

As with all rheological testing, adiabatic heating - that is heat generated due to working the specimen - can be significant. This is particularly the case with capillary extrusion rheometry [65]. Thus in melt spinning the effect of viscous dissipation in the extruder on temperature control may potentially be significant. However, as discussed earlier this technique can only be assumed to provide qualitative rather than quantitative values. Whorlow [14] commented that for stretching techniques uniaxial extensional measurements are limited to strain rates of approximately 10 s^{-1} due to adiabatic heating. Laun *et al* [43] estimated that for a LDPE the adiabatic temperature increase at a strain rate of 3 s^{-1} and for strains less than 4 would be smaller than 1 °C. They concluded that this would have a negligible effect on measurements. However this assumes that the deformation and thus the adiabatic heating is homogeneous. Where this is not the case the excessive localised deformation will generate a greater heating effect thus resulting in even greater localised deformation and likely specimen failure.

5.4.4 Specimen support

Apart from providing a controlled temperature environment the test medium must also provide sufficient support for the specimens otherwise the specimens will sag under their own weight. Silicone oil was used almost exclusively with the exception of when testing was carried out in air [e.g. 24, 60, 61, 65] or using a nitrogen cushion [27]. Specimens were either mounted vertically in the oil [e.g. 9, 49] or horizontally either submerged in or on the surface of the oil [e.g. 7, 10]. Utracki *et al* [38] commented that for low viscosity melts it is important to minimise buoyancy or gravitational effects by matching the densities of the polymer and the medium. They commented that for materials of viscosity higher than

10^7 Pa.s this is not a problem. However, for thermoplastics at typical processing temperatures it cannot be ignored and the use of an oil bath or other mechanism to support the specimen is required [19] (Cogswell commented when the viscosity < 10^6 Pa.s [20]). When specimens are mounted vertically or horizontally in the oil it is necessary to match the density of the oil with that of the specimen (both at the test temperature) so that deformation of the specimen prior to and during the test due to gravitational or buoyancy forces is minimal. When the specimen is floated on the surface of the silicone oil it is not necessary to match exactly the densities but the oil must be at least as dense as the specimen. However, floating the specimen on the oil may produce additional problems with temperature control.

5.4.5 Effect of the supporting medium on the specimen properties

A number of researchers using silicone oil as a test medium have investigated the effect of the oil on the properties of the polymer under investigation.

Hingmann [37] reported on the absorption of oil by the specimens, determined by weight measurements, during extensional testing. For PP it was found to increase by approximately 2% with the absorption being in a surface layer of ≈ 300 μm thickness. However, they reported that there was no effect on measured properties. They also identified that 6% oil content had no effect on flow properties [37].

Lanfray *et al* [36] commented that for PP and PS the specimens showed no evidence of swelling or change during the time of the experiments, as observed through low strain rate (Trouton ratio) measurements and also dynamic shear measurements.

In testing of glass fibre filled PP, Kamal *et al* [66] subjected the specimens to severe regimes of 24 hours at 200 °C in oil and measured a weight loss of less than 2% in Dow-1265 but a 2% increase in 15 minutes in Dow 210H. The latter exhibited swelling which was then followed by a weight loss indicating degradation of the polymer. The different silicone oils were used to match the different sample densities. However the authors commented, on the basis of the ratio of extensional viscosity and zero shear viscosity results and also on the superposition of stress growth results, that there was no appreciable effect of the oil on extensional viscosities.

Münstedt [9] reported that for PS after 2 hours exposure at 160 °C no silicone oil penetration was identified using IR spectroscopy. Furthermore there was no indication of oil penetration from zero shear viscosity measurements that are reportedly sensitive to penetration of low molecular components into the melt and thermal degradation. Similarly for LLDPE and LDPE, Schlund *et al* [50] concluded that silicone oil had no effect on results.

Thus although silicone oil was reported to be absorbed by PP, there was no reported effect on properties. However, there appears to be no comment on the failure behaviour of the polymers. Schlund *et al* [50], referring to work of Kurbanaliev [67] and Utracki [68], commented that the medium used in testing could have a significant effect on the ultimate properties of the melt in extension, specifically the maximum stress and strain at break for 1,2-polybutadiene and HDPE, LDPE and LLDPE. However the various mediums used by Kurbanaliev [67] were alcohols and the findings may not apply to the use of silicone oils. In the work presented by Schlund *et al* [50] it was shown that there was no effect within 30 minutes at 150 °C for LLDPE. This does highlight, however, that it is important to assess the effect of the medium on properties at failure in addition to the low strain behaviour.

Utracki *et al* [53] commented that testing at low strain rates ($\dot{\varepsilon} < 0.01$) may not be limited by the true melt strength of the specimen but by the test duration and thermal stability of the sample and degradation effects of the silicone oil (the duration of thermal stability was identified by dynamic viscosity measurements). At such low rates testing times increase dramatically, for example to test at a strain rate of 0.01 s^{-1} to a strain of 6 will take 10 minutes. Apart from degradation effects Laun [43] observed differences in behaviour of normal and stabilised LDPE indicating that cross-linking of the normal LDPE was occurring at 150 °C.

Meissner [62] commented that future developments of the uniaxial and multiaxial extensional rheometers should include eliminating the use of a supporting liquid and increasing the maximum test temperature. The approach by Meissner *et al* [27] in using a nitrogen cushion has the benefit that higher test temperatures, not limited by the thermal stability of the test medium (usually silicone oil), can be obtained and also that materials affected by silicone oils can be tested. However the potential for degradation effects and further cross-linking must still be assessed. A considerable benefit in using an oil bath is that heating times are small due to efficient heat transfer from the oil to the specimen.

5.4.6 Strain and strain rate

The predominant problem with Type C and D instruments is that the strain is limited by the length of the instrument, and to obtain a constant strain rate the clamp separation speed needs to be increased exponentially [57]. Rhi-Sausi *et al* [40] commented that strain rates above 0.1 s^{-1} are difficult to achieve using Type C and D instruments with screw-thread drive as high clamp speeds are required. However, values as high as 1 s^{-1} were obtained by Schlund *et al* [50] using a rotary motor as a winding device for a cable connected to the clamp, and 2 s^{-1} by Lanfray *et al* [36] and also by Lacaze *et al* [69] using twin screw-driven clamps. Typically the largest strains and strain rates obtained using Type C and D instruments were approximately 4 and 2 s^{-1} respectively. In comparison, using Type A and B instruments strains up to 7 were achieved by both Meissner *et al* [42] and Raible *et al* [29], and strain rates of up to 30 s^{-1} were achieved by Münstedt *et al* [30], 10 s^{-1} by Laun [43] and 1.5 s^{-1} by

Ide *et al* [45]. Measurements carried out by Münstedt *et al* [30] at 30 s^{-1} were made in approximately 0.1 seconds.

The use of a cam loading system to generate controlled stress tests limits the maximum strains that can be achieved (the cam being profiled to reduce the force linearly with the reduction in cross-sectional area of the specimen as it stretches). Larger strains require larger cams that in turn results in a large inertia component [10] and increased frictional problems thus increasing errors in the determination of both the stress and strain rate values. These factors become more significant at higher strains as the tensile force supported by the specimen decreases. These problems can be partially overcome by using two cams in series [10]. This system was shown to produce stress values deviating by no more than 5% over the range of deformations up to a strain of approximately 3.5 [10]. Various cam designs are described by Whorlow [14]. An additional practical problem associated with the use of cams is that they are designed for specimens of a specific length. Additional sources of error were investigated by various researchers. In constant strain rate testing, inhomogeneity of deformation due to the various reasons described previously may result in the actual strain rates and therefore strains differing from the set values. Correction to the strain rates of the order of 2% to 3% on the basis of optical measurements were made by Tanaka *et al* [44]. Gupta *et al* [16] commented that the specimen needs to be accelerated from rest to a finite velocity instantaneously at the start of the test if the deformation is to be truly constant strain rate. However, due to the inertia of the instrument [16, 19] and the response time of the control system this will not be the case. Consequently, for controlled strain rate measurements at short times and at high rates the strain rate is unlikely to be constant.

Ide *et al* [45] reported that they had made density-temperature corrections to the cross-sectional area of the specimens to obtain the true specimen diameters at the test temperature. It is important to know the initial diameter of the specimen at the test temperature. The magnitude of the correction made by Ide *et al* [45] ranged from 16% for HDPE and PP, 12% for LDPE and 5% for PS and PMMA.

Laun [70] briefly discussed the effect of instrument compliance on recoverable strain measurements indicating that the compliance will result in the strain recoil being overestimated, and the effect of machine inertia/control will result in underestimation of the recoverable strain in stress relaxation. Instrument compliance may also affect the measurement of strain and therefore strain rate, in particular when testing at high forces. Consideration should be given to ensuring that the compliance of the instrument (primarily the displacement of the force measuring device, on application of a force, affecting the effective specimen length) is negligible in comparison with the length of the specimen. This can effectively be checked by monitoring of the accuracy of deformation during the test by optical means (see below and Section 6.2.4).

The uniformity of deformation can be assessed, as carried out by Meissner [8], by cutting the stretched specimen into several sections at the end of the test. The homogeneity of the deformation can then be determined by measuring the weight of each section. Optical methods have also been employed to check on the uniformity of deformation [e.g. 32, 44, 61, 71].

See also Section 5.4.2 on clamping and its effect on uniformity of deformation.

5.4.7 Stress

The tensile force in a specimen undergoing extension will be a function of the specimen's initial cross sectional area, strain, strain rate, the material and its extensional stress growth characteristics (including temperature dependence). The consequence of this is that a wide range of forces will be encountered in measurements. At high strains, e.g. $\varepsilon = 6$ corresponding to an elongational ratio of ≈ 400, the force becomes very small due to the reduction in cross sectional area of the specimen [10]. The accuracy of the measurement of extensional properties is then limited by the resolution and stability of the force measuring device (referred to below as ‘transducer’) and also by the friction of the instrument and other factors discussed below. The decrease in force makes force transducer selection difficult but critically important to the accurate measurement of properties, particularly at high strains [19, 72]. Force transducers need to be of the appropriate range and also have good resolution and stability.

Instruments reviewed elsewhere [6] had maximum force ranges up to 10 N [9] although measurements of the order of 0.03 N peak force were identified for LDPE at $\dot{\varepsilon} = 0.005\ \mathrm{s^{-1}}$ and 150 $^{\circ}$C [42], and 0.002 N for LDPE at $\dot{\varepsilon} = 0.01\ \mathrm{s^{-1}}$ and 160 $^{\circ}$C [45]. Quoted values of the resolution of stretching instruments used for testing polymer melts were of the order of 10^{-3} N [9, 27] and 10^{-4} N [43]. Tensile forces were measured either using leaf springs combined with displacement transducers [e.g. 8, 27, 43] or strain gauges [e.g. 64], load cells [e.g. 9, 45], or the force was applied using cam loading systems [e.g. 10, 35, 73]. Sebastian *et al* [64] commented that immersion of the strain gauges in the oil bath resulted in significantly improved voltage stability in comparison with mounting in ambient.

The effect of errors on measurements due to instrument friction, interfacial tension, viscous or frictional drag and inertia of the specimen were investigated by various researchers. Friction of bearings can limit the lower range of instruments to materials with viscosities greater than approximately 10^4 Pa.s [73] or 10^5 Pa.s [19, 20]. Ishizuka *et al* [31] commented that frictional forces can become significant at high strains (estimated at ≈ 10% for $\varepsilon = 4$, $\dot{\varepsilon} = 0.6\ \mathrm{s^{-1}}$) as the tensile force on the specimen decreases considerably with increased strain.

Laun *et al* [43] showed, using a combined experimental and theoretical approach, that interfacial tension effects on tensile stress measurements of LDPE at 150 $^{\circ}$C (using silicone oil) were negligible at strain rates above $10^{-4}\ \mathrm{s^{-1}}$, but were dominant for strain rates less than

10^{-5} s^{-1}. A method by which a correction for interfacial effects can be made was presented. Interfacial tension will be more significant when using translating clamp (Type C and D) instruments for which relative speeds between the specimen and the oil will be greater.

Ide *et al* [45] estimated that the effect of fluid drag on the tensile force on the specimen was of the order of 2 - 3% for a low viscosity polymer and a silicone oil of 0.01 Pa.s. This effect was therefore ignored in their work. Ishizuka *et al* [31] indicated how the effect of drag on measurements can be taken into account.

Rhi-Sausi [40] presented evidence to indicate that inertia arising from acceleration of the specimen was not expected to be significant in their experiments that used a moving clamp Type C instrument. Ishizuka *et al* [31] concluded that the effect of inertia for their Type A instrument was of the order of 0.02% and was therefore negligible.

5.4.8 Calibration

Each component of the measurement, e.g. force, displacement and temperature, must be calibrated separately. The major problem in checking the calibration of an extensional rheometer is that there are no calibrants or reference materials that could be used, except possible limited sources established through intercomparisons [e.g. 77].

Particular problems associated with the use of such instruments will be the measurement of the specimen's dimensions at the test temperature at the start of the test, checking the uniformity of deformation, and the accuracy at low levels of force measurement encountered at low and high strain values. Dealy [19] discussed some of the problems associated with calibration of extensional rheometers. As additional means of checking the instrument, he commented that the minimum requirement should be to check that the extensional viscosity at very low extension rates was three times that of the zero shear viscosity (limiting shear viscosity at zero shear rate). This is known as the Trouton ratio. However, this does not check the instrument in the more industrially relevant high strain rate range. Dealy [19] commented that a better verification is to compare results obtained using different instruments, although this in itself is problematic [6, 75, also see Section 7.5].

Given the difficulty of such measurements it is essential to calibrate the individual components of the instrument and to perform, where possible, additional checks such as using a reference material or comparison with other instruments.

5.4.9 Summary

A comparison of the methods presented in the literature would indicate that the use of at least one rotating clamp is a desirable feature of an extensional rheometer. This avoids the restriction placed on measurements of the strain being fixed by the instrument's size rather than by the material's response - i.e. its failure. This artificial limitation on strain would most likely also limit the maximum strain rate that could be achieved using the instrument.

On the basis of a comparison of measurements using two different instruments the magnitude of the effect of end-errors introduced due to using a non-rotating clamp compared with rotating clamps was found to be negligible [30]. However, the effect of end-errors on the uniformity of the deformation was also observed to be potentially very significant [60]. Measurements made using different length to diameter ratios for the test specimens indicated that the length of the specimen should be at least 10 times its diameter [56].

Silicone oil was the most commonly used test medium. It provides both buoyancy for the specimen thus reducing any sag due to gravity and also acts as a heat transfer medium. The effect of silicone oil on the specimens was reported in several papers and although absorption was identified there was no observed effect on the measured properties. However the effect of the oil on the properties at failure was not reported, except for polyisobutylene in alcohols for which there was a significant effect [67]. The effect of silicone oil absorption on ultimate tensile properties of polymer melts requires further investigation.

The uniformity of temperature is an important factor in obtaining results at high strains. Considerable effort has been made to minimise temperature variations, for example the use of a copper trough [42] or double-skinned silicone oil baths [56, 58]. The use of a vertical test set-up and a silicone oil bath may result in the development of an undesirable temperature gradient due to density variations of the oil. Improved temperature homogeneity would be obtained for example by using a shorter oil bath. However in reducing the specimen length to improve on temperature uniformity the effect of any end-errors on the homogeneity of deformation would become more significant. In order to maintain an initial length to diameter ratio for the specimens of at least 10, specimens of small initial diameter could be used. Shorter specimens that are homogeneous may be easier to produce but they will also generate smaller forces (proportional to cross-sectional area) that may result in increased measurement errors.

The selection or design of the force measuring device is clearly of considerable importance, particularly for measurements at high strains at which the cross-sectional area of the specimen will be considerably smaller than it was at the start of the test. Good resolution and high accuracy at low values are important for accurate measurements at high strains.

To check the uniformity of deformation of the specimen and thus the quality of the test two methods have been used. The weight of several samples cut from the deformed specimen have been compared or the specimen's dimensions have been measured optically during the test. The latter obviously requires the use of a transparent window, that in itself might affect the temperature homogeneity of the specimen. However, the difficulties associated with measuring the diameter of a filament of molten polymer in an oil bath, both of which may well be transparent, may also result in unacceptable measurement errors.

The magnitude of the effect of various errors, for example due to frictional drag of the supporting medium were discussed (Section 5.4). Methods for correcting for the effect of these errors were referenced.

In summarising the current state of extensional measurement capability, flow measurements at strains up to 7, strain rates up to 30 s^{-1} and temperatures up to 350 ^{o}C have been reported although these conditions were not attained simultaneously. The availability of equipment to operate at these conditions is very limited. More typically, the upper limits of testing were of strains up to $\approx$ 4, strain rates up to $\approx$ 1 s^{-1} and temperatures up to $\approx$ 200 ^{o}C but such equipment is still of limited availability.

In conclusion, the more important aspects of instruments for measuring extensional viscoelastic properties of polymer melts are summarised as follows:

- Testing should be carried out using either constant strain rate or constant stress deformations in order to generate quantitative data that are independent of the test method (Section 3).

- The use of at least one rotating clamp overcomes the restriction that the maximum strain is limited by the length of the test equipment (Section 5.4.2).

- When using non-rotating clamps the effect of end-errors on the uniformity of deformation can be significant. The specimen length to diameter ratio should be at least 10 (Section 5.4.2).

- The attachment of etched specimens to clamps using adhesives (typically epoxy) has been found to be generally suitable (Section 5.4.2).

- Silicone oils have been commonly used and have been found to be suitable for use to support specimens and to act as heat transfer media, with no effect on extensional flow properties reported. However, there was no evidence in the literature of the effect of silicone oil on the specimen’s properties at failure. A forced gas heating systems has also been found to be suitable, but the use of a silicone oil bath may permit more rapid heating of the specimen (Sections 5.4.4 and 5.4.5).

- The homogeneity of specimens, in terms of their dimensions and also purity, affects the uniformity of their deformation. It is essential that this factor is controlled in order to obtain reliable data (Section 5.4.1).

- Temperature uniformity is essential for uniform deformation of the test specimen. It is essential that this factor is controlled in order to obtain reliable data (Section 5.4.3).

- The accurate measurement of force over a wide range is critical to the reliable determination of extensional properties over a wide range strains, in particular at very low and very high strains (Section 5.4.7).

- It is desirable to check the uniformity of deformation of specimens to assess the quality of each test (Section 5.4.6).

The relative importance of these factors and others discussed elsewhere in Section 5 is likely to differ from one material to another so general guidance is not possible. However, the uniformity of the specimen and test temperature are recognised as being critical to good measurements, but this should not undermine the importance of the other test parameters (e.g. force and strain rate) and their effect on the quality of the results. Quantification of some of these factors, i.e. the permitted tolerances, is provided in the latest version of the standard ISO/CD 20965 Plastics – Determination of the transient extensional viscosity of polymer melts, to which the reader is referred.

6. Measurement of extensional flow properties

6.1 General

Section 6 is aimed at illustrating various aspects of the measurement of the extensional properties of polymers using stretching methods, with reference to the NPL instrument and results obtained with it. It describes:

- the instrument (Section 6.2) and outlines the procedures adopted for using it to measure the extensional viscoelasticity of molten polymers (Sections 6.4 and 6.5),

- the analysis for interpreting the experimental data (Section 6.3), and

An assessment of the effect of uncertainties experimental data on the derived extensional viscosity values is presented in Section 8.

Many of the issues and analyses for this type of instrument are valid for the other controlled stain rate and controlled stress rheometers (Figure 1).

Use of this Guide must be in conjunction with the latest version of the Standard ISO/CD 20965 Plastics – Determination of the transient extensional viscosity of polymer melts.

6.2 Example extensional rheometer

6.2.1 General

An instrument for measuring the extensional viscoelasticity of polymer melts over a wide range of strain rates, developed by NPL, is shown in Figure 2. The operating principle of the instrument is to stretch a molten specimen between a rotating clamp and a fixed clamp (Type A instrument, Figure 1).

In this instrument the specimen deformation is achieved by the use of a rotating clamp that, by winding up the specimen, controlled the rate at which the specimen was stretched. The clamp's rotational position, and hence speed is controlled using a servo motor. In using a rotating clamp, measurements at constant strain rate can be achieved simply by using a constant rotation speed. The measurements are therefore 'controlled' [13]. Furthermore, the use of a rotating clamp results in an instrument that is not limited in its maximum strain capability. For an instrument that uses translating clamps the maximum strain that can be achieved is limited by the dimensions of the instrument. The rotating drum is raised during the test to prevent overlap of the specimen on winding more than once around the drum. The drum was roughened to prevent slip of the specimen occurring at the surface.

The fixed clamp is used to measure the tensile force developed in the specimen during stretching. This clamp is mounted on a leaf spring, the displacement of which is measured using a non-contacting optical transducer. The force is then calculated from the force - displacement characteristics of the leaf spring, determined through calibration using known weights. To improve the stability of the force measuring device compressed air was bled over it to prevent contamination of the optical system through condensation of silicone oil vapour. This was found to be effective and very desirable when testing at very low strain rates, and thus long test durations, and resulted in reduced drift in the sensor. To obtain different force sensor ranges and thus sensitivities, which is desirable for testing materials of different properties or at extremes of strain rate, leaf springs of different thickness were used: e.g. a thicker leaf spring for high strain rate measurements. Thus the performance of the force sensor was tuned to the measurement requirements.

The specimens used were typically either approximately 20 mm or 80 mm in length and 3 mm in diameter. Shorter specimens of smaller diameter could be used, provided the length to diameter aspect ratio of at least 10 is maintained, but this obviously reduces the magnitude of the tensile forces measured during testing. Two drum diameters of 40 mm and 80 mm were also available. By using the larger diameter winding drum and short specimen length, high strain rates up to a nominal value of ≈ 100 s^{-1} have been achieved using this instrument. Conversely, by using the smaller diameter winding drum and longer specimen length, low strain rates down to ≈ 0.001 s^{-1} have been achieved.

Testing has been carried out at up to 230 °C. The instrument has a maximum operating temperature of ≈ 260 °C, limited by the maximum operating temperature of the silicone oil into which the specimen is immersed.

6.2.2 Low strain rate testing

Particular difficulties that had to be addressed in developing the equipment to operate at low strain rates related to the control of specimen temperature and the measurement of low tensile forces. The heating system had to be developed to enable the temperature of the specimen to be controlled for the duration of the test. This was achieved by using a second oil bath to feed a heater coil in the test bath thereby providing heat input to counter the loss of heat from that bath to the surrounding environment. This solution was adopted as it provided the heat input to the test bath without causing excessive vibrational noise or excessive stirring of the silicone oil in the bath, which would otherwise swamp the measured force signal.

In testing at low strain rates, the tensile force generated in the specimen due to its deformation is very low. A thinner and thus less stiff leaf spring was used for testing at low rates to improve the accuracy of measurements. Also, improvement to the procedure for using the non-contacting force measuring device enabled the drift in the sensor due to contamination to be minimised. This resulted in greater confidence in defining the base line for the sensor which, because of the lower forces and longer durations of testing, became much more significant. Testing to failure, and thus testing to very long times, was important as it was necessary to determine a base line which was subsequently used to calculate the transient extensional viscosity values. As an indication of the maximum test duration, for materials that failed at a strain of three and were tested at 0.001 s^{-1} the test duration was 3000 s, or 50 minutes. Testing at low rates also presented problems for logging where relatively high logging rates were required at the start of the test, to capture the transients at the start, yet needed to continue for some considerable time thereafter.

6.2.3 High strain rate testing

High strain rate testing puts particularly strenuous demands on extensional measurement instrumentation in terms of the high speeds required and very short test durations. To obtain true tensile stress growth coefficient data one should have constant rate deformation of the specimen from the start of the test. For very high rate testing in particular this is not possible due to the inertia of the instrument and the very short test durations.

The performance of the motor over the first revolution was predicted for the NPL rheometer using the specification data for the drive motor and was also measured in calibration tests to determine its true response, Figure 3. The specification for the acceleration of the servo motor was 250 rev/s^2. Higher accelerations were theoretically possible using the motor, but were not achievable whilst keeping the motor under programmed control. The results demonstrated that the motor's performance was slightly worse than that predicted, presumably due to the added inertia elements of the specimen, drum and drum support. For tests at strain rates up to approximately 50 s^{-1} the error in time (and thus average rotation speed) taken to complete one revolution from a standing start was less than 5%. For the second rotation from start-up the error between the set and measured speeds was negligible.

In the more extreme case of testing at 100 s^{-1}, for tests up to a strain of 4 the predicted error in the time taken was approximately 30%. For shorter tests up to a strain of 3 this value increased to approximately 35%. These values fall below 10% for tests at a strain rate of approximately 50 s^{-1}. The strain at which the velocity reaches the set value for a test at 100 s^{-1} is estimated to be approximately 1.6, and 0.4 at 50 s^{-1}. Thus, as specimens normally failed at greater strain values than these, the strain rates at failure were equivalent to those set. Also, as a first approximation, only correction for the strain/time error need be applied for tests above approximately 50 s^{-1} as the magnitude of the errors are considered not to be too significant below that strain rate. For strain rates greater than 50 s^{-1} the magnitude of the correction can be estimated using linear interpolation of the above information.

For testing at high strain rates the data logging facilities captured data at high sampling rates up to 2000 per second. At 100 s^{-1} it would take 0.04 s to reach a strain of 4, thus giving 80 data points to define the shape of the test curve.

6.2.4 Measurement of strain rate using a video camera

It is desirable to check that the specimens deform as anticipated, given the set speeds of the instrument. For this purpose a video camera was used to record and hence measure the diameter of the specimen during testing. This information permitted the strains in the specimen during testing to be determined, and hence also the strain rates. This has demonstrated that the instrument performs well, with measured strain rates being approximately 95% of the set strain rates over a range up to 25 s^{-1}, Figure 4. Evaluation at higher strain rates was not possible as a shorter sample length of 20 mm length was used and it was not physically possible to get the camera into a suitable position to record the test.

All strain rates quoted below are nominal strain rates, without correction for acceleration and other effects (see Section 6.2.2). The implications of these errors for determination of extensional viscosity values are considered later.

6.3 Theory for the analysis of stretching flow data

The theory for the interpretation of the transient experimental data generated by the various types of constant strain rate instrument (Figure 1) is given in full in Appendix A2. In summary, for an instrument (Type A) with one fixed clamp and a rotating clamp of radius r rotating at ω rad/s with a specimen of length ℓ_o between the fixed and rotating clamps then the strain rate is constant for a fixed rotation speed.

The strain rate $\dot{\varepsilon}$ is given by

$$\dot{\varepsilon} = \frac{\omega \mathrm{r}}{\ell_o} \tag{7}$$

The strain ε is simply determined as the integral of strain rate with respect to time t and, as strain rate is constant, is given by

$$\varepsilon = \frac{\omega \mathrm{rt}}{\ell_o} \tag{8}$$

The cross-sectional area of the specimen is determined from the original cross-sectional area A_o of the specimen and its strain. Assuming conservation of volume on stretching, the transient tensile stress growth coefficient η_E^+ is given by

$$\eta_E^+ = \frac{\mathrm{F}\ \ell_o\, \mathrm{e}^{\left(\frac{\omega \mathrm{rt}}{\ell_o}\right)}}{\mathrm{A}_o\, \omega\, r} \tag{9}$$

where F is the force supported by the specimen. Equivalent expressions for other geometry types (see Figure 1) are presented in Appendix 2.

6.4 Specimen preparation

In carrying out specimen preparation, the operator is referred to the latest version of the standard ISO/CD 20965 Plastics – Determination of the transient extensional viscosity of polymer melts.

Specimens were prepared by extrusion using a capillary extrusion rheometer, as the preferred method, taking care to avoid entrapped air and extruding relatively slowly. Specimen lengths were selected from the extrudate to minimise variations in the diameter of the specimens and to avoid any defects that were visible. Typically the prepared specimens were a maximum of 100 mm in length and approximately 3 mm in diameter. Alternative methods of preparing specimens and the potential problems with these methods are discussed in Section 5.4.1.

The specimens were attached to small end-clamps before they were mounted onto the extensional rheometer. This approach enabled many specimens to be prepared at a time. The specimens were fixed to the end-clamps using an epoxy adhesive as follows. The ends of the specimen were treated by passing them through a butane flame. Care was taken to prevent any other part of the specimen, except that that was to be stuck, from being exposed to the flame. The ends were then dipped into the adhesive and attached to the end clamps. The specimen and end-clamps were then placed into an oven and the adhesive cured using a suitable time-temperature cycle. For the materials tested so far an epoxy adhesive (Ciba AV119) has proved to be suitable, with a cure cycle of 100 ^{o}C for 1 hour. The specimens were then allowed to cool before handling.

Normally the above procedure was found to be adequate. However, improved adhesion could potentially be obtained by dipping the ends, after flaming, into concentrated sulphuric acid for 30 seconds. However, care should be taken to prevent any other part of the specimen, except that to be stuck, from being exposed to the acid.

6.5 Experimental procedure

In summary, the following experimental procedure was developed for constant strain rate instruments. Many of the points are generic to testing using the various “types” of rheometer (see Figure 1), although some are stages are specific to the instrument developed by NPL. In carrying out experiments, the operator is referred to the latest version of the standard ISO/CD 20965 Plastics – Determination of the transient extensional viscosity of polymer melts, for specific details on testing such as the permitted tolerances on specimen dimensions, force measurement, strain and strain rate determination and variations in temperature along the specimen. Such values for the current version of the Standard are presented below. Use of this Guide must be in conjunction with the draft Standard.

Measure the diameter of the test specimen at at least five positions along its length and repeat after rotating the specimen by 90°. An average value for the diameter was then calculated from these ten measurements. For short specimens of 20 mm length, five measurements along the length may not be practically possible using a hand-held micrometer, in which case measurements should preferably be made at at least three positions.

Clamp the specimen in place on the rheometer between the drum and the leaf spring using the end-clamps stuck to the specimen.

Measure the length of the specimen between the clamps before immersing the equipment into the oil bath.

A specimen length to diameter aspect ratios greater than 10 should be used. Specimens typically had a length to diameter ratio of ≈ 30. However, a longer specimen will result in a reduction in the maximum strain rates that can be achieved. The magnitude of the end-errors can be assessed by using specimens of different length or diameter to produce different length to diameter aspect ratios. The effect on measured values can then be assessed.

After loading the specimen into the instrument, immerse the specimen into the silicone oil bath that had previously been allowed to reach equilibrium at the test temperature. The use of the oil bath provides temperature control and also support for the specimen during testing. The oil should be selected to provide the necessary temperature range for testing but also should be of a density that is similar to that of the materials to be testing thereby optimising the specimen support.

Measure the temperature in at least two positions along the length of the specimen to monitor temperature uniformity. The temperature should be measured near the specimen without contacting the specimen.

Allow the specimen to reach thermal equilibrium. Four minutes immersion time for testing at 150 °C was found to be suitable for specimens of 3 mm in diameter. The adequacy of the time allowed for the specimen to reach thermal equilibrium and the effect of the silicone oil on the specimen can be checked by varying the time for which the specimen is immersed in the oil bath before testing. The effect on measured values, obtained using different immersion times, can then be assessed.

In addition to the adequacy of time to reach thermal equilibrium, materials may be susceptible to degradation or further cross-linking and this may affect results. Such behaviour can be readily identified using an oscillatory rheometer, for example.

Peak forces measured in testing were up to ≈ 2 N for specimens approximately 3 mm in diameter. Peak forces for any configuration can be estimated as described in Appendix A3 (Equation A3.24) using data of the type presented, for example, in Figure 10. This will enable the operator to optimise the experimental set-up (where possible) to minimise errors, particularly in the measurement of force.

A correction for thermal expansion of the specimen diameter during heating may be made. Measurements of the density of a HDPE (HCA000) indicated a 20% decrease in density on heating from 25 °C to 150 °C and thus a corresponding increase in cross-sectional area, assuming the length is unchanged (due to internal stresses).

The diameter of the specimen may also be measured during the test by use of optical or cutting methods to derive true strains and strain rates and to assess the uniformity and accuracy of deformation. Obviously the cutting method results in the test being terminated once the cuts have been made and prevents data to failure from being obtained, and is thus not preferred. Such a check will also check for the effect of the compliance of the instrument on measured values.

Repeat testing is desirable, particularly on materials for which the operator has no previous experience, to establish the repeatability of the measurements and, in particular, that of the point of specimen failure. Repeat measurements do not, however, give an indication of the reproducibility and accuracy of the results.

When testing at very high strain rates correction or allowance for the inertia of the instrument may be necessary.

In accordance with the latest version of the Standard, ISO/DIS 20965 Plastics – Determination of the transient extensional viscosity of polymer melts:

- the diameter or width and thickness of the specimen, as appropriate, shall each be determined to and be uniform to within ± 2% of their average value,

- the length of the specimen between the clamps shall be determined to within 1% of its absolute value.

- the resolution of the force measuring device should preferably be no greater than 0,1%.

- the accuracy of force measurement shall be within ± 2% of the full scale value, although it is desirable and preferable, in particular for accurate measurements at low forces, that the accuracy of the force measurement device should be within ± 2% of absolute. However, this may be difficult to achieve in the lower part of the force transducer's range.

- the accuracy of strain determination or measurement shall be within ± 3% of the absolute value.

- the accuracy of rate of strain determination or measurement shall be within ± 3% of the absolute value.

- the spatial temperature uniformity shall be within ± 0,75 °C.

- the temporal temperature variation shall be within ± 1,0 °C of the set temperature.

- the temperature measuring device shall have a resolution of 0,1 °C and shall be calibrated using a device accurate to within ± 0,1 °C.

The user of this Guide should check, using the latest version of the Standard, whether these specifications have since been revised.

7. Experimental data for polymer melts

7.1 Introduction

To illustrate the various measurement issues, results obtained using the NPL constant strain rate instrument are presented and cover:

- typical experimental data, including the characterisation of three different polyethylenes (HDPE, LDPE and LLDPE) and three similar high density polyethylenes (Section 7.2)

- high strain rate experimental data (Section 7.3)

- low strain rate experimental data (Section 7.4)

Furthermore, a summary of results of an international intercomparison of stretching techniques is also presented (Section 7.5).

7.2 Typical experimental data

7.2.1 General

Tests were carried out on a range of polyethylenes at 150 °C using the extensional rheometer. Typically the specimen length was a maximum of ≈ 100 mm and the initial diameter was ≈ 3 mm. The experimental data of force versus strain are illustrated in Figure 5. As a constant strain rate was used in testing, the strain is directly proportional to time. In this case a strain of 1 corresponds to a time of ≈ 1 s. The trace shows that at the strain rate of 1.02 s^{-1} the force rose rapidly and peaked at ≈ 0.85 N in ≈ 0.6 seconds. The corresponding tensile stress growth coefficient curve, Figure 6, shows a continual increase in value up to a strain of ≈ 3 at which point the specimen failed. A strain of 3 corresponds to a reduction in the cross-sectional area of the specimen by a factor of ≈ ×20 and occurred within a duration of ≈ 3 seconds from the start of the test. The tensile stress growth coefficient data, Figure 6, take into account the exponential decrease in the cross-sectional area of the specimen (see Equations A2.4 and A2.5). The failure of the specimen is more easily observed in the tensile stress growth coefficient data than in the force data.

These curves highlight various experimental difficulties. For example, the maximum load to which the specimen was subjected corresponded to ≈ 0.85 N (equivalent to the force exerted by ≈ 85 g under gravity), Figure 5. At the point of failure the load had dropped to ≈ 0.1 N. Thus the forces measured during stretching of the specimen are relatively small, in particularly at large strains due to the considerable reduction in the cross-sectional area of the specimen. As the strain increases the percentage error in the measurement of force will increase significantly with consequent effects on the errors in the derived values of the extensional properties.

The use of different specimen length to diameter ratios in the range ≈ 7 - 30 (different lengths but of the same diameter of ≈ 3.2 mm) indicated there was little effect at strains above 0.5, Figure 7. However, at low strains of the order of 0.1 there was a possible correlation of aspect ratio with measured values. The use of shorter specimens resulted in higher tensile stress growth coefficient values, although the total variation was within approximately ± 10%. This trend is as expected as the effect of end-errors would increase for shorter specimens.

The repeatability of measurements, for example Figure 8, is shown to be good with variation less than approximately ± 5% for strains above 0.1.

Results of extensive testing of a HDPE (HGH000[2]) over three decades of strain rate from 0.01 s^{-1} to 10 s^{-1} are presented in Figures 9 and 10. When plotted as a function of time the data obtained at different strain rates overlap at low times forming an apparent master curve, Figure 10. The effect of increasing the strain rate is to shift to earlier time the point at which the curve departs from the master curve. When plotted as a function of strain the effect of strain rate is seen as a shift of data predominantly in the tensile stress growth coefficient axis, with lower strain rates corresponding to higher tensile stress growth coefficient values, Figure 9. For the strain rate of 10 s^{-1} it is observed that failure occurred within ≈ 0.25 seconds from commencement of the test indicating the need for high sampling rates to obtain adequate data, Figure 10.

Viscous heating of the specimen due to the work done in stretching, often a source of significant experimental error in shear flow testing, has been (over)estimated on the basis of data presented in Figure 5 and shown to be small, Appendix A4. The effect of viscous heating on the data presented here is therefore considered to be negligible.

7.2.2 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. Tensile stress growth coefficient data for the three polyethylenes are presented in Figures 11 to 14 and also combined in Figure 15 for a strain rate of ≈ 0.7 s^{-1} only.

These materials exhibited significant differences in their extensional strain hardening behaviour. The LDPE exhibited greatest strain hardening with tensile stress growth coefficient values increasing significantly with increasing strain. In comparison, the LLDPE exhibited no further strain hardening beyond a strain of ≈ 0.5. The HDPE exhibited an intermediate level of strain hardening. For these materials an equilibrium extensional viscosity (or *tensile viscosity)* could be determined for the LLDPE but not for the HDPE or LDPE.

It is considered that the high degree of scatter for the LLDPE data, Figure 14, is due to the low rate, low viscosity test approaching the lower limit of the force measuring sensor used. Both the low rate and low viscosity contribute to the low magnitude of the measured force values.

The peak tensile stress growth coefficient values, obtained prior to failure of the specimens, for these three polyethylenes and also for an additional HDPE (HGH000) have been plotted as a function of strain rate and clearly indicate the rate dependence of their behaviour, Figure 16. The behaviour of the LLDPE is relatively independent of the rate

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[2] NPL references to the materials, e.g. HGH000, are used to identify different grades of materials.

while the HDPEs and the LDPE have similar rate dependencies. The difference in tensile stress growth coefficient values was over two decades demonstrating a significant difference in the behaviour of these materials.

It is noted that the shear viscosities and extensional viscosities of the materials HDPE, LDPE and LLDPE exhibit different trends. In order of decreasing shear viscosity they rank LLDPE, LDPE and then HDPE [5], whereas in order of decreasing extensional viscosity they rank LDPE, HDPE and then LLDPE, Figure 15. The latter trend is exhibited by both the converging flow [5] and tensile stretching methods. This highlights the importance of extensional flow measurements in fully characterising the flow behaviour of materials, especially where those materials are to be used in processes in which the deformation is predominantly extensional. 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.

7.2.3 Measurement of three similar high density polyethylenes

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. Results for three similar HDPEs, each having PE100 rating and referred to as HGZ000, HHA000 and HHB000, 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 17. These shear flow 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 ≈ 50%, and HGZ000 and HHA000 had similar values but HGZ000’s were perhaps marginally higher.

The extensional rheometer developed at NPL was used to test these materials. Testing was carried out at three strain rates 0.1 s^{-1}, 1 s^{-1} and 7.9 s^{-1} at 200 °C, Figure 18. 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 ≈ 30%, and the difference between HGZ000 and HHA000 was ≈ 10%.

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 being 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, may be the more problematic of the materials for injection moulding, requiring higher pressures to force material through the gate region for example.

7.3 Low strain rate data

Low strain rate testing of various polyethylenes (HDPE, LDPE and LLDPE) has clearly illustrated the measurement difficulties that are prevalent in such testing. Data obtained at low strain rates are presented, for example, in Figure 19 [75]. Data at strain rates as low as 0.001 s^{-1} on HDPEs have since been obtained [75]. For the LDPE (HGE000) considerable strain hardening was exhibited, Figure 19. Transient extensional stress growth coefficient values exhibited a decade increase in magnitude on departing from the master curve[3]. However, at the lowest strain rate used (0.002 s^{-1}) no apparent strain hardening was observed. The reason for the absence of strain hardening is unclear. It may have been due to premature failure of the specimen. The lack of strain hardening means that the specimen is relatively unstable and should any necking occur it is likely to fail prematurely. However the repeat test showed almost identical behaviour at long times indicating the high level of repeatability of the behaviour.

The peak force measured in the 0.002 s^{-1} strain rate test was less than 0.005 N (equivalent to 0.5 gf), thus indicating the small forces that need to be measured. The resolution of the force measuring device was 0.000024 N. Scatter in data due to vibrational noise, in part from the drive system, was more apparent when testing at the lowest strain rates but could be reduced further by modifying the filtering of the force sensor signal. These values thus demonstrate the severe demands placed on such an instrument for use when testing at low strain rates.

7.4 High strain rate data

Results for various polyethylenes (HDPE, LDPE and LLDPE) clearly demonstrate measurement issues related to high strain rate testing. For the LDPE (HGE000) strain rates up to 100 s^{-1} were achieved with test times less than 0.1 s, Figure 20. It is noted that strains are the natural logarithm of the extension ratio and thus a strain rate of 100 s^{-1} is significantly greater than that of 10 s^{-1} in terms of the rate of deformation occurring: a strain rate of 1 s^{-1} corresponds to stretching the sample to 2.7 times its length per second, a

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[3] The master curve refers to the superimposition of data obtained at different strain rates when plotted as transient stress growth coefficient versus time. Additional strain hardening refers to the departure of data from this master curve at high strain values.

rate of 10 s^{-1} corresponds to 22000 times its length per second, and 100 s^{-1} to 2.7 $x10^{43}$ times its length per second. However, in practice the times for which these latter deformation rates occur is significantly shorter than one second and thus the total strains are also significantly shorter.

The repeatability of results is shown to be quite good, Figure 20, and even at the highest strain rate of 100 s^{-1} [76]. The feature of the extensional behaviour that is hardest to achieve repeatably is the point of failure of the specimen. It is reasonable to assume that where repeat testing is carried out the highest strain to failure is the most reliable: any premature failures being due to inhomogeneities in the specimens.

It was noted that the oscillatory waveform evident in some of the results was particularly pronounced at a test rate of 40 s^{-1}, Figures 20 and 21 [76]. This was considered to be due to the fact that the resonant frequency of the force measuring system, which was approximately 40 Hz, corresponded with the effective rate of loading at this strain rate. The resonance appeared to be less pronounced at 100 s^{-1}. Data obtained in the presence of resonance was in agreement with data obtained using a longer specimen for which resonance was not such a significant problem [76]. The main cause for concern with the resonance is its effect on the accuracy of the values obtained at short time scales that are comparable with the period of oscillation of the resonance. Resonance of the force sensor is considered difficult to overcome due to the need for a sensor of low force capability, and thus low stiffness. However, the use of specimens of different length may provide a means to minimise the effect.

From the repeat measurements, Figure 20, the use of the short length specimens of $\approx$ 20 mm was shown not to be problematic at strains in excess of $\approx$ 0.5 at 25 s^{-1} (i.e. > 0.02 s), Figure 20 [the use of short specimens is essential for obtaining high strain rate data]. Below this values of strain the results differ more markedly due to the very short times and low forces that they correspond to, along with the resonance of the force sensor. Although not conclusive, but indicated by HGE000, Figure 20 and [76], an examination of the viscosity versus time curves showed that at times less than 0.01 s the lowest value extensional viscosity data corresponded to that obtained at the highest rates. This is consistent with the effects of the initial acceleration of the drum in that the forces generated will be lower as the strains are lower than expected. As a consequence the derived extensional viscosity values will also be lower. On this basis, when examining viscosity versus time plots for high rate data it is more appropriate to assume that, at short times, the data obtained at the lower strain rates are more reliable.[4]

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[4] Tensile stress growth coefficient versus time plots are found useful as data obtained at different strain rates tend to form a master curve, with departure from that master curve occurring at high strains due to additional strain hardening.

7.5 Results of an intercomparison of stretching techniques

The results of an intercomparison of stretching methods comprising 6 laboratories indicated that the variation in tensile stress growth coefficient values was estimated to be up to approximately ± 60%, Figures 22 and 23 [77]. Measurements were performed mainly over a strain rate range of 0.01 s^{-1} to 1 s^{-1}, and measured tensile stress growth coefficient values were predominantly in the range 5 x 10^4 Pa.s to 3 x 10^6 Pa.s. The observed variation in peak tensile stress growth coefficient values was up to approximately ± 100% indicating the greater difficulty in determining these failure values. A high density polyethylene that was stable at 190 °C for in excess of two hours was used as the intercomparison material. Samples of the material were provided to each of the participants who produced their own specimens. Measurements were made at 150 °C and 190 °C [77].

NOTE: The quoted variation in tensile stress growth coefficient values excludes data at short times/low strains for which the variation is greater due to instrument inertia effects and that the resolution of the measuring equipment may be insufficient for reliable measurement of the low forces generated. Measurement errors will be strongly dependent on the magnitude of the force to be measured which, in turn, will be strongly dependent on the strain. The values quoted are indicative for the various instruments that were involved in the intercomparison.

8. Estimate of uncertainties in extensional testing

8.1 Intermediate strain rate testing

Due to the considerable stretching that the specimen normally undergoes during testing, particularly on reaching high strain values, there is a very significant decrease in the cross-sectional area of the specimen, accompanied by a similarly large decrease in the measured force. As a consequence there is a significant increase in the uncertainty in the measurements. To understand and quantify the level of confidence that one can put in the accuracy of the results obtained, it is important that the uncertainties in the measurements, in particular that due to the accuracy and resolution of the force measurement, are known.

An analysis of the uncertainties of extensional measurements is presented in Appendix A3 [78]. The equations can be used to calculate the uncertainties in the derived tensile stress growth coefficient values, given the uncertainties in each of the components of the measurement. The analysis can thus be used to identify the critical parameters that generate the uncertainties in the measurements and can thus be used to improve on the measurement method.

The example presented, Appendix A3 and Figure 24, is based on a HDPE (HGH000) at 150 °C and a strain rate of $\approx 1\ s^{-1}$ (Figure 10). It illustrates the significant increase in measurement uncertainties at high strains due, predominantly, to the increase in the uncertainty in the measurement of force resulting from the significant reduction in the cross-sectional area of the specimen. The estimated uncertainties increased dramatically at high strain values, Figure 24. Expanded uncertainties representing 95% confidence limits are presented and were estimated to be of the order of ± 20% at a strain of 3.3, ± 50% at a strain of 4.8 and ± 100% at a strain of 5.7.

8.2 Low strain rate extensional viscosity testing

At low strain rates the uncertainties in the low forces generated at low strain values are potentially significant. The analysis was used to estimate the uncertainties in testing for the HDPE (HFU000) at $0.001\ s^{-1}$, Figure 25. The results show measured tensile stress growth coefficient data along with calculated values, assuming a power-law fit, and 95% confidence limits to the calculated data. The relative combined uncertainty is also presented. To provide an improved analysis the strain range had been split into two regions, that above and that below a strain value of approximately 0.4. This enabled different power-law parameters to be used in each region, thereby more accurately fitting both the master curve and additional strain hardening parts of the experimental data.

The principal difficulty in carrying out the uncertainty analysis for these low rate tests was in establishing the value for the uncertainty in the force measurement at the very low values of force involved. A value for the error in measurement of the force was determined from the calibration curve for the force sensor. These uncertainty calculations were very sensitive to the magnitude of that chosen value, and consequently the uncertainties in measurement of transient stress growth coefficient values may differ quite markedly from those plotted. However, the results for the uncertainty calculations appear to be reasonable (if not excessively large for the lower 95% limit at low strain values) and reflect the relatively high confidence in results of transient stress growth coefficient data at low strains based on the fact that the low rate data lie on or near the master curve generated by data obtained at higher strain rates, e.g. Figure 19 [75].

8.3 High strain rate extensional viscosity testing

As demonstrated earlier, the uncertainties in extensional viscosity determination grow rapidly at large strains primarily due to the rapid decrease in cross-sectional area of the specimen with a resultant rapid decrease in the measured force. At high strain rates acceleration issues also become significant, as discussed in Section 6.2.3. Due to the acceleration behaviour of the instrument the strain achieved in a given time is less than that that would be achieved if an instantaneous acceleration were assumed. Furthermore, the difference in the measured to set strain rates, as observed using the video camera, has an effect on the accuracy of the derived results. The analysis of the uncertainties in extensional testing was used to estimate the uncertainties in testing for the HDPE (HFU000) at 25.5 s^{-1} and 100 s^{-1}, taking into account these additional errors, Figures 26 and 27. The errors in the strain rate due to the acceleration of the drum and the visual observations has been accounted for by assigning a 15% or 40% error to the angular speed of the drum for set strain rates of 25.5 s^{-1} and 100 s^{-1} respectively. The contribution due to the visual observations was set at 5%, the remainder being due to the acceleration of the drum. The plots, Figures 26 and 27, show the measured tensile stress growth coefficient data along with calculated values, assuming a power law fit, and 95% confidence limits to the calculated data. The results show a rapid increase in the uncertainties as strains increases beyond a value of approximately 1, with the effect becoming more significant at higher strain rates. The 95% confidence limits to the data are not true confidence limits because the errors due to the acceleration and strain rates are systematic. The effect of both the acceleration phase and the error in strain rates observed using the video camera would be to result in lower transient stress growth coefficient values being determined than actual. Thus the upper 95% limit gives an indication of the maximum shift in data that would be necessary to correct for these effects, rather than being due to random uncertainties in the various parameters.

9. Summary

Extensional flow characterisation yields information about the materials not revealed by shear flow measurements. A comparison of extensional viscoelasticity data with shear viscosity data [5] 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.

Guidance and recommendations for the measurement of the extensional flow properties of polymer melts, including at very high and very low strain rates, have been presented. Analyses to determine tensile stress growth coefficient values and an analysis of the uncertainties in data have also been presented. The method developed by NPL has been used to illustrate the measurement of the transient extensional flow behaviour of polymer melts, using a range of polyethylenes, and has highlighted the measurement issues that need to be addressed in order to obtain reliable data. Results of an international intercomparison are also presented.

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Figures

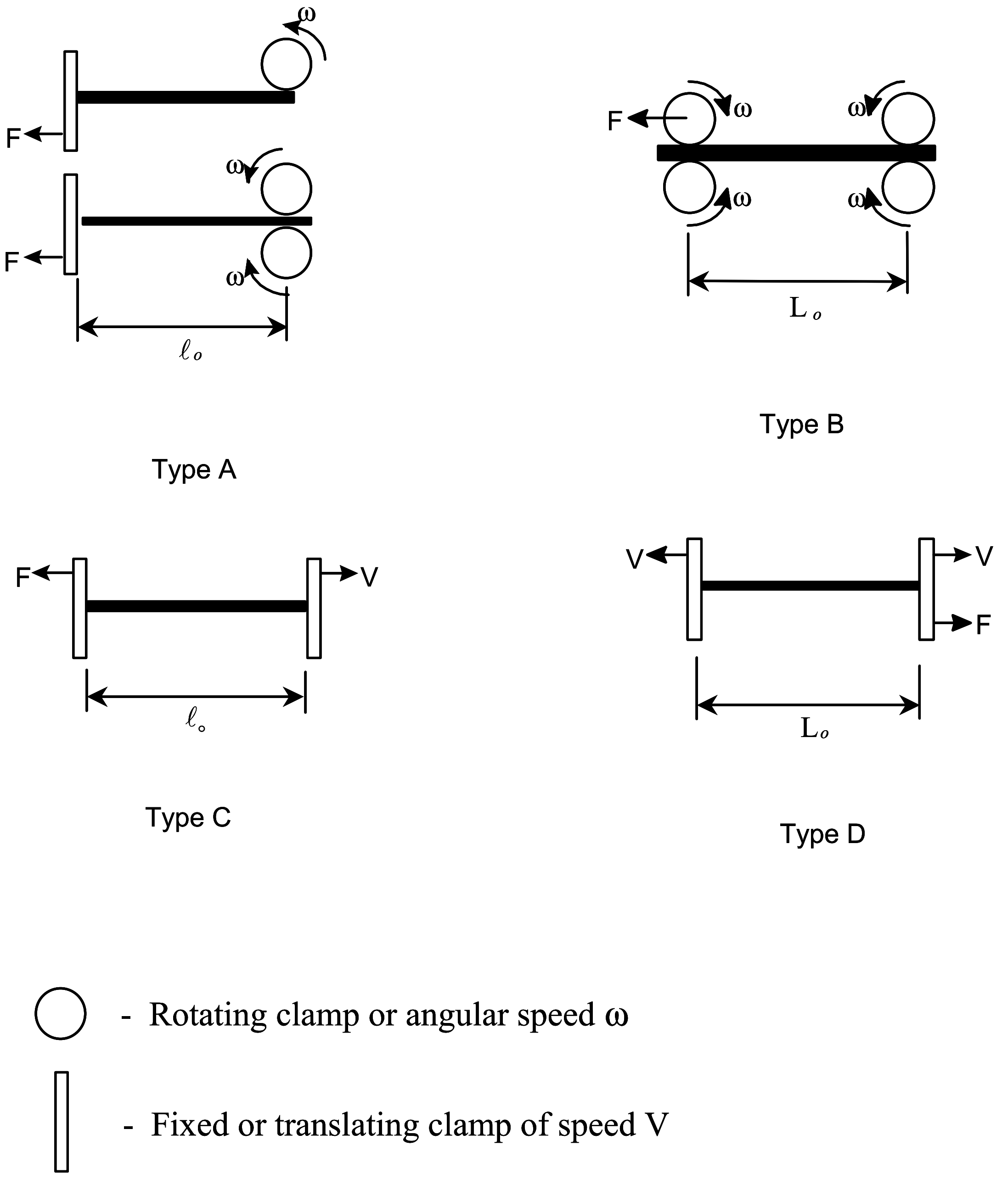

Figure 1: Schematic of test instrument Types A to D.

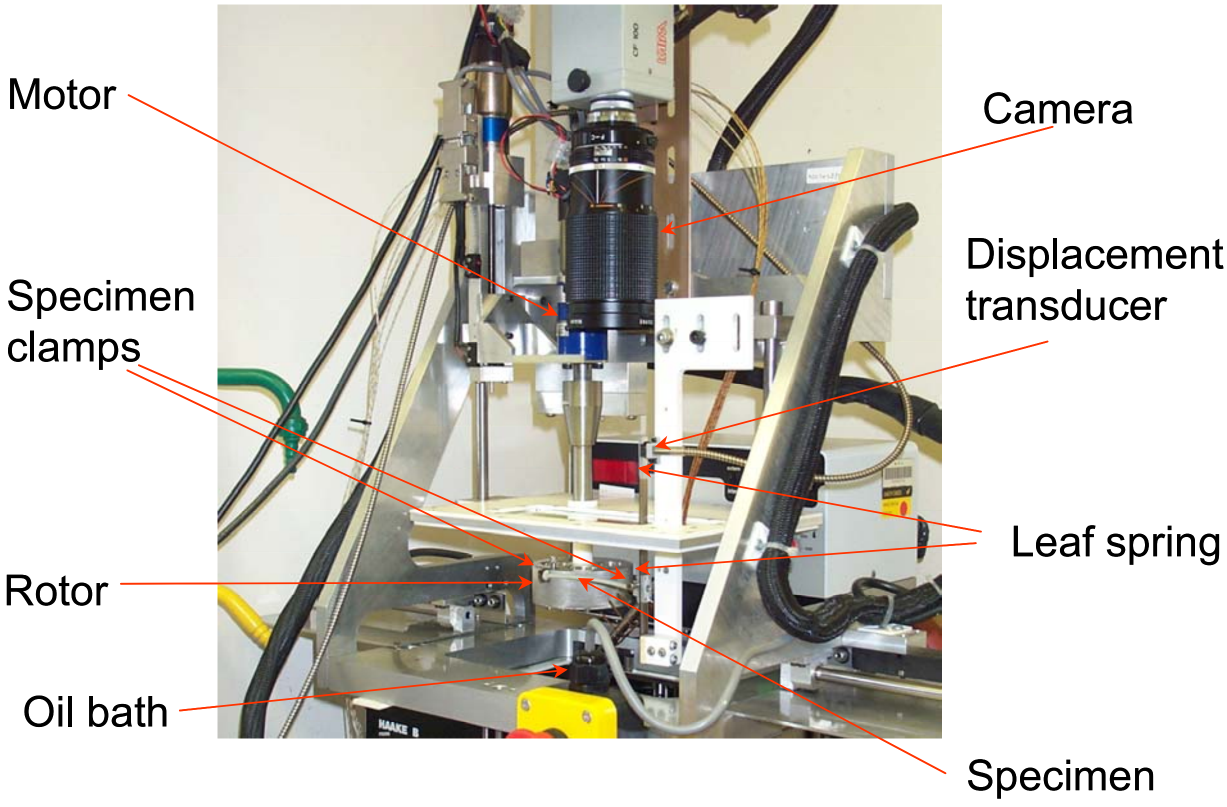

Figure 2: The NPL extensional rheometer showing the sample clamped between the rotating drum and force measuring device, suspended above the silicone oil bath.

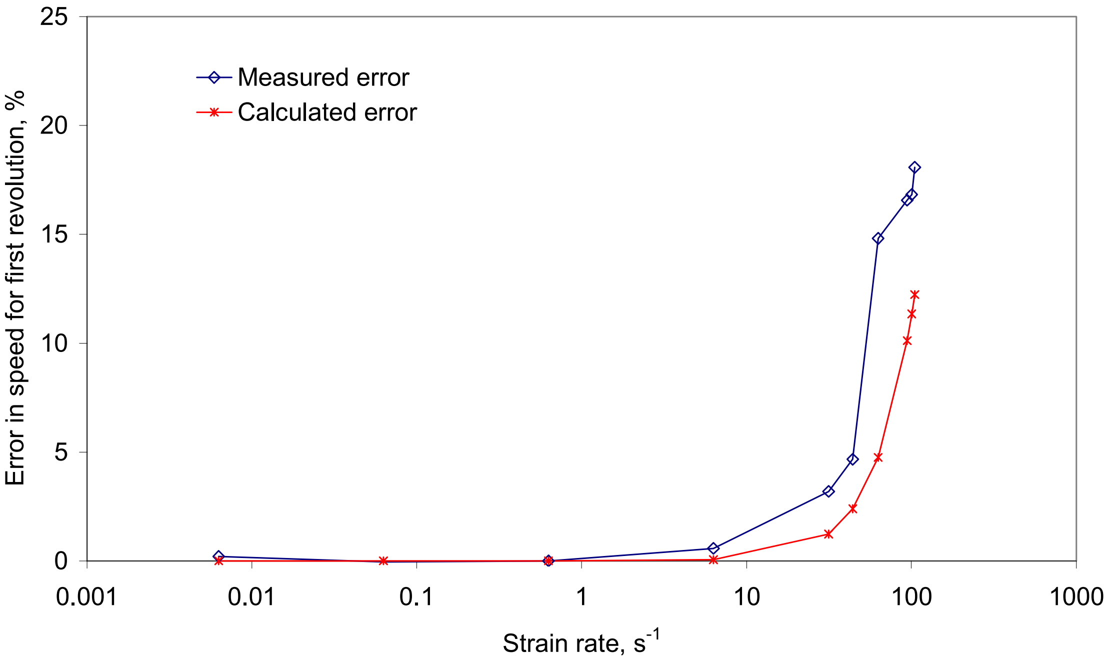

Figure 3: Error in rotation speed due to drum acceleration (using specimen length of ≈ 20 mm and drum radius of 40 mm).

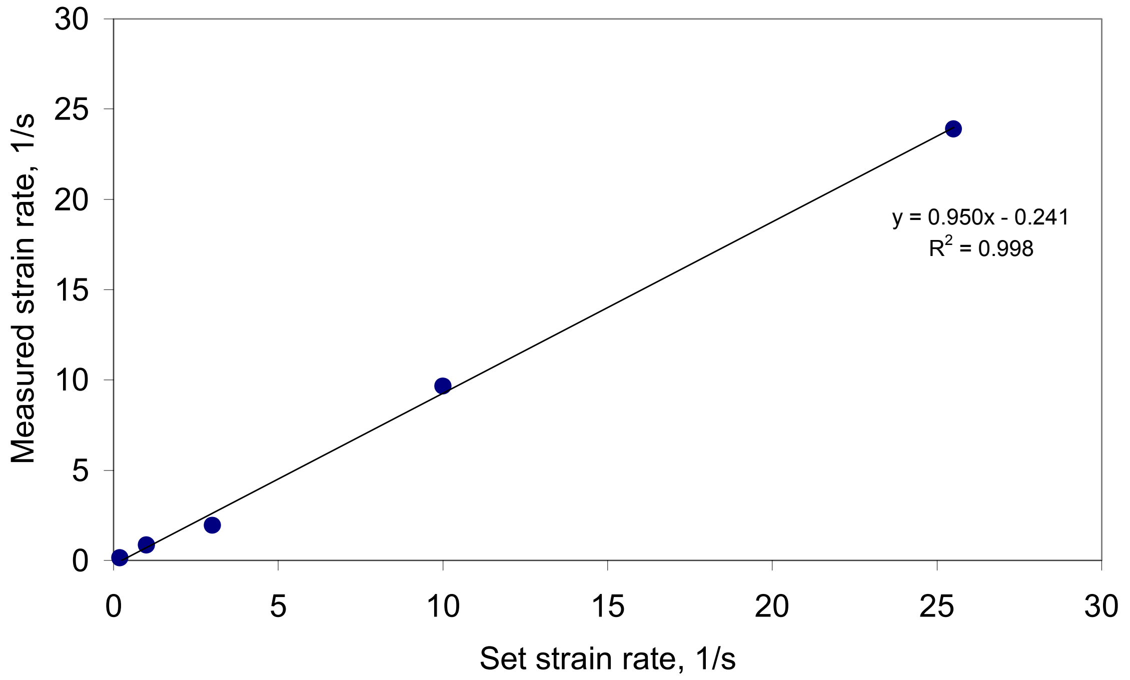

Figure 4: Comparison of set with optically measured values of strain rate for the extensional rheometer, the measured values being determined using a video camera.

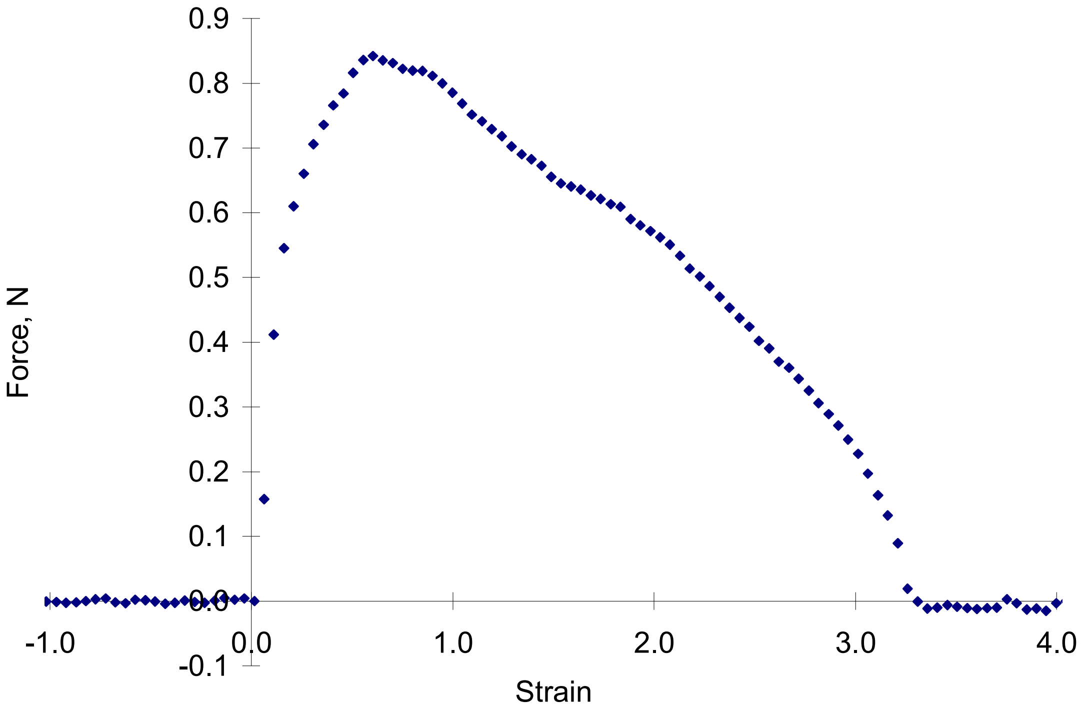

Figure 5: Extensional testing of a HDPE at a strain rate of 1.02 s^{-1} and 150 ^{o}C showing the raw force versus strain data necessary for determining extensional parameters (HGH_EXT_074).

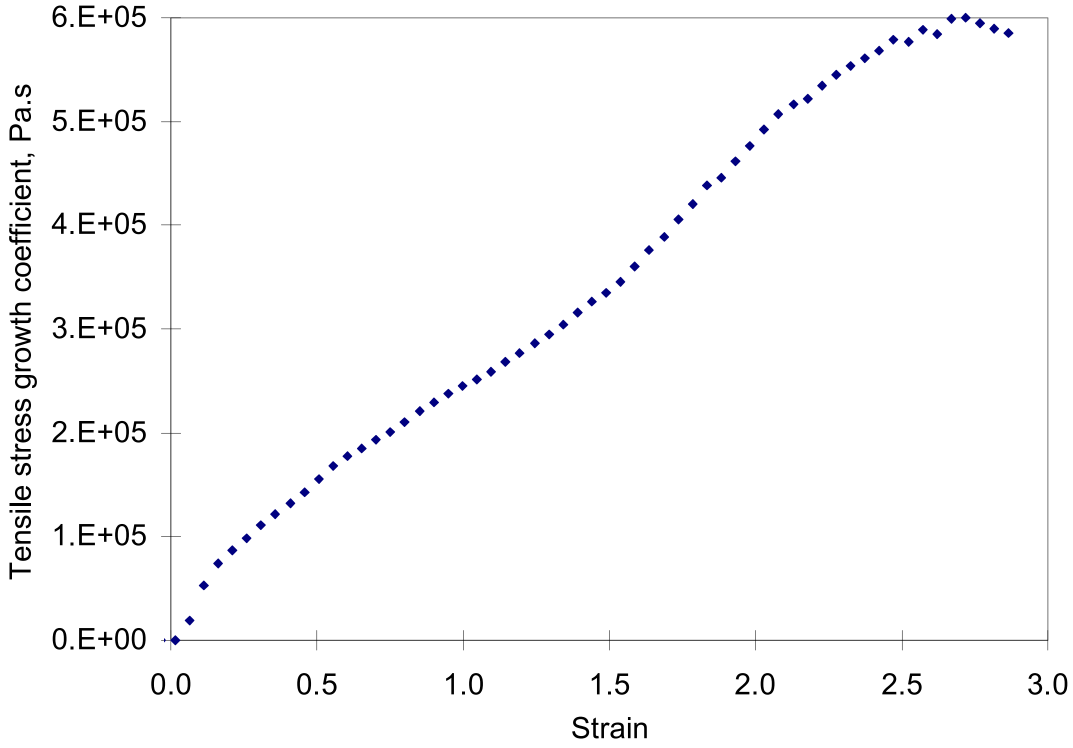

Figure 6: Transient extensional viscosity (or tensile stress growth coefficient) versus strain plot for the same test as presented in Figure 7 (HGH_EXT_074).

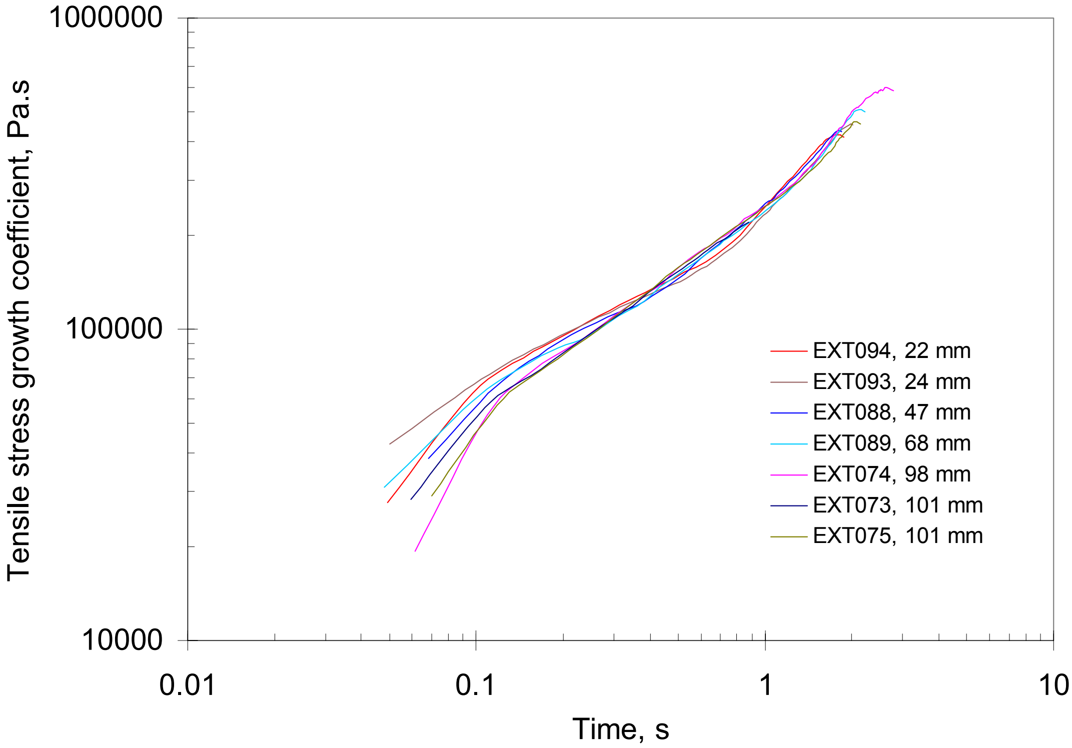

Figure 7: Effect of specimen length in the range 22 mm to 101 mm on the tensile stress growth behaviour of a HDPE (HGH000) at 150 °C and a strain rate of 1 s^{-1}.

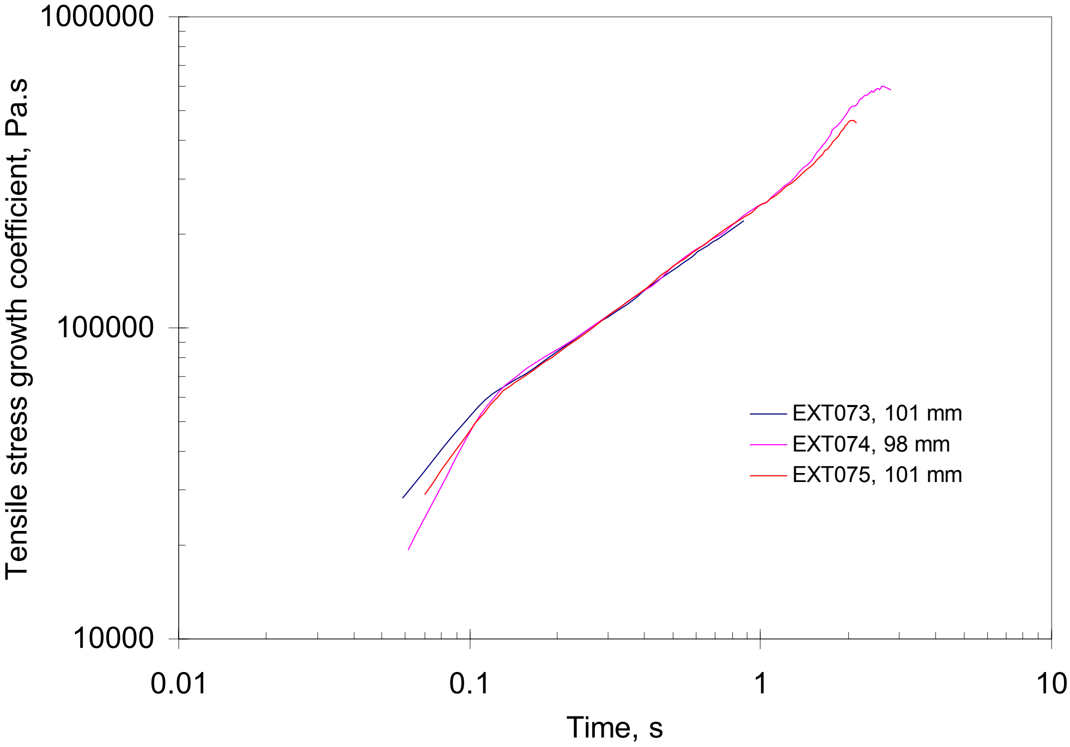

Figure 8: Effect of immersion time in silicone oil on extensional flow behaviour of a HDPE (HGH000) at 150 °C and a strain rate of 1 s^{-1} (EXT073 & EXT075: 4 minutes, EXT074: 8 minutes).

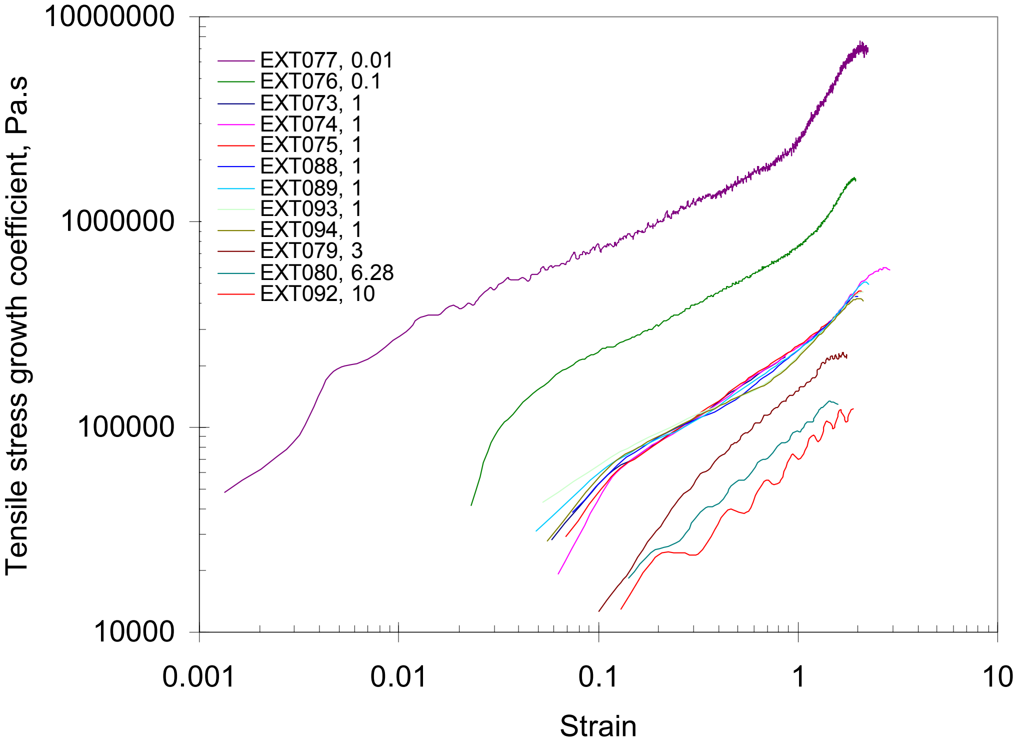

Figure 9: Effect of strain rate from 0.01 s^{-1} to 10 s^{-1} on tensile stress growth behaviour of a HDPE (HGH000) at 150 °C.

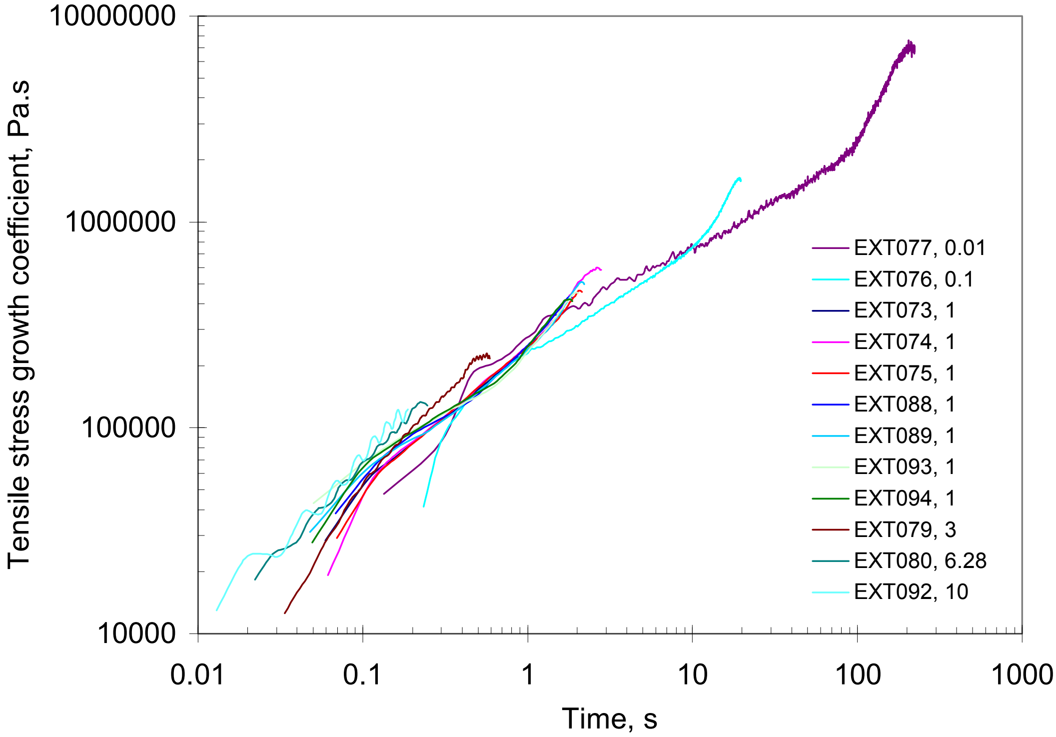

Figure 10: Effect of strain rate from 0.01 s^{-1} to 10 s^{-1} on tensile stress growth behaviour of a HDPE (HGH000) at 150 °C.

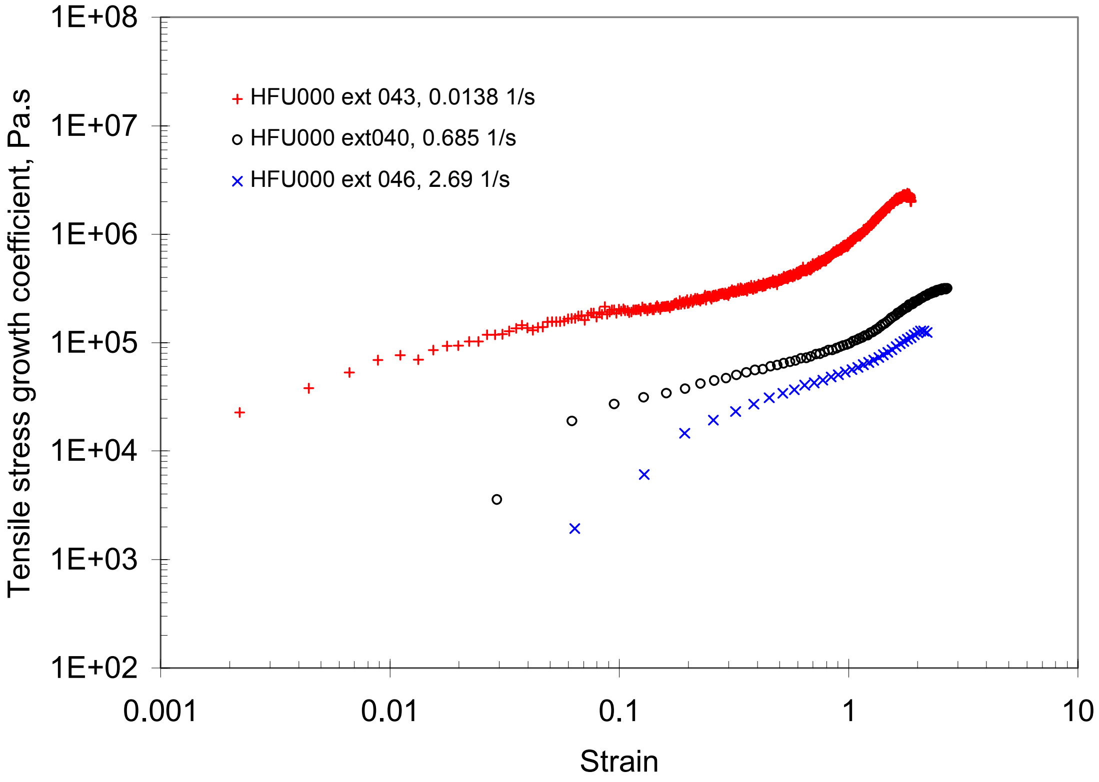

Figure 11: Extensional flow measurements on a HDPE (HFU000) at 150 ^{o}C plotted as a function of strain, indicating the effect of strain rate on the flow behaviour.

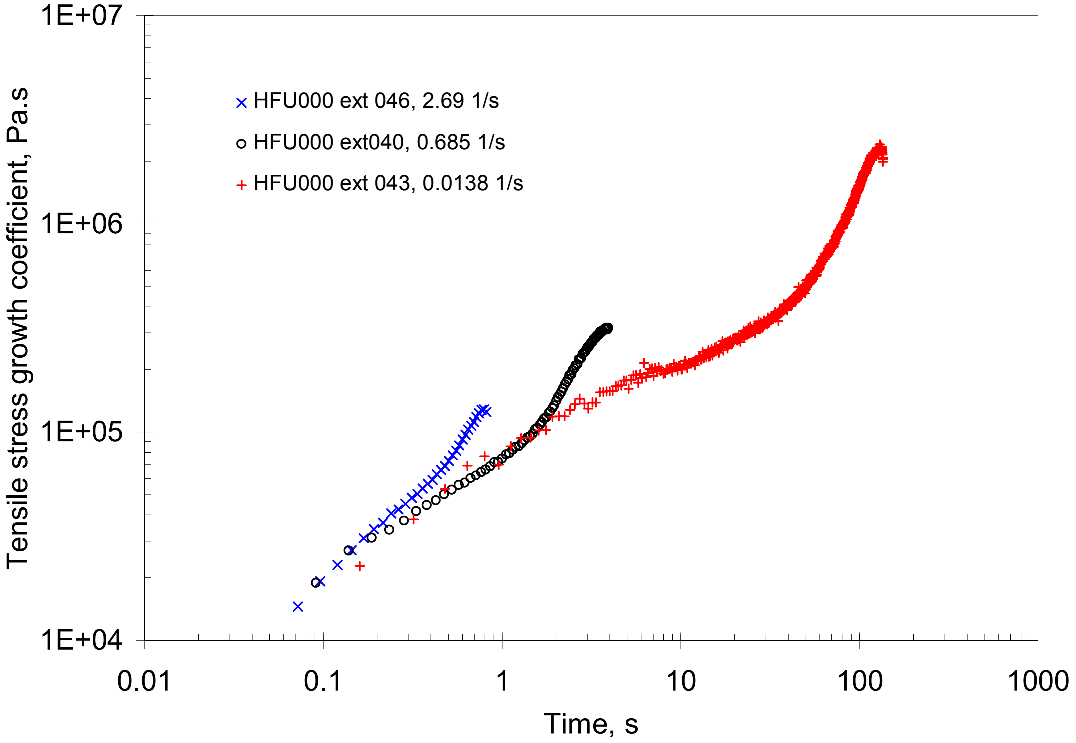

Figure 12: Extensional flow measurements on a HDPE (HFU000) at 150 °C indicating the overlap at short times of data obtained at different strain rates.

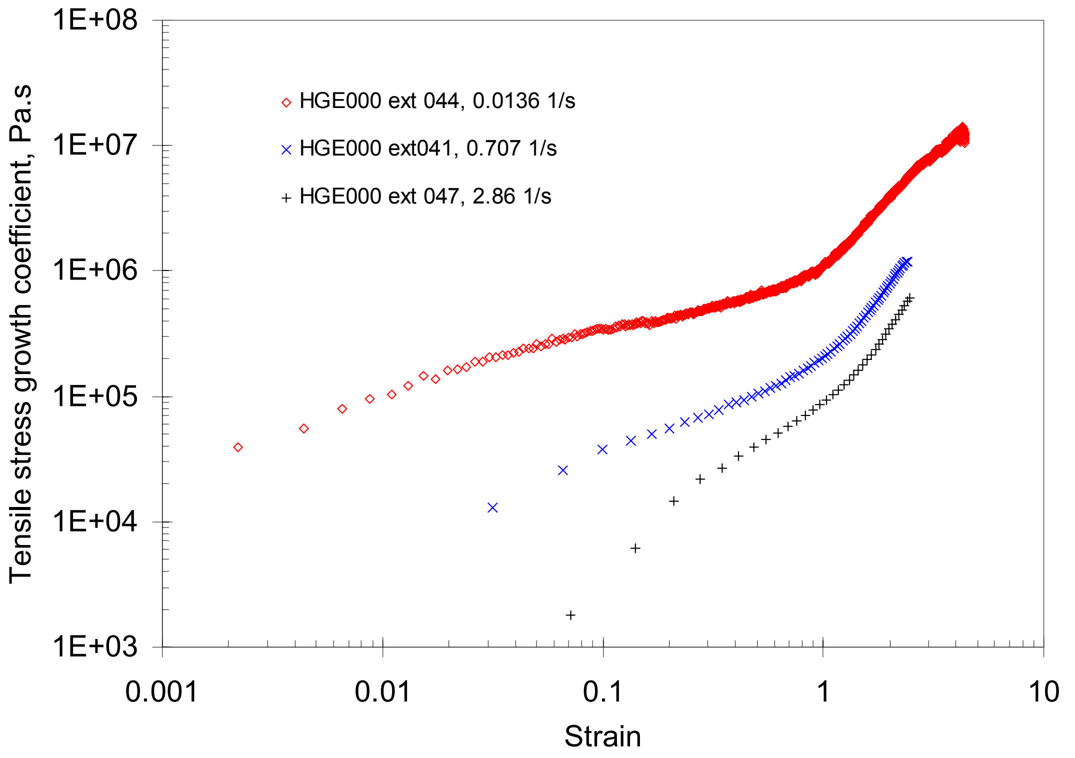

Figure 13: Transient extensional viscosities for LDPE (HGE000) at 150 °C at three strain rates.

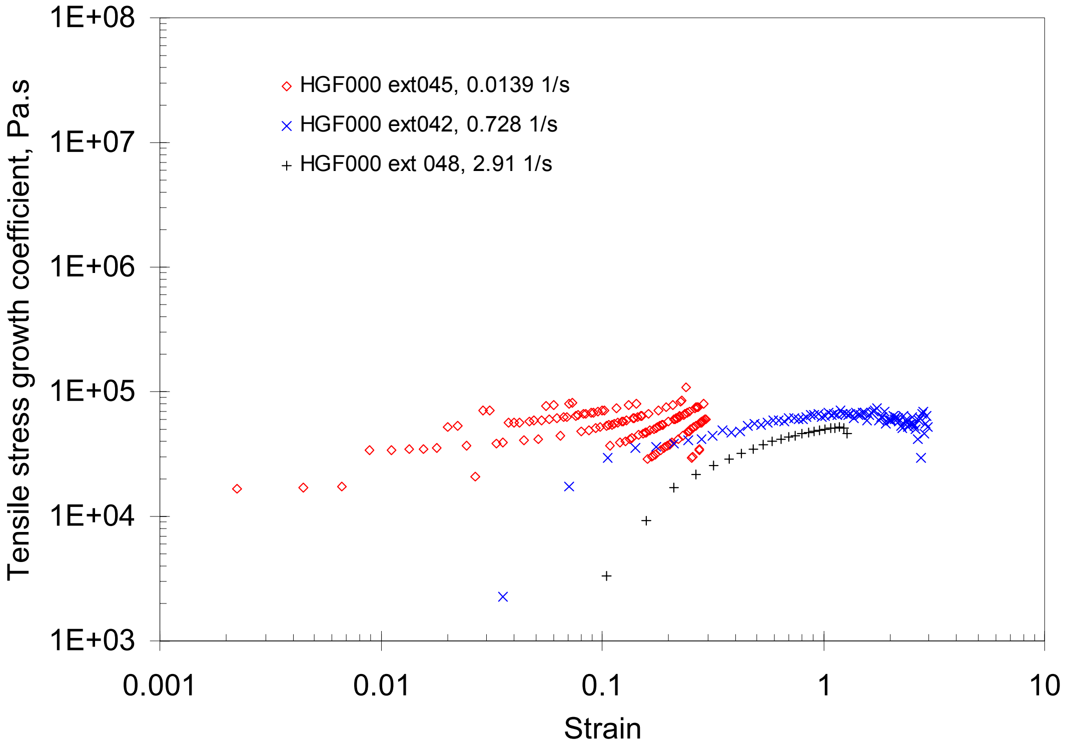

Figure 14: Transient extensional viscosities for LLDPE (HGF000) at 150 °C at three strain rates.

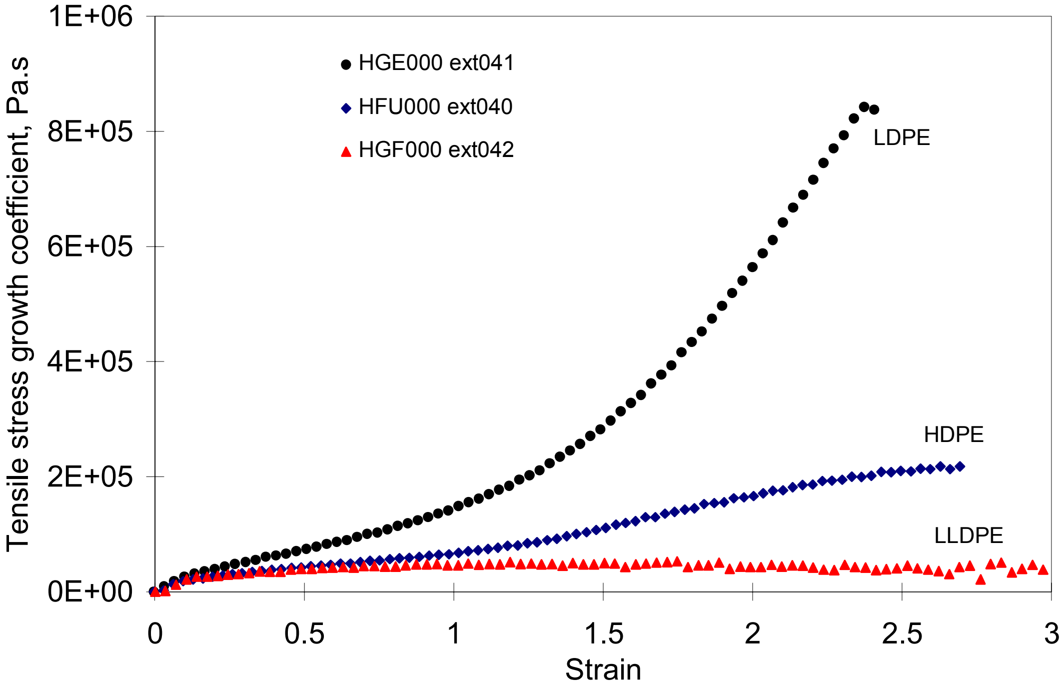

Figure 15: Extensional flow measurements on three polyethylenes at 150 ^{o}C exhibiting significant differences in strain-hardening behaviour at a strain rate of ≈ 0.7 s^{-1}.

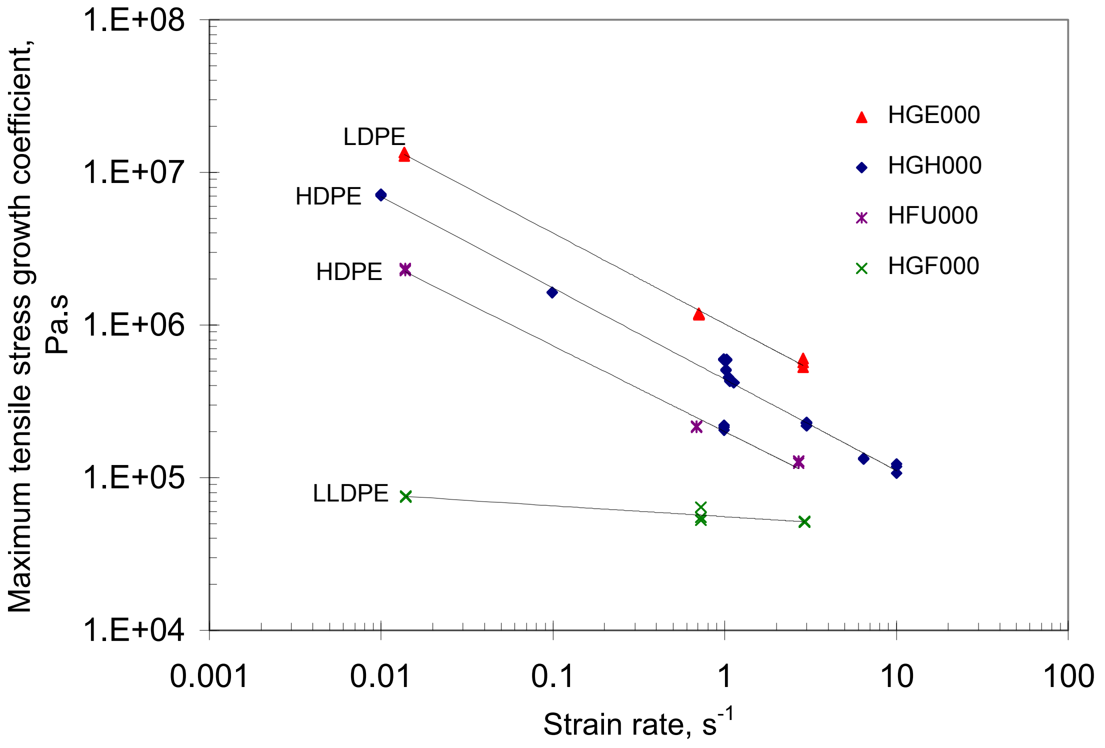

Figure 16: Comparison of maximum tensile stress growth coefficient values obtained by a stretching method for four polyethylenes LDPE (HGE000), LLDPE HGF000), HDPE (HFU000) and HDPE (HGH000) at 150 °C.

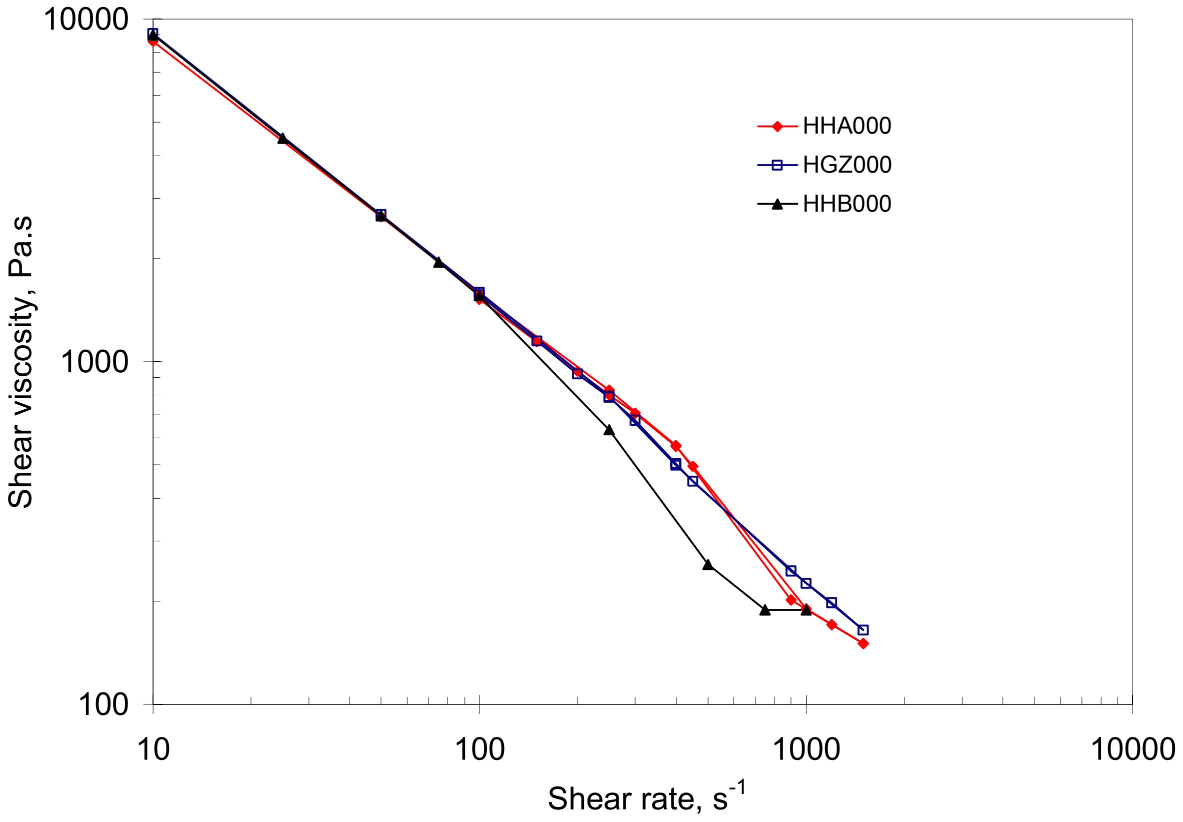

Figure 17: Comparison of shear viscosities of three polyethylene materials at 200 °C, suggesting melt fracture at shear rates above 100 s^{-1}.

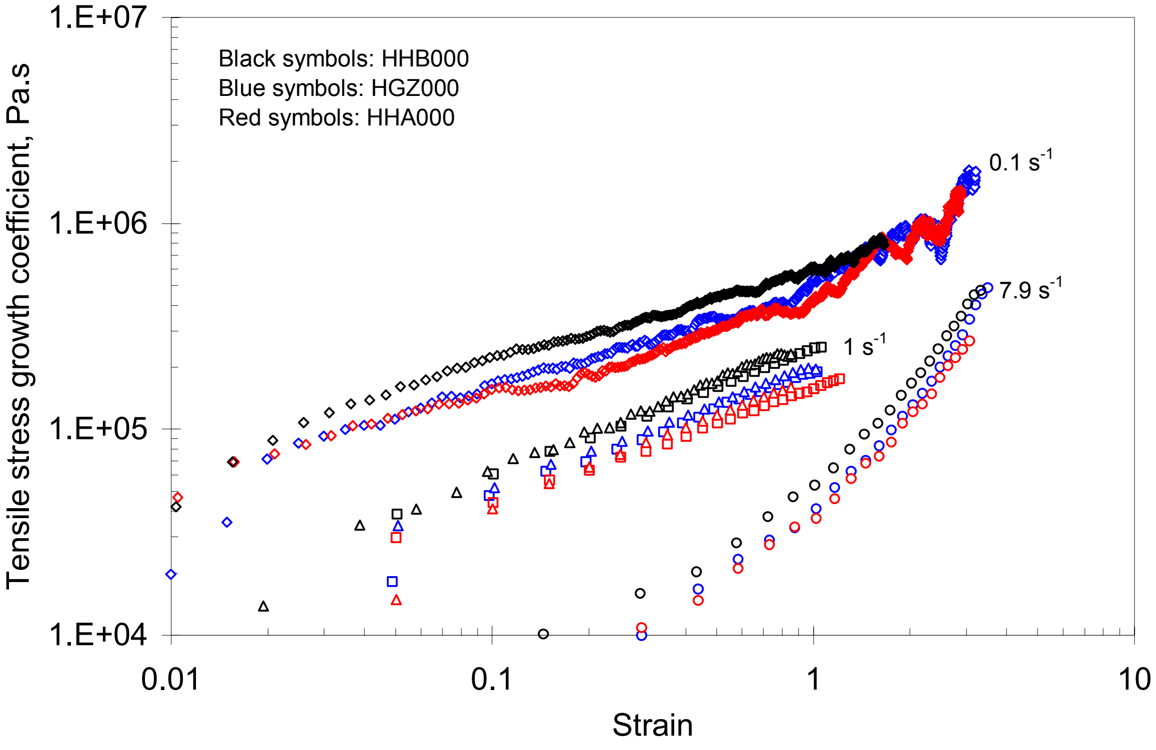

Figure 18: Comparison of the tensile stress growth coefficient behaviour of three high density polyethylenes at three different strain rates at 200 °C.

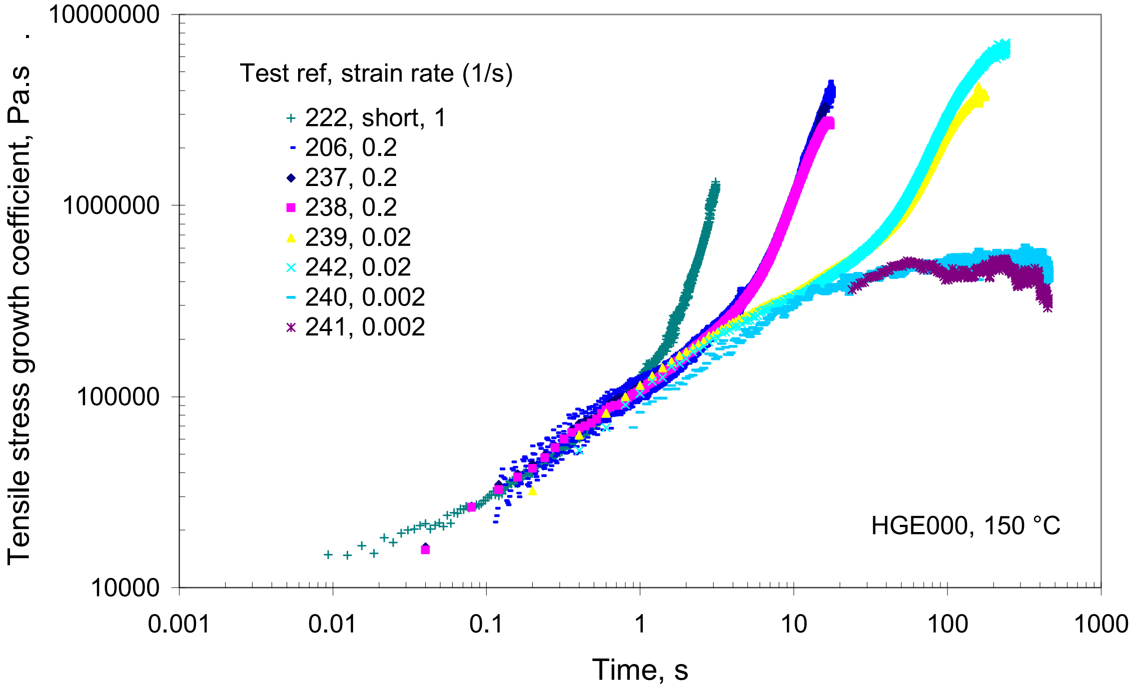

Figure 19: Extensional viscosity data as a function of time for a LDPE (HGE000) at 150 °C.

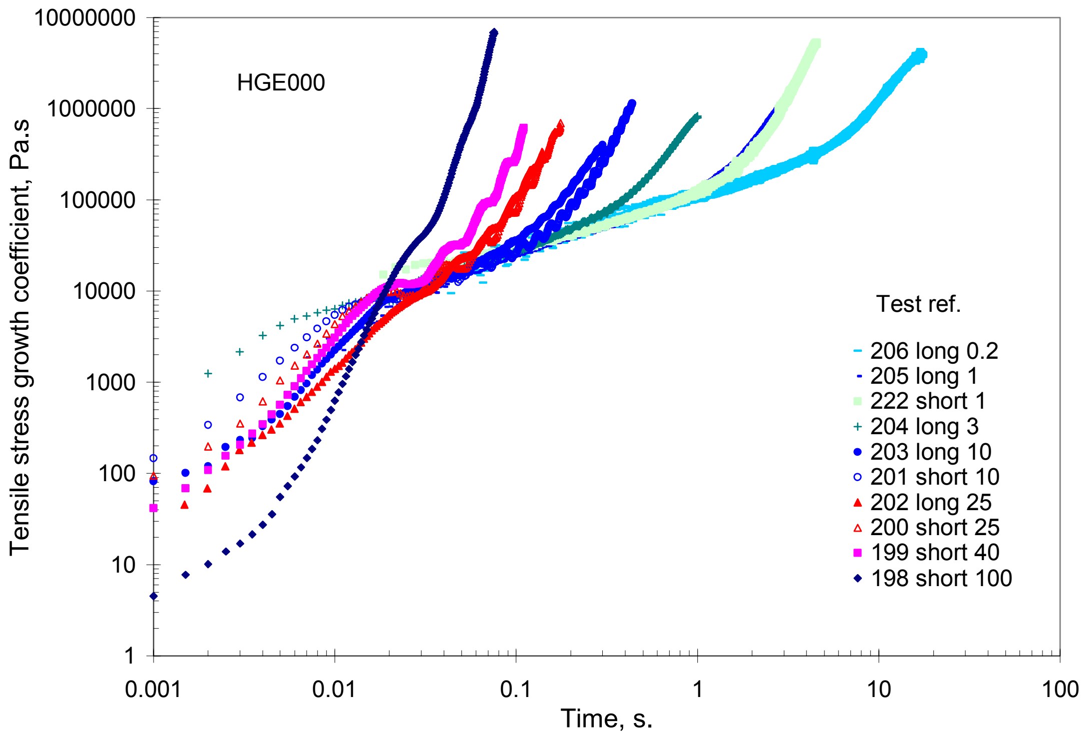

Figure 20: Extensional viscosity data as a function of time for a LDPE (HGE000) at 150 °C. The legend presents: test reference, specimen length (long or short) and strain rate.

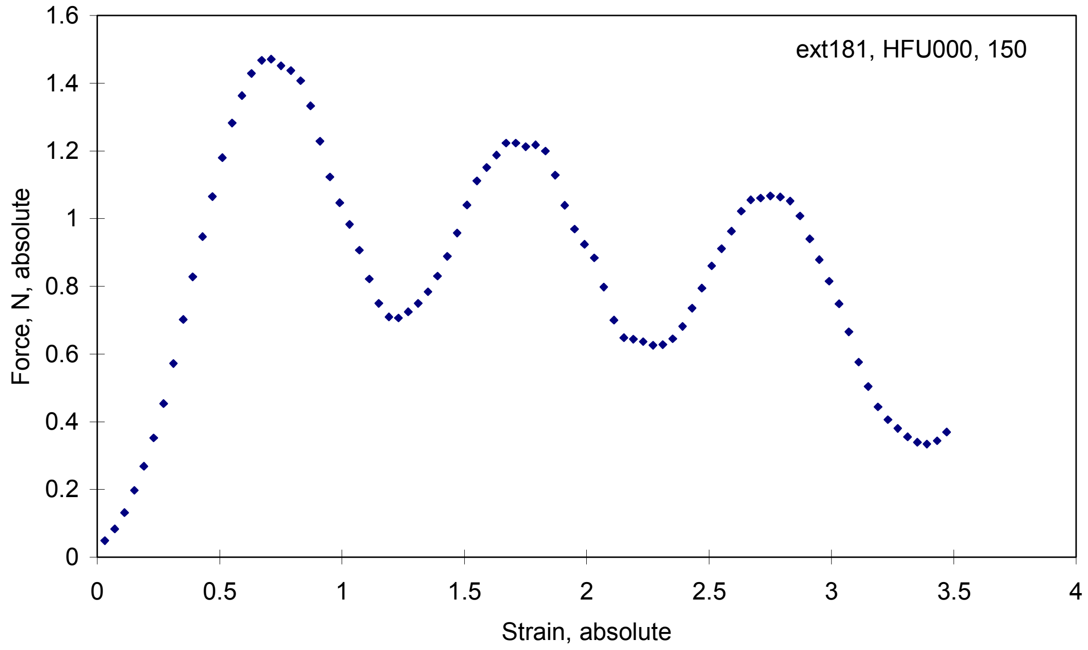

Figure 21: Resonance of the force measuring system as observed when measuring the extensional viscosity of a HDPE (HFU000) at 150 °C. Strain rate = 40 s^{-1}, thus a strain of 1 corresponds to a time interval of 0.025 seconds.

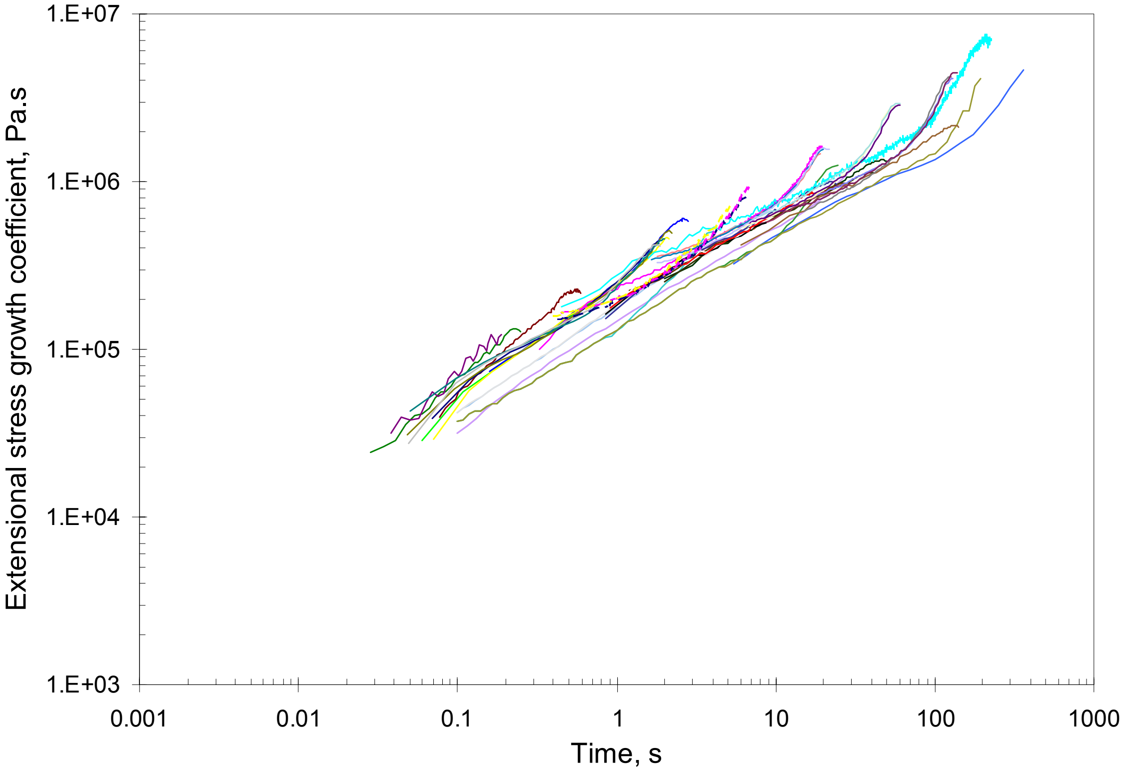

Figure 22: Intercomparison of stretching flow methods for a HDPE (HGH000) at 150 ºC: laboratory 3 and short time laboratory 8 data removed.

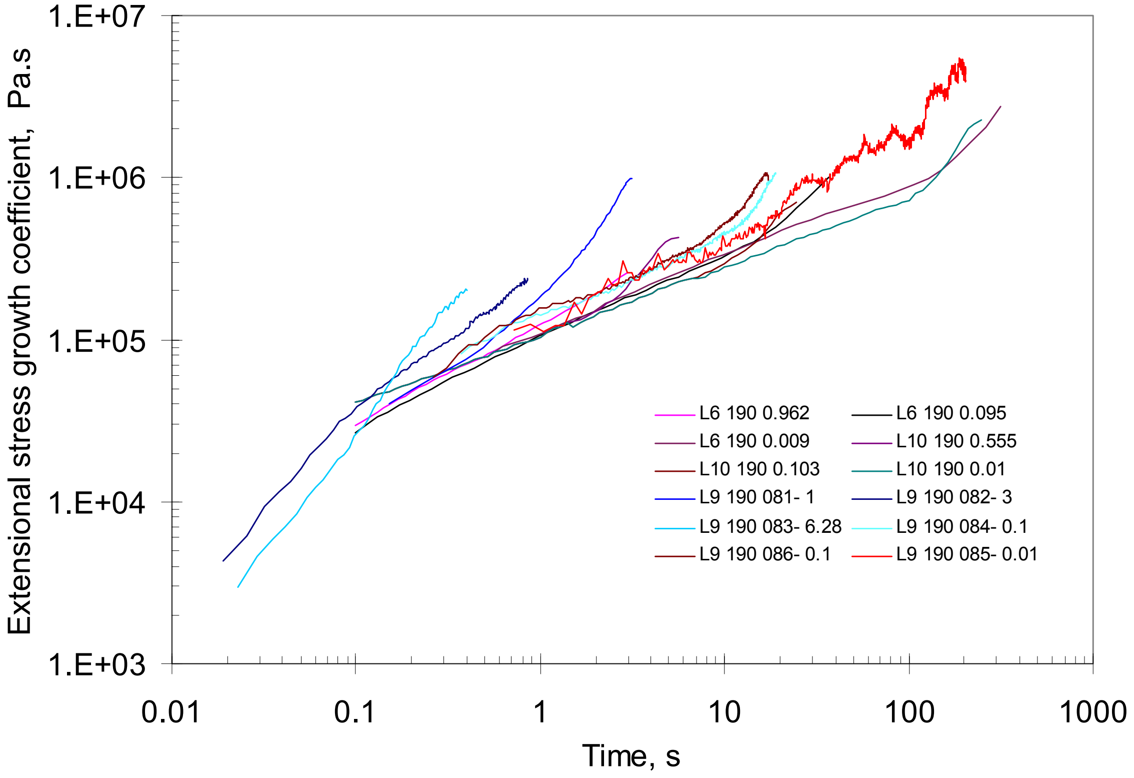

Figure 23: Intercomparison of stretching flow methods for a HDPE (HGH000) at 190 ºC: laboratory 3 data removed.

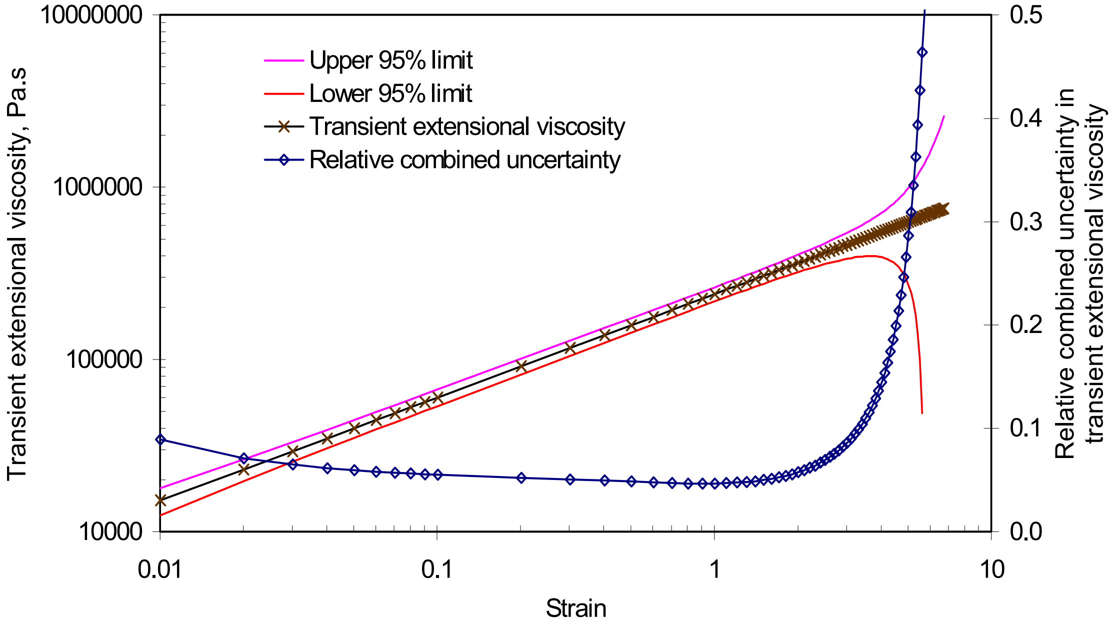

Figure 24: Analysis of the uncertainties in transient extensional viscosity values yielding 95% upper and lower confidence bands, based on a HDPE at 150 °C and 1 s^{-1}. The relative combined uncertainty values presented (right-hand y-axis) are for one standard deviation.

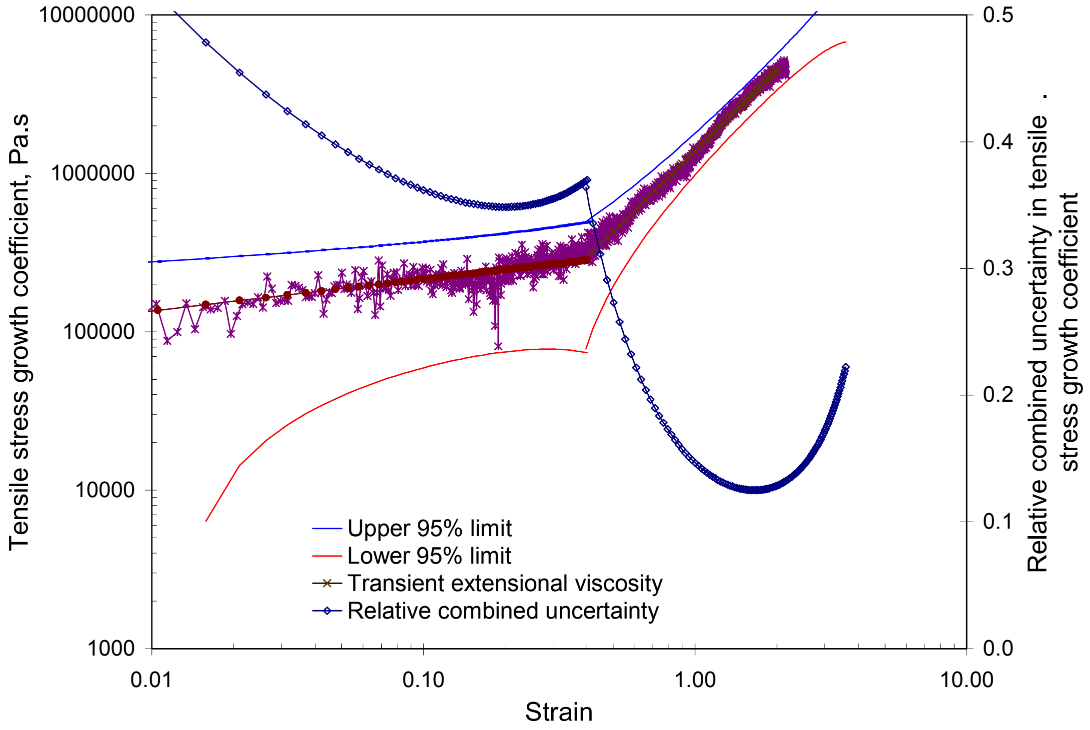

Figure 25: Assessment of the uncertainties in extensional viscosity when testing at low strain rates – HFU000 at 0.001 s^{-1} (test reference ext228).

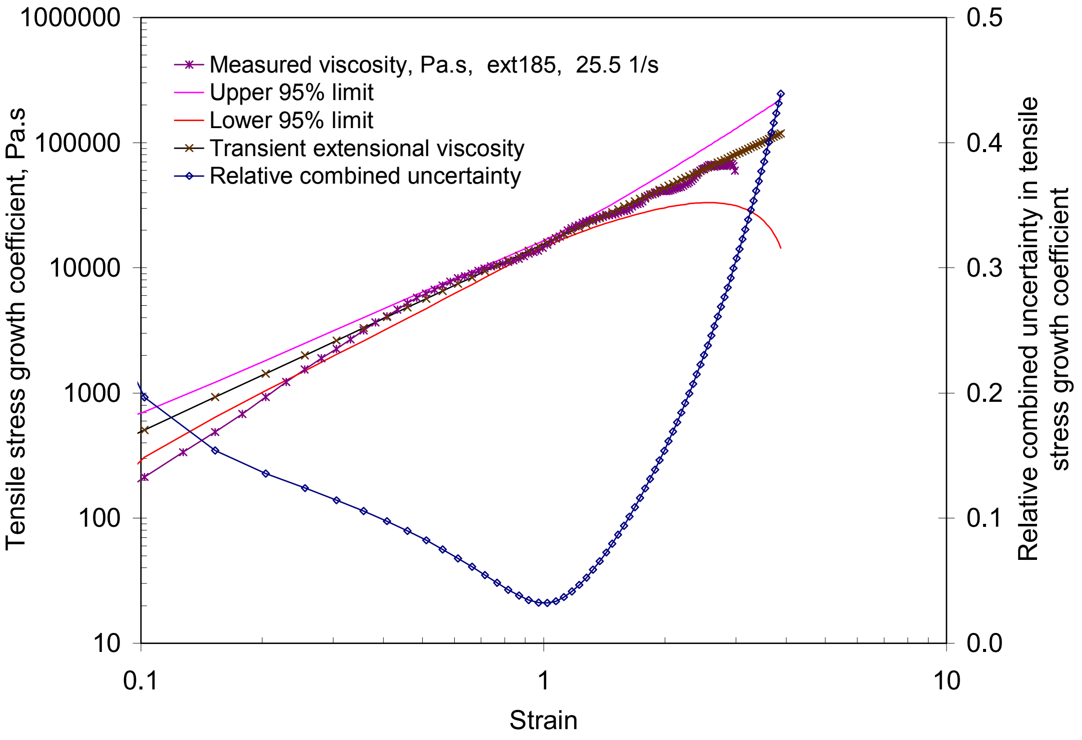

Figure 26: Assessment of the uncertainties in extensional viscosity when testing at high rates – HFU000 at 25.5 s^{-1} (test ref ext185).

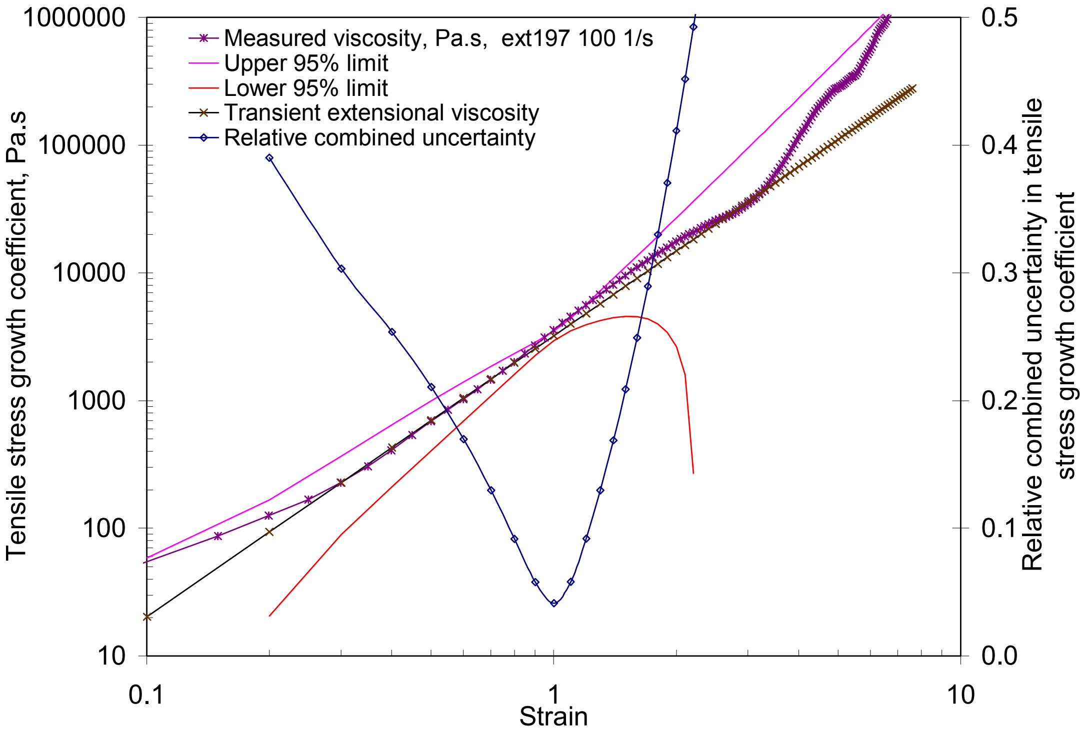

Figure 27: Assessment of the uncertainties in extensional viscosity when testing at high strain rates – HFU000 at 100 s^{-1} (test ref ext197).

Appendix A1 Definitions of strain, strain rate, stress and material properties functions in tensile (simple) extension

The following definitions are given by Whorlow [14] for strains and strain rates. Further descriptions are also given by, for example, Gupta *et al* [16] and Dealy [19, 23].

A1.1 Elongation ratio

The *Elongation ratio (ER)* is the ratio of the current length ℓ to the initial length ℓ_o of the specimen:

$$ER = \ell / \ell_o. \qquad (A1.1)$$

It is dimensionless.

A1.2 Cauchy ε_C and Hencky ε strains

The *Cauchy strain* ε_C is given by the ratio of the change in length $\delta\ell$ to the initial length ℓ_o of the specimen:

$$\varepsilon_C = \delta\ell / \ell_o. \qquad (A1.2)$$

The *Hencky strain* ε (also referred to as the natural or true strain) is given by the natural logarithm of the elongation ratio:

$$\varepsilon = \ln(\ell / \ell_o) \qquad (A1.3)$$

Thus the elongation ratio is related to the Cauchy strain by

$$ER = 1 + \varepsilon_C \qquad (A1.4)$$

i.e. $\ell / \ell_o = 1 + \delta\ell / \ell_o.$ (A1.5)

To illustrate the difference between these measures of strain a specimen that was stretched to 10 times its original length has an elongation ratio of 10, a Cauchy strain of 9 and a Hencky strain of 2.3. These strains are dimensionless.

A1.3 Cauchy $\dot{\varepsilon}_c$ and Hencky $\dot{\varepsilon}$ strain rates

The *Cauchy strain rate* is given by

$$\dot{\varepsilon}_c = 1/\ell_o \times \partial\ell/\partial t \qquad (A1.6)$$

and the *Hencky strain rate* by

$$\dot{\varepsilon} = 1/\ell \times \partial\ell/\partial t \qquad (A1.7)$$

In describing and modelling plastics processing the Hencky strain is preferred as the rate of strain of an element of fluid within the flow is independent of its original length and is determined only from the velocity field of that element. It is thus a more suitable characteristic of the flow.

These strain rates have units of reciprocal seconds (s^{-1}).

A1.4 Extensional flows: velocity fields

Terminology and definitions for materials functions describing the response of viscoelastic fluids to various shearing and extensional deformations are presented by Dealy [22]. Mathematical descriptions of extensional flows were presented, for example, by Meissner *et al* [79] and Walters [15].

For *tensile (simple or uniaxial) extension* where $\dot{\varepsilon}$ is the true strain rate and ε is the true strain, defined as

$$\varepsilon = \ln(\ell / \ell_o) \qquad \text{(A1.8)}$$

then in rectangular coordinates:

$$v_1 = \dot{\varepsilon}\ x_1 \qquad \text{(A1.9)}$$

$$v_2 = -\tfrac{1}{2}\dot{\varepsilon}\ x_2 \qquad \text{(A1.10)}$$

$$v_3 = -\tfrac{1}{2}\dot{\varepsilon}\ x_3 \qquad \text{(A1.11)}$$

where $\dot{\varepsilon} \geq 0$.

Alternatively, in cylindrical coordinates:

$$v_z = \dot{\varepsilon}\ z \qquad \text{(A1.12)}$$

$$v_r = -\tfrac{1}{2}\dot{\varepsilon}\ r \qquad \text{(A1.13)}$$

where $\dot{\varepsilon} \geq 0$.

A1.5 Material properties

Following the notation presented by Dealy [22], and prepared by the Nomenclature Committee of the Society of Rheology, for start-up flow in tensile (simple) extension at constant (Hencky) strain rate $\dot{\varepsilon}$ the following definitions are given. Equivalent expressions for cessation of steady tensile extension, tensile creep, tensile recoil and tensile step strain are presented by Dealy [22].

A1.5.1 Net tensile stress

The *net tensile stress* σ_E is defined by

$$\sigma_E = \sigma_{11} - \sigma_{22} = \sigma_{11} - \sigma_{33} = \sigma_{zz} - \sigma_{rr} \qquad \text{(A1.14)}$$

where σ_{ii} is a stress tensor in either rectangular or axisymmetric co-ordinates. It has units of pascal (equivalent to N/m^2). The tensile stress growth function is indicated by σ_E^+ where the + indicates start-up rather than cessation of flow.

A1.5.2 Tensile stress growth coefficient

The *tensile stress growth coefficient* η_E^+ is defined by:

$$\eta_E^+(t, \dot{\varepsilon}) = \sigma_E / \dot{\varepsilon} \qquad \text{(A1.15)}$$

where t is time. It has units of pascal.seconds (Pa.s).

A1.5.3 Tensile viscosity

The *tensile viscosity* η_E is defined by:

$$\eta_E(t, \dot{\varepsilon}) = \lim_{t \to \infty} [\eta_E^+(t, \dot{\varepsilon})] \qquad \text{(A1.16)}$$

It is the limiting tensile stress growth coefficient value and represents an equilibrium extensional viscosity if a steady value is achieved. However for materials that do not exhibit a steady state behaviour the use of an ‘equilibrium extensional viscosity’ such as this is obviously not appropriate.

Appendix A2 Analysis of extensional rheometer testing

A2.1 Definitions and terminology

ν - velocity of separation of the ends of the specimen, $m.s^{-1}$

ω - angular velocity of drum, $rad.s^{-1}$

r - drum radius, m

t - time, s

ℓ_o - original specimen length, m

ℓ - specimen length at time t, m

A_o - original specimen cross-sectional area, m^2

A - specimen cross-sectional area at time t, m^2

F - force, N

ε - Hencky strain

$\dot{\varepsilon}$ - Hencky strain rate, s^{-1}

σ_E - net tensile stress, Pa

η_E^+ - tensile stress growth coefficient, Pa.s

The Hencky strain and strain rate and the tensile stress growth coefficient are defined in Appendix A1.

A2.2 Analysis of extensional flow

Rearranging the Hencky strain

$$\varepsilon = \ln\left(\frac{\ell}{\ell_o}\right) \tag{A2.1}$$

yields

$$\ell = \ell_o \, e^{(\varepsilon)} \tag{A2.2}$$

or, for constant Hencky strain rate $\dot{\varepsilon}$,

$$\varepsilon = \dot{\varepsilon}\, t \tag{A2.3}$$

where t is time. Thus

$$\ell = \ell_o \, \mathrm{e}^{(\dot{\varepsilon} t)} \tag{A2.4}$$

Assuming conservation of volume of the specimen then

$$\ell_o A_o = \ell A \tag{A2.5}$$

The tensile stress, expressed as the ratio of the force to area,

$$\sigma_E = \frac{\mathrm{F}}{\mathrm{A}} \tag{A2.6}$$

can thus be rewritten, using Equation A2.5, as

$$\sigma_E = \frac{\mathrm{F}\ell}{\mathrm{A}_o \ell_o} \tag{A2.7}$$

Substitution for ℓ using Equations A2.2 and A2.4 yields

$$\sigma_E = \frac{\mathrm{F\,e}^{(\varepsilon)}}{\mathrm{A}_o} \tag{A2.8}$$

or

$$\sigma_E = \frac{\mathrm{F\,e}^{(\dot{\varepsilon} t)}}{\mathrm{A}_o} \tag{A2.9}$$

respectively. Thus the tensile stress growth coefficient, the ratio of tensile stress to Hencky strain rate, is given by

$$\eta_E^{+} = \frac{\mathrm{F\,e}^{(\varepsilon)}}{\mathrm{A}_o \dot{\varepsilon}} \tag{A2.10}$$

or

$$\eta_E^{+} = \frac{\mathrm{F\,e}^{(\dot{\varepsilon} t)}}{\mathrm{A}_o \dot{\varepsilon}} \tag{A2.11}$$

A2.3 Analysis of the deformation in extensional rheometers: Type A

The following is an analysis of testing carried out using the NPL rheometer (Type A in Figure 1). This analysis does not assume steady state behaviour. Analysis of flow in other configurations can be similarly derived. For the specimen the effective length is given by the separation between the clamps which in the case of the NPL instrument is between the fixed clamp and the point at which a line from the fixed clamp meets the drum at a tangent (ℓ_o in Figure 1). The effective specimen length in the test region remains unchanged during the test as the specimen is wound onto the drum. For the extensional rheometer using a single rotating drum the instantaneous velocity of separation of the ends of the specimen, ν, is given by

$$\nu = \omega r \qquad \text{(A2.12)}$$

where ω is the angular speed of the rotating clamp and r is its radius. The velocity of separation can also be written as

$$\nu = \partial \ell / \partial t \qquad \text{(A2.13)}$$

Thus, by using the definition of Hencky strain rate

$$\dot{\varepsilon} = 1/\ell \times \partial \ell / \partial t \qquad \text{(A2.14)}$$

the strain rate in the specimen is given by

$$\dot{\varepsilon} = \frac{\omega r}{\ell_o} \qquad \text{(A2.15)}$$

This assumes that the length ℓ_o is constant. The specimen length will change with force due to the compliance of the force transducer but this was considered to be minimal for the configuration used.

Integrating Equation A2.15 with respect to time yields the strain

$$\varepsilon = \int_0^t \dot{\varepsilon}\ \mathrm{d}t \qquad \text{(A2.16)}$$

Thus

$$\varepsilon = \frac{\omega r t}{\ell_o} \qquad \text{(A2.17)}$$

Thus the tensile stress is given, using Equation A2.8, by

$$\sigma_E = \frac{F\, e^{\left(\frac{\omega r t}{\ell_o}\right)}}{A_o} \qquad \text{(A2.18)}$$

and the tensile stress growth coefficient, using Equation A2.10, by

$$\eta_E^{+} = \frac{F\, \ell_o\, e^{\left(\frac{\omega r t}{\ell_o}\right)}}{A_o\, \omega\, r} \qquad \text{(A2.19)}$$

A2.4 Analysis: Type B

For the experimental configuration in which there are two sets of rotating clamps, each having the same angular speed ω (Type B, Figure 1), then the analysis presented for type A is valid except that

$$v = 2\omega r \tag{A2.20}$$

Thus the strain rate is given by

$$\dot{\varepsilon} = \frac{2\omega r}{\ell_o} \tag{A2.21}$$

and the strain by

$$\varepsilon = \frac{2\omega rt}{\ell_o} \tag{A2.22}$$

The net tensile stress is given by equation (A2.9) and the tensile stress growth coefficient by equation (A2.11), substituting for strain rate as appropriate.

A2.5 Analysis: Type C

In the case of a single translating clamp (Type C, Figure 1), to obtain a constant strain rate $\dot{\varepsilon}$ then the speed of the clamp V, where

$$V = \frac{\partial \ell}{\partial t} \tag{A2.23}$$

is given, using equation (A2.4), by

$$V = \ell_o \dot{\varepsilon}\ \mathrm{e}^{(\dot{\varepsilon}\, t)} \tag{A2.24}$$

where ℓ_0 is the original specimen length. For a specified constant strain rate, the strain is given by equation (A2.3), the net tensile stress by equation (A2.9) and the tensile stress growth coefficient by equation (A2.11).

A2.6 Analysis: Type D

In the case of using two clamps moving with the same speed (Type D, Figure 1), to obtain a constant strain rate $\dot{\varepsilon}$ then the speed of the clamps V, where

$$2V = \frac{\partial \ell}{\partial t} \tag{A2.25}$$

is given, using equation (A2.4), as

$$V = \frac{\ell_0 \dot{\varepsilon}\ e^{(\dot{\varepsilon} t)}}{2} \tag{A2.26}$$

where ℓ_o is the original specimen length. For a specified constant strain rate, the strain is given by equation (A2.3), the net tensile stress by equation (A2.9) and the tensile stress growth coefficient by equation (A2.11).

Appendix A3 Analysis of the uncertainties of measurement of transient extensional viscosity

A3.1 Introduction

Due to the considerable stretching that the specimen normally undergoes during testing, particularly on reaching high strain values, there is a very significant decrease in the cross-sectional area of the specimen, accompanied by a similarly large decrease in the measured force. As a consequence there is a significant increase in the uncertainty of the measurements. To understand and quantify the level of confidence that one can put in the accuracy of the results obtained, it is important that the uncertainties in the measurements, in particular that due to the accuracy and resolution of the force measurement, are known.

An example analysis is presented of the uncertainties in transient extensional viscosity measurements for an extensional rheometer having a single rotating clamp and a fixed clamp, Type A. The equations can be used to calculate the uncertainties in the derived tensile stress growth coefficient values, given the uncertainties in each of the components of the measurement. Similar analyses can be carried out for other instrument configurations by following the principles presented. For further details see [6].

A3.2 Uncertainty analysis

Following a more rigorous approach, used for example by Kandil (2), the combined uncertainty $u_c(y)$ of the measurand y (the quantity to be measured) can be determined from the partial derivatives of the function and the uncertainties in the parameters. Assuming that individual uncertainty sources are uncorrelated, the combined uncertainty $u_c(y)$ can be computed using the root sum squares:

$$u_c(y) = \sqrt{\sum_{i=1}^{m} [c_i u(x_i)]^2} \qquad \text{(A3.1)}$$

where c_i is the sensitivity coefficient (partial derivative) associated with an input quantity x_i and $u(x_i)$ is the uncertainty in that quantity.

The combined uncertainty $u_c(y)$ corresponds to one standard deviation and therefore has an associated confidence level of approximately 68%. Assuming a normal distribution then it is assumed that an expanded uncertainty U for 95% confidence level can be determined using a coverage factor of 2 (i.e. equivalent to 2 standard deviations). The relative uncertainty is the ratio of the uncertainty in the parameter to the value of the parameter.

A3.3 Uncertainty in strain rate

The strain rate $\dot{\varepsilon}$ in the specimen is given (Appendix 2) by

$$\dot{\varepsilon} = \frac{\omega \mathrm{r}}{\ell_o} \tag{A3.2}$$

where ω is the angular speed of the winding drum, r is the radius of the winding drum and ℓ_o is the original specimen length. The sensitivity coefficients, or partial derivatives, of the strain rate are thus

$$\frac{\partial \dot{\varepsilon}}{\partial \omega} = \frac{\mathrm{r}}{\ell_\mathrm{o}} \tag{A3.3}$$

$$\frac{\partial \dot{\varepsilon}}{\partial r} = \frac{\omega}{\ell_\mathrm{o}} \tag{A3.4}$$

and

$$\frac{\partial \dot{\varepsilon}}{\partial \ell_\mathrm{o}} = \frac{-\omega r}{\ell_\mathrm{o}^{2}} \tag{A3.5}$$

In what follows, a subscripted u denotes an uncertainty in the subscripted quantity. Thus, using equation A3.1, the combined uncertainty in strain rate $u_{\dot{\varepsilon}}$ is given by the root sum of the squares of the product of the partial derivatives and the corresponding uncertainties

$$(u_{\dot{\varepsilon}})^2 = \left(\frac{r}{\ell_o} u_\omega\right)^2 + \left(\frac{\omega}{\ell_o} u_r\right)^2 + \left(\frac{-\omega r}{\ell_o^{2}} u_{\ell_o}\right)^2 \tag{A3.6}$$

and the combined relative uncertainty in strain rate is given, by dividing through by $\dot{\varepsilon}$, by

$$\left(\frac{u_{\dot{\varepsilon}}}{\dot{\varepsilon}}\right)^2 = \left(\frac{u_\omega}{\omega}\right)^2 + \left(\frac{u_r}{r}\right)^2 + \left(\frac{u_{\ell_o}}{\ell_o}\right)^2 \tag{A3.7}$$

A3.4 Uncertainty in strain

The strain ε is given (Appendix 2) by

$$\varepsilon = \frac{\omega \mathrm{rt}}{\ell_o} \tag{A3.8}$$

where t is time. Thus, in a similar fashion to that for strain rate, the combined relative uncertainty in strain is given by

$$\left(\frac{u_\varepsilon}{\varepsilon}\right)^2 = \left(\frac{u_\omega}{\omega}\right)^2 + \left(\frac{u_r}{r}\right)^2 + \left(\frac{u_t}{t}\right)^2 + \left(\frac{u_{\ell_o}}{\ell_o}\right)^2 \tag{A3.9}$$

A3.5 Uncertainty in transient extensional viscosity

Given the expression for transient extensional viscosity η_E (Appendix 2):

$$\eta_E = \frac{\mathrm{F}\ell_{\mathrm{o}} e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{A_o \omega\, r} \tag{A3.10}$$

then the sensitivity coefficients c_i are given by

$$\frac{\partial \eta_E}{\mathrm{dF}} = \frac{\ell_{\mathrm{o}} e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{A_o \omega\, r} \tag{A3.11}$$

$$\frac{\partial \eta_E}{\partial \mathrm{A_o}} = \frac{-F\ell_{\mathrm{o}} e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{A_o{}^2 \omega\, r} \tag{A3.12}$$

$$\frac{\partial \eta_E}{\partial\, \mathrm{r}} = \frac{F\ell_o}{A_o \omega}\left(\frac{\text{-}e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{r^2} + \frac{\omega\,\mathrm{t}\, e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{\ell_{\mathrm{o}} r}\right) \tag{A3.13}$$

$$\frac{\partial \eta_E}{\partial\, \omega} = \frac{F\ell_o}{A_o r}\left(\frac{\text{-}e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{\omega^2} + \frac{\mathrm{r\,t}\, e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{\ell_{\mathrm{o}} \omega}\right) \tag{A3.14}$$

$$\begin{aligned}\frac{\partial \eta_E}{\partial\, \ell_{\mathrm{o}}} &= \frac{F}{A_o \omega\, r}\left(e^{\left(\frac{\varpi \mathrm{r\,t}}{\ell \mathrm{o}}\right)} + \frac{\ell_o \omega\,\mathrm{r\,t}\, e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{-\ell_o^2}\right) \\ &= \frac{F}{A_o \omega\, r}\left(e^{\left(\frac{\varpi \mathrm{r\,t}}{\ell \mathrm{o}}\right)} - \frac{\omega\ \mathrm{r\,t}\, e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{\ell_{\mathrm{o}}}\right)\end{aligned} \tag{A3.15}$$

and

$$\frac{\partial \eta_E}{\partial\, \mathrm{t}} = \frac{F e^{\left(\frac{\omega \mathrm{r\,t}}{\ell \mathrm{o}}\right)}}{A_o} \tag{A3.16}$$

The combined relative uncertainty in the tensile stress growth coefficient values is then evaluated as

$$\left(\frac{\mathrm{u}_{\eta_E}}{\eta_E}\right)^2 = \left(\frac{\partial \eta_E}{\partial \mathrm{F}}\frac{u_{\mathrm{F}}}{\eta_E}\right)^2 + \left(\frac{\partial \eta_E}{\partial A_o}\frac{\mathrm{u}_{\mathrm{A_o}}}{\eta_E}\right)^2 + \left(\frac{\partial \eta_E}{\partial \mathrm{r}}\frac{\mathrm{u_r}}{\eta_E}\right)^2 + \left(\frac{\partial \eta_E}{\partial \omega}\frac{\mathrm{u}_{\omega}}{\eta_E}\right)^2 + \left(\frac{\partial \eta_E}{\partial \ell_{\mathrm{o}}}\frac{\mathrm{u}_{\ell_{\mathrm{o}}}}{\eta_E}\right)^2 + \left(\frac{\partial \eta_E}{\partial \mathrm{t}}\frac{\mathrm{u_t}}{\eta_E}\right)^2 \tag{A3.17}$$

where the partial derivatives are given in Equations A3.11 to A3.16.

A3.6 Evaluation of uncertainty in transient extensional viscosity measurements

The values of the terms in the above uncertainty budgets, except for the force terms, are relatively easy to determine. The difficulty with the force term is that the force decreases considerably at high strain values and thus the relative uncertainty in the force will increase significantly as the test progresses. To determine the effect of its contribution to the combined uncertainty in the determination of the transient extensional viscosity values further analysis, presented below, is required.

The force F supported by the specimen during testing is given by

$$F = \sigma \ A \qquad \text{(A3.18)}$$

where A is the cross-sectional area of the specimen and σ is the tensile stress. Using the definition of transient extensional viscosity η_E^+ (or tensile stress growth coefficient), Appendix 1, then

$$\sigma = \eta_E{}^+ \dot{\varepsilon} \qquad \text{(A3.19)}$$

Thus the force

$$F = \eta_E{}^+ \dot{\varepsilon} \ A \qquad \text{(A3.20)}$$

or, using Equations A2.2, A2.5 and A2.17 of Appendix 2,

$$F = \eta_E{}^+ \dot{\varepsilon} \ A_o e^{\left(\frac{-\omega r t}{\ell_o}\right)} \qquad \text{(A3.21)}$$

Assuming that the transient extensional viscosity as a function of time can be fitted reasonably well using a power law model (see Figure 8 of reference 1) then the equation of a straight line fit to the data is of the form

$$\log(\eta_E{}^+) = m \log(t) + \log\left(\eta_{E,o}{}^+\right) \qquad \text{(A3.22)}$$

where $\eta_E{}^+ = \eta_{E,o}{}^+$ at t = 1 s and $\eta_{E,o}{}^+$ and m are constants. Equation A3.22 can be rewritten as

$$\eta_E{}^+ = \eta_{E,o}{}^+ \ t^m \qquad \text{(A3.23)}$$

The force over the test duration is thus approximated by

$$F = \eta_{E,o}{}^+ \ t^m \ \dot{\varepsilon} \ A_o e^{\left(\frac{-\omega r t}{\ell_o}\right)} \qquad \text{(A3.24)}$$

Given the values of $\eta_{E,o}{}^{+}$ and m, the force F can be determined as a function of time and, assuming that all strain rate data fall on the same master curve, also as a function of strain if one ignores any additional strain hardening phenomenon. Thus the relative uncertainty in the force F, given the decreasing magnitude of force at high strains, can be reasonably accurately determined. However the combined uncertainty in transient extensional viscosity values must be quoted as a function of time rather than as a single value.

Analysis is presented based on the case of a high density polyethylene HDPE (HGH000) at 150 °C and a strain rate of ≈ 1 s^{-1}, Table 1 and Figure 10. The transient extensional viscosity values are plotted along with 95% confidence limits derived using Equation A3.17 and a coverage factor of 2, Figure 24. It clearly illustrates the significant increase in measurement uncertainties at high strains due, predominantly, to the increase in the uncertainty in the measurement of force resulting from the significant reduction in the cross-sectional area of the specimen at high strain values. Expanded uncertainties representing 95% confidence limits were estimated to be of the order of ± 20% at a strain of 3.3, ± 50% at a strain of 4.8 and ± 100% at a strain of 5.7. The initial decrease in uncertainty, up to a strain of 1, is due to the low forces being generated in the specimen at the start of the test resulting in a high relative uncertainty for force.

Table 1: Example values and uncertainties in the extensional testing parameters

$\eta_{E,o}{}^{+}$ = 2.4 x10^5 Pa.s and m = 0.6 estimated for an HDPE (HGH000) at 150 ^{o}C at a strain rate of ≈ 1 s^{-1}.

Quantity	Symbol	Units	Type	Probability distribution	Divisor	Quantity value x	Quantity range, ±	Standard uncertainty u_x	Relative uncertainty u_x/x
Initial specimen diameter 1	d_1	m	A	Normal	1	0.0033	0.0001	0.0001	0.030
Initial specimen diameter 2	d_2	m	A	Normal	1	0.0033	0.0001	0.0001	0.030
Force	F	N	B	Rectangular	$\sqrt{3}$	variable	0.016	0.0092	variable
Effective specimen length	ℓ_o	m	B	Rectangular	$\sqrt{3}$	0.1	0.003	0.0017	0.017
Angular speed	ω	rad/s	B	Rectangular	$\sqrt{3}$	5.0	0.015	0.0087	0.0017
Drum radius	r	m	B	Rectangular	$\sqrt{3}$	0.02	0.00075	0.00043	0.022
Time	t	s	B	Rectangular	$\sqrt{3}$	variable	0.024	0.014	variable
Initial area*	A_o	m^2	A	Normal	1	8.6 x 10^{-6}	3.7 x 10^{-7}	3.7 x 10^{-7}	0.043

Note: * Indicates calculated values based on the assumption of independent orthogonal measurements of the specimen.
The standard uncertainty is obtained by dividing the quantity range by the divisor.

Appendix A4 Effect of viscous dissipation in the specimen during stretching on measurements

It is assumed, for the purposes of this calculation, that the force exerted on the specimen F for the duration of the test is constant and is equal to the maximum force exerted on the specimen. Furthermore the specimen is of initial length ℓ_o, initial radius R_o and undergoes a strain ε in a Type C or D configuration (Figure 1). The final specimen length is given (Equation A2.2) by

$$\ell = \ell_o e^{\varepsilon} \qquad \text{(A4.1)}$$

Thus the work done W_F is given by

$$W_F = (\ell_o e^{\varepsilon} - \ell_o)F \qquad \text{(A4.2)}$$

The work done per unit volume of the specimen is thus

$$w_F = \frac{(\ell_o e^{\varepsilon} - \ell_o)F}{\pi\ R_o^{\ 2}\ell_o} \qquad \text{(A4.3)}$$

Assuming the thermal behaviour

$$w_\theta = \rho\ C_p \Delta\theta \qquad \text{(A4.4)}$$

where ρ is the density, C_p is the specific heat capacity and Δθ is the temperature rise in the specimen then, given the energy balance

$w_F = w_\theta$,

and thus

$$\Delta\theta = \frac{(\ell_o e^{\varepsilon} - \ell_o)F}{(\rho C_p)(\pi\ R_o^{\ 2}\ell_o)} \qquad \text{(A4.5)}$$

Using the values based on the HDPE (HGH000), Figures 5,

$F = 1$ N
$\ell_o = 0.1$ m
$R_o = 1.5 \times 10^{-3}$ m
$\varepsilon = 3$
$\rho = 900$ kg/m^3
$C_p = 2300$ J/(kg.K)

then the temperature rise Δθ is estimated to be ≈ 1.3 °C.

This estimate assumes that the force was constant over the duration of the test, that all the energy was dissipated as heat and that no heat was dissipated from the specimen. These assumptions are all invalid. For example, Figure 5 clearly shows that although the peak force was approximately 1 N the average force would have been lower. Also no account of the energy stored elastically in the specimen or heat dissipated from the specimen was made. All of these factors would result in a reduction of the temperature increase. For a HDPE (HGH000) the shear flow temperature dependence was estimated to be less than 1 %/^{o}C, and the extensional flow properties were of the same order [74]. As the dissipation would be evenly distributed through the specimen the effect on measurements is considered to be negligible.

Although a C or D type configuration was used in this analysis, for ease of visualisation of the problem and simplicity of the analysis, the predicted temperature rise will be the same for other configurations where the same strain rate, strain and force values are used.

For other materials and testing conditions the temperature rise may not be so insignificant. A more accurate calculation of the temperature rise can be carried out by summing the work done, i.e. force x displacement for increments in specimen length, over the test duration using the force and length equations, Equations A3.24 and A4.1 respectively.