

Review of extensional viscoelasticity measurement techniques for polymer melts

M. Rides, C.R.G. Allen and S. Chakravorty
Centre for Materials Measurement and Technology
National Physical Laboratory
Teddington, Middlesex
United Kingdom
TW11 0LW

ABSTRACT

In many forming processes the polymer melt undergoes predominantly extensional deformation, for example in blow moulding and film blowing. Thus extensional flow measurement methods are necessary to characterise most appropriately the melt behaviour. Such data can be used for the development and selection of materials and for improving processing. A critical review of techniques for measuring the extensional viscoelastic behaviour of polymer melts is presented. The methods reviewed principally cover stretching flow methods in which a specimen is stretched between 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°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 ≈ 4 , strain rates up to $\approx 1 \text{ s}^{-1}$ and temperatures up to $\approx 200^\circ\text{C}$ but such equipment is still of limited availability. The more important aspects of instrument design for measuring extensional viscoelastic properties of polymer melts at conditions above these more typical limits were identified. Limitations in measurement capability due to instrument design and also inhomogeneity of specimen deformation were highlighted. The latter may be due to inadequate specimen preparation and/or non-uniformity of specimen temperature.

© Crown copyright 1996
Reproduced by permission of the Controller of HMSO

ISSN 1361-4061

National Physical Laboratory
Teddington, Middlesex, UK, TW11 0LW

Extracts from this report may be reproduced provided the source is acknowledged.

Prepared as a deliverable of milestone 3 of the project
MMP11: Measurement of extensional viscoelastic properties of polymers,
a project of the Department of Trade and Industry EAM programme
on measurements related to the processability of materials

Approved on behalf of Managing Director, NPL
by Dr M K Hossain, Director, Centre of Materials Measurement and Technology

CONTENTS

1	INTRODUCTION	1
2	SURVEY OF EXTENSIONAL TECHNIQUES	1
2.1	General	1
2.2	Industrial requirements	2
2.3	Historical background and developments on related materials	3
2.4	Classification of techniques	4
2.5	Discussion and comparison of uniaxial stretching techniques	4
2.6	Materials	7
2.7	Specimen preparation	8
2.8	Clamping...	9
2.9	Temperature control and testing medium	10
2.10	Strains, strain rates and stresses of testing	13
2.11	Other extensional flow measurement techniques and considerations	15
3	DISCUSSION	16
4	CONCLUSIONS.	17
5	ACKNOWLEDGEMENTS	17
6	REFERENCES	18

Table 1: Summary specification of instruments for extensional viscoelasticity measurements presented in the literature

Figures

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

1 INTRODUCTION

This survey was carried out as part of the first stages of a project on measurement of the extensional viscoelastic properties of polymers (project reference MMP11, milestone 3). This project is one of a number of projects funded by the Engineering, Automotive and Metals Division (EAM) of the Department of Trade and Industry on measurements related to the processability of materials. This project aims at the development of methods for characterising the extensional viscoelastic behaviour of polymer melts. The first two specific objectives of the project are:

To review existing methods for measuring the parameters that describe the viscoelastic behaviour of polymeric materials under stress fields that are predominantly extensional,

- and to develop new methods, as required, that will allow measurements to be made at extensional strain rates comparable with those encountered during industrial processing.

The objective of the first stage of the project milestone M3 is to review existing techniques for extensional viscoelasticity measurements. This will help to establish the specification of the instrument that is to be developed at the NPL so that it can most appropriately and effectively meet the needs of UK industry.

This review of the literature targets the measurement of the uniaxial extensional properties of polymer melts. It commences with general information and comments on extensional flow measurements, including a summary of industrial requirements, a historical note and brief discussion of related non-polymer melt and non-uniaxial work [sections 2.1 - 2.3]. This is followed by a classification, general description and comparison of the various techniques used [2.4 - 2.5]. This is followed by a more detailed examination of the following topics: the materials investigated in the literature [2.6]; preparation of test specimens [2.7]; methods used for clamping [2.8]; methods used to control the temperature of the test specimen [2.9]; the effect of the supporting medium (typically silicone oil) on the polymer [2.9]; the control and/or measurement of strain, strain rate and stress [2.10]; and finally additional points on alternative methods and calibration of instruments [2.11]. Experimental investigations reported in the literature are tabulated in Table 1. Definitions of strains, strain rates, stress and material properties functions are presented in Appendix A1. The critical features of such instruments are then discussed in a broader context [3] and conclusions are finally drawn [4].

2 SURVEY OF EXTENSIONAL TECHNIQUES

2.1 GENERAL

This review of uniaxial extensional techniques for viscoelastic measurements of polymer melts addresses free surface methods, ie where a specimen is stretched without the constraint of a fixed surface or channel in contact with the specimen along its length. The constrained category of methods includes the commonly referred to "converging flow" or "contraction flow" techniques (see for example references 1 and 2). Constrained methods will be reviewed in a later report as part of the project MMP11. Although this review is principally of uniaxial extensional test methods for polymer melts, reference to other relevant aspects of extensional flow testing are made.

2.2 INDUSTRIAL REQUIREMENTS

The industrial needs for extensional viscoelasticity testing, specifically in terms of the strains, strain rates and temperatures were assessed by Rides (3). It was concluded that there was need for measurements typically up to strains of 2 and strain rates of up to 5 s^{-1} at temperatures of up to 260°C . Cogswell (4) summarised the industrial needs for measurement techniques in terms of their practicality, performance and cost. He commented that techniques that were then suitable for adoption by industry, specifically the fibre spinning (1, 5) and converging flow (1, 2) methods, were qualitative. There was a requirement for quantitative, accurate techniques that were practicable, that extended the range of measurements to strain rates greater than 1 s^{-1} and were suitable for materials of viscosity lower than 10^5 Pa.s . This review also addresses whether this position, expressed by Cogswell in 1972 (4), has changed.

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 (6) and it may not be possible to approximate it as a constant force process. The most relevant and appropriate measurements to make should be measurements of true materials properties (constant strain rate or constant stress) rather than those that produce results that are dependent on the measurement method (constant force or constant speed).

In contrast to shear flows where reference is normally made only to steady shear flow behaviour, extensional flow is normally described as a transient behaviour. The extensional viscosity may vary as a function of both strain (or time) 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* behaviour (for example reference 7, 8), or an *equilibrium extensional viscosity* value (typically at large strains, for example references 9, 10, 11), or a *maximum* in the stress value (8) - see Figure 1 and for definitions see Appendix A1. An equilibrium extensional viscosity is dependent on strain rate but not on strain or time. The observation of *equilibrium* or *maximum* values in extensional viscosity is discussed later with reference to the literature.

Petrie (12) makes the very valid point that industrial flows are rarely of pure elongation, long duration, constant strain rate and not affected by their flow history. Industrial processes tend to be transient extensional flows. Therefore he questioned the need to be able to determine the tensile (or equilibrium) extensional viscosity rather than, perhaps, the tensile stress growth behaviour (i.e. the change in stress with time for a constant strain rate). If the determination of equilibrium extensional viscosity values is difficult and expensive and the benefits of so doing are limited then the driving force to do so is also limited. However, in terms of modelling of flow processes there may be benefits in obtaining equilibrium extensional viscosity values to improve the process of selecting the most suitable constitutive equation and also for determining the values of the parameters of that equation (13).

2.3 HISTORICAL BACKGROUND AND DEVELOPMENTS ON RELATED MATERIALS

Extensional flow properties of materials were, according to Ballman (14), first studied by Trouton (15) in 1906 on pitch, tar and wax materials. It was observed that the extensional viscosity values were approximately three times the shear viscosity values, yielding what is now commonly referred to as the Trouton ratio. Research into the measurement of extensional flow properties was progressed in the early 1960s with work on PS reported by Ballman (14) in 1965. Significant advances of the study of the extensional properties of polymer melts were then made by Cogswell (16) in 1968 and Meissner (17) in 1969 with the development of new instruments. A period of activity followed in the 1970's (predominantly but not exclusively by Meissner, Münstedt and Laun) that was based largely on the instruments of Cogswell (16) and Meissner (17) or developments thereof.

Since the early work of Cogswell (16) and Meissner (17) several articles and reviews describing the various extensional viscoelastic methods for polymer melts have been written in both journals and texts (1, 4, 5, 8, 18-23). The reviews of Meissner (8, 20) focused in particular on the techniques developed by Meissner (7) and Münstedt (9).

Multiaxial extensional viscosity measurements of materials are not discussed here. The reader is referred to Meissner (8), Meissner et al (20, 24), Berger et al (25), Whorlow (21), Dealy (19) and Petrie (5) for further details.

A significant contribution to the understanding of extensional viscoelasticity measurements, albeit specifically addressing the measurement of polymer solutions, was made through recent international collaborations. The work on the measurement of the shear and extensional viscoelastic properties of three specific polymer solutions M1, A1 and S1 has been reported by various researchers:

M1 - 0.244% polyisobutylene in a mixed solvent of 7% kerosene in polybutene, exhibiting constant shear viscosity with a dominant relaxation time of 0.3 s (references 26 (26 papers) and 27);

A1 - 2% polyisobutylene in a mixture of cis- and trans-decalin, exhibiting shear thinning with a dominant relaxation time of 5 s (references 28 and 27); and

S1 - 2.5% polyisobutylene in a mixture of decalin and polybutene, exhibiting shear thinning with a dominant relaxation time similar to M1 (reference 29 (7 papers)).

The experimental methods that were used are described in the various papers and also by James et al (30) and Gupta et al (1). Comparison of results for the polymer solutions A1 (28) and M1 (30) clearly indicated substantial differences in extensional properties obtained using the different methods: the extensional viscosity varied by three decades over the two decades of strain rate with results presented over virtually the entire region. Petrie (12) commented that the discrepancy was because "the experimental methods were not reproducing the conditions equivalent to the formal definition of extensional viscosity" - ie the tests generated flow fields that were not well approximated by the mathematical descriptions used to interpret the experimental data. James et al (30) commented that the rheometers used in the study of the fluid M1 should be viewed as yielding "*an* extensional viscosity rather than *the* extensional viscosity". This volume of work has clearly highlighted some of the difficulties facing the measurement of extensional flow properties. The validity of these comments should be considered when such techniques are applied to polymer melts.

2.4 CLASSIFICATION OF TECHNIQUES

Extensional techniques can be described, following Nazem et al (31), as "controllable" or "non-controllable" (21, 22). "Controllable" implies that the instantaneous values of strain rate and stress are uniform throughout the specimen and that the strain rate or stress is held constant with time. The results of non-controllable methods, for example fibre spinning and converging flow analysis (1, 2, 5) tend to be difficult to interpret (19).

As described in the above section stretching techniques can be categorised as either: constant strain rate, constant stress, constant force or constant speed (constant change of length of the specimen per unit time). The first two are potentially "controllable" measurements whereas the latter two are "non-controllable". Constant force and constant speed measurements are difficult to interpret as they are neither constant stress nor constant strain (21). This is emphasised by Gupta et al (1) who commented that uniaxial stretching methods are the preferred methods for obtaining fundamental data but can only be performed on very viscous polymers and at low strain rates using constant strain rate or constant stress, but not constant force or speed methods. Cogswell (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.

Within each of the above categories further subcategories or "types" are chosen here for convenience to describe the various techniques, Figure 2. References to these "types" are made below and in Table 1.

- Type 1: Two rotating clamps.
(Each clamp consists of either a single or pair of rotating elements. Clamps may be smooth or textured.)
- Type 2: Single rotating clamp and 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 3: Moving (i.e translating, non-rotating) clamp(s).
(Where a single moving clamp is used the other clamp is fixed in position.)
- Type 4: Haul-off or fibre spinning from an extruder.

2.5 DISCUSSION AND COMPARISON OF UNIAXIAL STRETCHING TECHNIQUES

The principal requirement for extensional measurements is for methods that provide true material properties data at high values of strain, strain rate and temperature. The difficulties that this requirement poses are now addressed.

General limitations of stretching techniques

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 (1) commented that the major difficulties in using instruments for constant strain rate measurements are due to temperature and stretch rate uniformity and also gravitational effects.

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 (16) commented that the constant stress method is suited to melts with viscosities greater than 10^5 Pa.s. Rhi-Sausi et al (32) reported that improvements to this type of instrument were made but did not quantify the magnitude of those improvements. Gupta et al (1) 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 (1). Petrie (5) 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 (1) 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 (1) 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 latest development by Meissner et al (33) 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 (33) was a type 1 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 fitting 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.

Instruments of type 1, 2 or 3 may be "controllable" or "non-controllable", whereas those of type 4 are "non-controllable". Because of the requirement for true materials properties type 4 methods are less desirable although they have specific advantages over the other types. For this reason they are largely considered separately below.

The most fundamental difference between types 1, 2 and 4 and type 3 is that the strains achievable using type 3 instruments are limited by the physical sizes of the instrument and the specimen length (19). For types 1, 2 and 4 the gauge length is constant and the strain rate is proportional to the take-up speed of the clamp(s). 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 (4) commented that translating clamp techniques (type 3) are limited to maximum strains of approximately 2. However, since then strains up to a value of 4 have been achieved using such methods (see Table 1, references 34, 35). Higher strain values cannot simply be obtained using type 3 devices by employing shorter initial specimen lengths as end-effects may become more significant. This factor is addressed in detail in section 2.8. 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 (proportional to the cross sectional area of the specimen) thus increasing errors in the measurement. To obtain higher strain values, up to a maximum value of 7, type 1 and 2 instruments with rotating clamps have been successfully used (see Table 1, references 10, 33, 34, 36, 37).

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 (5), 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 (36) 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 - 6 beyond which the stress decreased. Münsterdt et al (34) 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.

Cogswell (4) 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 transitional behaviour of the material. Axtell et al (38) 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 (38) added that transient data from constant strain rate tests are often more representative of deformations occurring in polymer processing and therefore more desirable. It is clear that if constant strain rate methods are to be used then strains greater than 4 are required to fully characterise the material, including its steady state behaviour if it exists.

Meissner et al (36) commented that an important part of testing was the separation of the total deformation into its elastic (recoverable) and inelastic (irrecoverable) components. This can be obtained by cutting the specimen at the end of deformation and measuring the recovery (8). He commented that the rotary clamp is an important element in the versatility and usefulness of extensional rheometers. It is noted however that the use of rotating clamps is not a pre-requisite to being able to carry out stress relaxation or strain recovery measurements.

One of the more striking recent advances in techniques is that presented by Leitlands (39) who developed an instrument, along the lines of the type 1 instrument of Meissner (7, 8), that can generate an unsteady extensional flow comprising a periodic deformation superimposed onto a constant strain rate deformation. A type 2 unsteady extensional device has since been developed by Tanaka et al (40) along similar lines to that of Leitlands (39). These instruments demonstrate that through the use of microprocessor control of motors any feasible stress or strain rate function ($\sigma > 0, \dot{\epsilon} > 0$) can be imposed.

Melt spinning techniques

Isothermal and non-isothermal spinning measurements of both polymer melts and solutions are reviewed in detail by Petrie (5) and more briefly by Cogswell (4) and others (1, 21, 23).

The spinning technique, for example using a haul-off device beneath an extrusion rheometer, is neither of homogeneous deformation (4, 19) nor normally isothermal although isothermal measurements have been carried out (41). It is, in terms of the definition presented above, "non-controllable". The strain rate increases in moving along the spin-line (11) and is not purely extensional near the exit of the extruder die (11). The authors commented that results from such tests are not expected to be quantitatively correct. However Gupta et al (1) commented that such a technique can be used to measure melts of lower viscosities than by type 1, 2 and 3 stretching techniques.

The advantages and disadvantages of the spinning experiment are presented in detail by Revenu et al (41). They commented that type 1 and 3 instruments in which the specimen is extended under constant strain rate or stress are very expensive, difficult to operate and not suitable for all polymers, especially those with low viscosities. They commented that fibre

spinning is more suitable for such materials, but acknowledged that the properties data obtained from fibre spinning will at best be approximations of true materials properties. The flow is inhomogeneous and is complicated by extrudate swell and molecular orientation in the die entry region (41). The authors commented that spinning is widely used only for qualitative evaluation of the drawability of polymers. In comparing the result of spinning experiments with transient extensional viscosity results the authors (41) concluded that although values agreed well the factors of extrudate swell, draw resonance and filament breakage limit the quality of the spinning results and the range of strain rates over which they were obtained.

2.6 MATERIALS

Polymer melts that have been covered by the work reported herein are listed below, where the number in brackets indicates the number of references for which results on that material were presented:

LDPE [29]
 HDPE [11]
 LLDPE [9]
 PS [18]
 PP [10]
 PMMA [2]
 ABS
 blend of PS/LDPE
 blend of HDPE/LDPE
 blend of LLDPE/LLDPE
 blend of LLDPE/LDPE
 CaCO₃ filled PS
 ethylene based ionomer melts E/MMA
 fibre-filled PS
 glass fibre filled PP
 graft-copolymer LDPE-g-PS
 PA6
 PES
 PET
 PVC
 SBS copolymer
 VLDPE

In testing fibre-filled PP (up to 40% weight of fibres) Kamal et al (42) reported that the noise of the stress growth signal was about $\pm 5 - 15\%$ compared with $\pm 2\%$ for the unfilled material.

Results for polymer melts that were available in the literature were collated by Ide et al (43) and Petrie (5). Ide et al (43) listed the investigators who have carried out studies of various materials by isothermal melt spinning, constant elongation rate, constant stress and constant force methods: LDPE (10 references), PS (8 refs), HDPE (5 refs), PP (3 refs), PMMA (1 ref.), elastomers (2 refs) and nylon 6 (1 ref.). Petrie (5) tabulated papers presenting uniaxial extensional results obtained by constant stress, constant strain rate, constant force and constant speed methods for PS (16 refs), LDPE (10 refs), HDPE (7 refs), PET (2 refs), PP (3 refs), PMMA (6 refs), PVC (6 refs), and also for elastomers and other materials. Petrie (5) also tabulated work on isothermal and non-isothermal spinning of polymer melts and solutions. Many of the papers are also presented in Table 1.

2.7 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: stress relaxation of the specimen (due to internal stresses locked in the specimen during its preparation) prior to and during testing, non-uniformity of the specimen dimensions, and impurities, including trapped air causing stress concentrations and possible degradation.

Methods for preparing specimens were principally extrusion (9, 10, 11, 38, 40, 43, 45-52) or transfer moulding which was predominantly carried out under vacuum (42, 53-58). Injection moulding (41, 59), compression moulding (33, 60) and machining from larger samples (14, 38) 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. Vinogradov (61) reported that annealing PS specimens made by extruding from a capillary rheometer into water had no effect on their stress growth behaviour. Ballman (14) used birefringence to identify whether specimens were free of stress.

Hingmann et al (45), 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 (54) 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 that should be independent of the initial orientation.

On the subject of impurities in the specimens, for example, Meissner (8) reported that dissolved air in PS will result in the formation of bubbles in the specimen on re-melting. The specimen were therefore produced under vacuum (8). This was used in conjunction with transfer moulding by a number of researchers to prepare specimens.

The homogeneity of deformation that is necessary particularly for obtaining data at high strains is critically dependent on specimen homogeneity. Meissner et al (36) commented that further increases in the strain to failure above a value of 7 are probably limited due to inhomogeneity of the specimen.

Meissner et al (36) commented that a disadvantage of type 1 compared with type 3 instruments is that larger specimen sizes are typically required. In testing of materials that are under development this may be a problem. However this difference in specimen size may not be significant compared with the sample size required for capillary extrusion measurements. Larger specimen sizes are potentially more susceptible to impurities and non-uniformity.

2.8 CLAMPING

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 applied to the sample). Two main approaches are used, either fixing the specimen to a single translating or rotating clamp, or by drawing the specimen between two counter-rotating rotors that may be textured to improve friction between the specimen and the rotors. The former type is referred to herein as "fixed" clamps. The use of either type of system has advantages and disadvantages.

Translating or single rotating clamps (fixed clamps)

In using fixed clamps the specimen can be 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 (16). However, this has the disadvantage that it may also affect the temperature uniformity of the specimen which, as is reported later, is important for uniformity of the deformation of the specimen. The alternative and more commonly adopted approach was 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 (18) commented that the use of adhesives seems to produce minimum end-effects and that epoxy adhesives appear to be suitable for polyolefins (18). Alternative approaches included the use of clamps similar to artist's pencil grips (i.e collet-type clamps) as used by Vinogradov et al (61), self-locking clamps (62, 63), and also the use of a hoop-shaped specimen mounted over two PTFE posts so that gripping of the specimen is not required (38). 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 (64-66).

Meissner (20) commented that type 3 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 introduces end-effects (10) that are more significant for specimens of small length to diameter aspect ratio. It is also noted that 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-effects 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 (67). 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 (61) observed that for PS specimens of length to radius 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. Cogswell (4) stated that for a length to radius ratio of at least 10 the end-effects will be comparable to the scatter in results.

Everage et al (65) used a modified Rheometrics Mechanical Spectrometer (rotational rheometer) using its off-set drive and torque measuring system to perform extensional measurements. Forced air heating was used as slumping of the specimen was not found to be a problem, 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 2 cm only could be used. Dealy (19), referring to Garfield et al (68), 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 (66)). Connelly et al (66) related the degree of non-

uniformity of the stretching process to the Weissenberg number characterising the flow. Petrie (5) commented that according to Pearson (personal communication referenced in (5)) the non-uniformity of the stretching of PS was most significant at low strain rates and high temperatures.

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. Meissner (8) emphasised that it is important to do this check in order to assess the accuracy of the test. Optical methods have also been employed to check on the uniformity of deformation (e.g. 40, 51, 66, 69).

Counter-rotating clamps

The use of a counter-rotating clamp system (for example Meissner (7)) has the advantages that constant Hencky strain rates (Appendix A1) 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-effect as polymer is not allowed to build up in the vicinity of the clamp. The deformation of the specimen is therefore homogenous along the specimen length (4, 36). 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. In one case the effect was reported to be up to 10% (36). The counter-rotating clamp arrangement of Meissner (7) was further developed (33) in which two pairs of tank tracks are used to stretch the specimen.

Laun et al (37) commented that difficulties in using two pairs of counter-rotating clamps, due to the need to both drive and measure the tension thorough 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), ie low tensile forces.

Münstedt et al (34) made steady state measurements of extensional viscosity of LDPE and HDPE using both type 1 and 3 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.

2.9 TEMPERATURE CONTROL AND TESTING MEDIUM

Temperature control

With the exception of four papers all the work reported using "controllable" methods were at temperatures equal to or less than 200 °C. The four exceptions to this were for testing up to: 250 °C for PA and 350 °C for PES (37), 210 °C for PP (70), 220 °C for LLDPE and HDPE/LDPE blends (58), and 220 °C for PP (49). All of these papers reported the use of silicone oils as mediums except for Meissner et al (33) in which a cushion of nitrogen was used to support the specimen. With reference to the use of oils, Muller (62) 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 (62) 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 will 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 behaviour of the specimen. 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. As described in section 2.8, Meissner (8) commented that it is important to check the uniformity of deformation of testing.

The importance of good temperature control has been acknowledged by various researchers. For example, to achieve good temperature control Muller et al (62) 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 $\pm 0.2^\circ\text{C}$. 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 (72) also used a double-layer heating arrangement. Axtell et al (38) commented that the use of an oil bath has the advantage over hot-air in that better stability is obtainable. In isothermal spinning measurements the temperature control was achieved using an oven beneath the extruder (41). However the majority of spinning measurements reported were carried out under non-isothermal conditions.

Meissner et al (36) 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 which was reported to be better than $\pm 0.1^\circ\text{C}$ at 150°C (36). With these improvements strains of up to 7 were obtained. Meissner et al (36) 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 (73). 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. For stretching techniques of the types 1, 2 and 3 Whorlow (21) commented that uniaxial extensional measurements are limited to strain rates of approximately 10 s^{-1} due to adiabatic heating. Laun et al (37) 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.

The role of the medium in supporting the specimen

Apart from providing a controlled temperature environment the test medium must also provide sufficient support for the specimen. Silicone oil was used almost exclusively with the exception of when testing was carried out in air (eg 41, 65, 66, 74) or using a nitrogen cushion (33). In the papers presented in Table 1 the specimens were either mounted vertically in the oil (eg 9, 53) or horizontally either submerged in or on the surface of the oil (e.g. 7, 10). Utracki et al (55) 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 (Cogswell commented when the viscosity $< 10^6$ Pa.s (4)) it cannot be ignored and the use of an oil bath or other mechanism to support the specimen is required (19). 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 was 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.

Effect of the test 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 (45) 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% by weight with the absorption being in a surface layer of ≈ 300 μ m 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 (45).

Lanfray et al (70) 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 (42) 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 or thermal degradation. Similarly for LLDPE and LDPE, Schlund et al (54) 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 appeared to be no comment on the failure behaviour of the polymers. Schlund et al (54), referring to work of Kurbanaliev (75) and Utracki (71), 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 (75) were alcohols and the findings may not apply to the use of silicone oils. In the work presented by Schlund et al (54) 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 (58) commented that testing at low strain rates ($\dot{\epsilon} < 0.01$) 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 (37) observed differences in behaviour of normal and stabilised LDPE indicating that crosslinking of the normal LDPE was occurring at 150 °C.

Meissner (20) 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 (33) 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 crosslinking must still be assessed.

2.10 STRAINS, STRAIN RATES AND STRESSES OF TESTING

Strains and strain rates

The predominant problem with type 3 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 (62). Rhi-Sausi et al (32) commented that strain rates above 0.1 s^{-1} are difficult to achieve using a type 3 instrument with screw-thread drive as high clamp speeds are required. However, values as high as 1 s^{-1} were obtained by Schlund et al (54) using a rotary motor as a winding device for a cable connected to the clamp, and 2 s^{-1} by Lanfray et al (70) and also Lacaze et al (76) using twin screw-driven clamps. Surprisingly, Vinogradov (61) reported a maximum value of 6 s^{-1} using a screw-driven device, although the next highest strain rate reported therein was $8.7 \times 10^{-3} \text{ s}^{-1}$. Typically the largest strains and strain rates obtained using a type 3 instrument were approximately 4 and 2 s^{-1} respectively. In comparison, using type 1 and 2 instruments strains up to 7 were achieved by both Meissner et al (36) and Raible et al (77), and strain rates of up to 30 s^{-1} were achieved by Münstedt et al (34), 10 s^{-1} by Laun (37) and 1.5 s^{-1} by Ide et al (43). Measurements carried out by Münstedt et al (34) 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 rates 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 (21). 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 - 3% on the basis of optical measurements were made by Tanaka et al (40). Gupta et al (1) 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 machine (1, 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 (43) 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 (43) ranged from 16% for HDPE and PP, 12% for LDPE and 5% for PS and PMMA.

Laun (78) 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.

Stresses

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. $\epsilon = 6$ 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, 23). Force transducers need to be of the appropriate range and also have good resolution and stability.

Instruments presented in Table 1 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 $\dot{\epsilon} = 0.005 \text{ s}^{-1}$ and 150 °C (36), and 0.002 N for LDPE at $\dot{\epsilon} = 0.01 \text{ s}^{-1}$ and 160 °C (43). Quoted values of the resolution of stretching instruments (excluding type 4) used for testing polymer melts were of the order of 10^{-3} N (9, 33) and 10^{-4} N (37). Tensile forces were measured either using leaf springs combined with displacement transducers (e.g. 8, 33, 37) or strain gauges (e.g. 72), load cells (e.g. 9, 43), or the force was applied using cam loading systems (eg 10, 16, 44). Sebastian et al (72) 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 (44) or 10^5 Pa.s (4, 19). Ishizuka et al (49) commented that frictional forces can become significant at high strains (estimated at $\approx 10\%$ for $\epsilon = 4$, $\dot{\epsilon} = 0.6 \text{ s}^{-1}$) as the tensile force on the specimen decreases considerably with increased strain.

Laun et al (37) showed, using a combined experimental and theoretical approach, that interfacial tension effects on tensile stress measurements of LDPE at 150 °C (using silicone oil) were negligible at strain rates above 10^4 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 3) instruments for which relative speeds between the specimen and the oil will be greater.

Ide et al (43) 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 neglected in their work. Ishizuka et al (49) indicated how the effect of drag on measurements can be taken into account.

Rhi-Sausi (32) 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 3 instrument. Ishizuka et al (49) concluded that the effect of inertia for their type 2 instrument was of the order of 0.02% and was therefore negligible.

2.11 OTHER EXTENSIONAL FLOW MEASUREMENT TECHNIQUES AND CONSIDERATIONS

Several researchers have used flow birefringence measurements to determine stress levels during extensional measurements. These have been reviewed by Mackay et al (79).

Alternative approaches to measuring the extensional viscoelastic behaviour of polymer melts were pursued by Takaki et al (80) and Karin (81) although these methods were "non-controllable" as defined earlier. Takaki et al (80) used a drop weight approach in which a specimen clamped at the top was suspended vertically in an oil bath with a supported weight attached to the bottom. When the specimen had reached the test temperature the bath was draining and the support for the weight removed thus allowing it to fall thereby stretching the specimen. Problems described were due to slumping of the specimen and also the specimen pulling off of the clamps, although details of the clamps were not presented. These were problems particularly for low molecular weight specimens and at higher temperatures. Karin (81) reported a novel approach of using a melt centrifugal pelletizer in which polymer was extruded, under centripetal force, through holes and was then deformed and subsequently quenched. From the dimensions of the quenched, deformed pellet and the testing conditions used extensional viscosity values were determined. The author commented that this technique is suited to very high strain rates (up to 43 s^{-1}) and short extension times (< 1 second).

In order to overcome some of the difficulties faced by the experimentalist in performing extensional measurements Petrie (12) suggested that mathematical analysis could be used to take into account, rather than neglecting the differences between reality (the experiments) and the ideal (the basis by which analysis of experiments is currently made). He indicated however that this would not necessarily be a trivial exercise.

Calibration

The major problem in checking the calibration of an extensional rheometer is that there are no calibrants or reference materials that could be used. 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, and also ensuring the uniformity of deformation. Dealy (19) discussed some of the problems associated with calibration of extensional rheometers. He commented that the minimum requirement should be to check that the

ratio of extensional viscosity at very low extension rates was three times that of the zero shear viscosity (limiting shear viscosity at zero shear rate). 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 (see reference to intercomparison of A1 and M1 fluids, section 2.3).

3 DISCUSSION

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 (type 1 and type 3) the magnitude of the effect of end-errors introduced due to using a non-rotating clamp was found to be negligible (34). However, the effect of end-errors on the uniformity of the deformation was also observed to be very significant (65). 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 (61).

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 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 (75). 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 (36) or double-skinned silicone oil baths (61, 63). 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, which 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 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. Methods for correcting for the effect of these errors were referenced.

In summarising the current state of extensional measurement capability, and addressing the industrial requirements expressed by Cogswell (32) (see Section 2.2), flow measurements at strains up to 7, strain rates up to 30 s^{-1} and temperatures up to 350°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 ≈ 4 , strain rates up to $\approx 1 \text{ s}^{-1}$ and temperatures up to $\approx 200^\circ\text{C}$ but such equipment is still of limited availability.

4 CONCLUSIONS

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.
- The use of at least one rotating clamp overcomes the restriction that the maximum strain is limited by the length of the test equipment.
- When using non-rotating clamps the effect of end-errors on the uniformity of deformation can be significant. Evidence indicates that the specimen length to diameter ratio should be at least 10.

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

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. However, the effect of the oil on the sample properties at failure is questioned and needs investigation.

- Temperature uniformity is essential for uniform deformation of the test specimen.

The homogeneity of specimens, in terms of their dimensions and also purity, affects the uniformity of their deformation.

It is desirable to check the uniformity of deformation of specimens to assess the quality of each test.

5 ACKNOWLEDGEMENT

The work reported in this paper was carried out as part of a project on measurement of the extensional viscoelastic properties of polymers (MMP11, milestone 3). This project is part of a programme of underpinning research financed by the Engineering, Automotive and Metals Division (EAM) of the Department of Trade and Industry on measurements related to the processability of materials.

6 REFERENCES

- 1 R.K. Gupta and T. Shridar, *Elongational Rheometers*, in *Rheological Measurement*, Ed. A.A. Collyer and D.W. Collyer, Elsevier Applied Science, London, 1988.
- 2 D.M. Binding, *Contraction Flows and New Theories for Estimating Extensional Viscosity*, in *Techniques in Rheological Measurements*, Ed. A.A. Collyer, Chapman and Hall, London, 1993.
- 3 M. Rides, NPL Report CMMT(A)38, Industrial need for extensional viscoelasticity measurements, September 1996 (submitted for approval).
- 4 F.N. Cogswell, *Trans. Soc. Rheol.*, 16 (1972) pp.383-403.
- 5 *Elongational Flows*, C.J.S. Petrie, Pitman, London, 1979.
- 6 J.R.A. Pearson, *Mechanics of Polymer Processing*, Elsevier Applied Science, London, 1985.
- 7 J.M. Meissner, *Trans. Soc. Rheol.*, 16 (1972) pp.405-420.
- 8 J. Meissner, *Chem Eng. Commun.*, 33 (1985) pp.159-180.
- 9 H. Mnstedt, *J. Rheology*, 23 (1979) pp.421-436.
- 10 H.M. Laun and H. Mnstedt, *Rheol. Acta*, 15 (1976) pp.517-524.
- 11 H.M. Laun and H. Schuch, *J. Rheology*, 33 (1989) pp.119-175.
- 12 C.S. Petrie, *Rheol. Acta*, 34 (1995) pp.12-26.
- 13 M. Rides, Numerical simulation of the extrudate swell behaviour of polymer melts, National Physical Laboratory Report CMMT(A)31, June 1996.
- 14 R.L. Ballman, *Rheol. Acta*, 4 (1965) pp.137-140.
- 15 F.T. Trouton, *Proc. Roy. Soc.*, A77 (1906) pp.426.
- 16 F.N. Cogswell, *Plastics and Polymers*, April 1968, pp.109-111
- 17 J. Meissner, *Rheol. Acta*, 8 (1969) pp.78-88.
- 18 J.M. Dealy, *Polym. Eng. Sci.*, 11 (1971) pp.433-445.
- 19 J.M. Dealy, *J. Non-Newtonian Fluid Mech.*, 4 (1978) pp.9-21.
- 20 J. Meissner, *Polym. Eng. Sci.*, 27 No. 8 (1987) pp.537-546.
- 21 *Rheological Techniques*, R.W. Whorlow, Ellis Horwood, London, 1992.
- 22 *Rheometry*, K. Walters, Chapman and Hall, London, 1975.
- 23 *Rheometers for Molten Plastics*, J.M. Dealy, Van Nostrand Reinhold Company, London, 1982.
- 24 J. Meissner, S.E. Stephenson, A. Demarmels and P. Portman, *J. Non-Newtonian Fluid Mechanics*, 11 (1982) pp.221-237.
- 25 L. Berger and J. Meissner, *Rheol. Acta*, 31 (1992) pp.63-74.
- 26 *Proceedings of an International Conference on Extensional Flow*, Combloux, France, March 20-23 1989. Special Issue of *J. Non-Newtonian Fluid Mechanics*, July 1990.
- 27 V. Tirtaatmadja and T. Sridhar, *J. Rheology*, 37 (1993) pp.1081-1102.
- 28 N.E. Hudson and T.E.R. Jones, *J. Non-Newtonian Fluid Mechanics*, 46 (1993) pp.69-88.
- 29 *Proceedings of a meeting on The Rheometry of Polymer, from the solution to the melt*, Abbaye Royale de Fontevraud, May 12-15 1993. Special Issue of *J. Non-Newtonian Fluid Mechanics*, May 1994.
- 30 D.F. James and K. Walters, A Critical Appraisal of Available Methods for the Measurement of Extensional Properties of Mobile Systems, in *Techniques in Rheological Measurements*, Ed. A.A. Collyer, Chapman and Hall, London, 1993.
- 31 F. Nazem and C.T. Hill, *Trans. Soc. Rheol.*, 15 (1974) pp.87-.
- 32 J. Rhi-Sausi and J. Dealy, *Polym. Eng. Sci.*, 16 (1976) pp.799-802.
- 33 J. Meissner and J. Hostettler, *Rheol. Acta*, 31 (1994) pp.1-21.
- 34 H. Mnstedt and H.M. Laun, *Rheol. Acta*, 20 (1981) pp.211-221.
- 35 P.K. Agrawal, W.K. Lee, J.M. Lorntson, C.I. Richardson, K.F. Wissbrun and A.B. Metzner, *Trans. Soc. Rheol.*, 21 (1977) pp.355-379.
- 36 J. Meissner, T. Raible and S.E Stephenson, *J. Rheology*, 25 (1981) pp.1-28.
- 37 H.M. Laun and H. Mnstedt, *Rheol. Acta*, 17 (1978) pp.415-425.
- 38 F.H. Axtell and B. Haworth, *Polymer Testing*, 9 (1990) pp.53-70.

- 39 V.V. Leitlands, Intern. J. Polymeric Mater., 8 (1980) pp.233-242.
- 40 K. Tanaka, K. Koyama and S. Kurita, J. Rheology 37(4) (1993) pp.609-620.
- 41 J. Revenu, J. Guillet and C. Carrot, J. Rheology, 37 (1993) pp.1041-1056.
- 42 M.R. Kamal, A.T. Mutel and L.A. Utracki, Polym. Composites, 5 (1984) pp.289-298.
- 43 Y. Ide and J.L. White, J. Applied Polym. Sci., 22 (1978) pp.1061-1079.
- 44 H. Mnstedt, Rheol. Acta, 14 (1975) pp.1077-1088.
- 45 R. Hingmann and B.L. Marczinke, J. Rheology, 38 (1994) pp.573-587.
- 46 M. Takahashi, T. Isaki, T. Takigawa and T. Masuda, J. Rheology, 37 (1993) pp.827-846.
- 47 L. Li, T. Masuda and M. Takahashi, J. Rheology, 34 (1990) pp.103-116.
- 48 W. Minoshima and J.L White, J. Non-Newtonian Fluid Mech., 19 (1986) pp.251-274.
- 49 O. Ishizuka and K. Koyama, Polymer, 21 (1980) pp.164-170.
- 50 Y. Suetguga and J.L.White, J. Applied Polym. Sci., 28 (1983) pp.1481-1501.
- 51 T. Takahashi, H. Toda, K. Minagawa, J-I. Takimoto, K. Iwakura and K. Koyama, J. Applied Polym. Sci., 56 (1995) pp.411-417.
- 52 P. Riha, J. Watanabe, K. Minagawa and K. Koyama, Proc. Fourth European Rheology Conference, Sevilla, Spain, 4-9 September 1994, pp.374-376.
- 53 L.A. Utracki and A.M. Catani, Polym. Eng. Sci., 25 (1985) pp.690-697.
- 54 B. Schlund and L.A. Utracki, Polym. Eng. Sci., 27 (1987) pp.380-386.
- 55 L.A. Utracki and P. Sammut, Polym. Eng. Sci., 30 (1990) pp.1019-1026.
- 56 D.M. Kalyon and D.S. Czerwonka, Plast. Rubber Proc. and Applications, 14 (1990) pp.29-33.
- 57 B. Schlund and L.A. Utracki, Polym. Eng. Sci., 27 (1987) pp.1523-1529.
- 58 L.A. Utracki, M.R. Kamal and N.M. Al-Bastaki, Conference Proc., ANTEC 1984, New Orleans, pp.417-420.
- 59 R. Fulchiron, V. Vernay and G. Marin, J. Non-Newtonian Fluid Mech., 48 (1993) pp.49-61.
- 60 F.N. Cogswell and D.R. Moire, Polym. Eng. Sci., 14 (1974) pp.573-576.
- 61 G.V. Vinogradov, V.D. Fikhman, B.V. Radushkevich and A.Ya. Malkin, J. Polym. Sci., A-2, 8 (1970) pp.657-678.
- 62 R. Muller and D. Froelich, Polymer, 26 (1985) pp.1477-1482.
- 63 D. Froelich, B. Muller and Y.H. Zang, ACS, Rubber Div., 128th Meeting - Fall Cleveland, Ohio, October 1985, paper 66, pp.23.
- 64 E. Kamei and S. Onogi, Appl. Polym. Symp., 27 (1975) pp.19-46.
- 65 A.E. Everage and R.L. Ballman, J. Applied Polym. Sci., 20 (1976) pp.1137-1141.
- 66 R.W. Connelly, L.G. Garfield and G.H. Pearson, J. Rheology 23(5) (1979) pp.651-662.
- 67 S.H. Spielgelberg, D.C. Ables and G.H. McKinley, J. Non-Newtonian Fluid Mechanics, 64 (1996) pp.229-267.
- 68 L.G. Garfield, G.H. Pearson and R.W. Connelly, Annual meeting, Society of Rheology, New York (1977) Abstract E15.
- 69 T. Takahashi, J. Watanabe, K. Minagawa and K. Koyama, Polymer 35 (1994) pp.5722-5727.
- 70 Y. Lanfray and G. Marin, Rheol. Acta, 29 (1990) pp.390-399.
- 71 L.A. Utracki and J. Lara, Paper presented at the "Table ronde internation. Ecoulement élongationnels, La Bresse, France, Jan 23-28, 1983 (Reference 17 of Schlund et al (54)).
- 72 D.H. Sebastian and J.R. Dearborn, Polym. Eng. Sci., 23 (1983) pp.572-575.
- 73 M.R. Kamal and H Nyum, Polym. Eng. Sci., 20 (1980) pp.109-119.
- 74 C.W. Macosko and J.M. Lornston, Society of Plastic Engineers. Technical Paper, 1973, vol 19 pp.461-467.
- 75 M.K. Kurbanaliev, G.V. Vinogradov, V.E. Dreval, A.Ya. Malkin, Polymer, 23 (1982) pp.100-104.
- 76 J.M. Lacaze, G. Marin and Ph. Monge, Rheol. Acta, 27 (1988) pp.540-545.
- 77 T. Raible, S.E. Stephenson, J. Meissner and M.H. Wagner, J. Non-Newtonian Fluid Mech., 11 (1982) pp.239-256.

- 78 H.M. Laun, J. Rheology, 30 No.3 (1986) pp.459-501.
- 79 M.E. Mackay and D.V. Boger, Flow visualisation in rheometry, in Rheological Measurement, Ed. A.A. Collyer and D.W. Clegg, Elsevier Applied Science, London, 1988.
- 80 T. Takai and D.C. Bogue, J. Applied Polym. Sci., 19 (1975) pp.419-433.
- 81 H.G. Karian, Conference Proc., ANTEC 1984, New Orleans, pp.412-416.
- 82 J.M. Dealy, J. Rheology 39(1) January/February 1995, pp.253-265.
- 83 L.E. Bailey, D.G. Cook, J. Pronovost and A. Rudin, Polym. Eng. Sci., 34 (1994) pp.1485-1491.
- 84 C.D. Han and J.Y. Park, J. Applied Polym. Sci., 19 (1975) pp.3257-3276.
- 85 R. Hingman and B.L. Marczinke, Proc. Fourth European Rheology Conference, Sevilla, Spain, 4-9 September 1994, pp.374-376.
- 86 M. Kobayashi, T. Takahashi, J. Takimoto and K. Koyama, Polymer 36 (1995) pp.3927-3933.
- 87 K. Koyama and O. Ishizuka, Polym. Process Eng., 1 (1983) pp.55-70.
- 88 P.J.R. Leblans, J. Samplers and H.C. Booij, Integration of fundamental polymer science and technology, Elsevier Applied Science Pub. Ltd, 1986
- 89 T. Matsumoto and D.C. Bogue, J. Polym. Sci.: Polym. Phys. Ed., 15 (1977) pp.1663-1674.
- 90 T. Takahashi, Y. Takagi, K. Minagawa, K. Iwakura and K. Koyama, Polymer 35 (1994) pp.4472-4473.
- 91 M.H. Wagner, Rheol. Acta, 18 No.6 (1979) pp.681-692.

Table 1: Summary specification of instruments for extensional viscoelasticity measurements presented in the literature (Bracketed values indicate the quoted range of the instrument that was not demonstrated by the results presented)

Ref.	Author date	Apparatus description	Deformation mode	Material	Temperature °C	Strain	Strain rate s^{-1} , maximum value or range where quoted (or stress values)	Clamping	Media
35	Agrawal et al 1977	Uniaxial extensional flow apparatus, type 3, horizontal, cam defining profile	constant $\dot{\epsilon}$	PS LDPE	150 - 165	≈ 4	0.04 - 0.7	two split half-clamps.	silicone oil
38	Axtell et al 1990	Rutherford elongational rheometer, type 3, horizontal	constant $\dot{\epsilon}$ constant σ constant force constant speed	PET	80 - 110 (20 - 280)	2.56	0.001 - 1 (10^{-5} - 1)	PTFE holders	silicone oil Dow-Corning 210H (290 °C max)
83	Bailey et al 1994	Spinning method type 4, horizontal	constant force, non-isothermal	LDPE LLDPE	-	≈ 3.7	1 - 18 (apparent values)	tensioning / winding device	air cooling / non-isothermal
14	Ballman 1965	Materials Technology Tensile Tester No. VHS-21 type 3	constant $\dot{\epsilon}$	PS	150	0.24	7.8×10^{-4} - 2.2×10^{-2}	threaded ends	oven
16	Cogswell 1968	Elongational rheometer type 3, cam loading	constant σ	LDPE PMMA	120 - 150	2	≈ 0.07 - 0.04 ($\sigma = 10^3$ - 10^7 Pa)	water cooled clamps	oil
60	Cogswell et al 1974	Constant load creep type 3, horizontal	constant σ	Poly-isobutylene	20	1	estimated < 0.01		
66	Connelly et al 1979	Modified Rheometrics Mechanical Spectrometer using offset drive and torque transducer system, type 2, horizontal	constant $\dot{\epsilon}$	PS	132 - 155	≈ 3.2	0.016 - 0.16	traditional solid's clamping 2 cm specimen length	forced air heating system
65	Everage et al 1976	Modified Rheometrics Mechanical Spectrometer using offset drive and torque transducer system, type 2, horizontal	constant $\dot{\epsilon}$	PS	155	≈ 3.4	0.01 - 1.2	traditional solid's clamping 2 cm specimen length	forced air heating system
63	Froelich et al 1985	Metravib Extensional rheometer / flow birefringence, type 3, vertical but with two moving clamps, 450 mm total travel, see ref 62.	constant $\dot{\epsilon}$	PS	140 double silicone oil bath.	3.2	0.01 - 0.5 (2×10^{-4} - 2)	self locking clamps	silicone oil or oven for high viscosity polymer
59	Fulchiron et al 1993	Viscostrain elongational rheometer (Metravib) type 3 (vertical ?)	constant $\dot{\epsilon}$	PP	180	≈ 2.5	0.5 - 2	-	PDMS oil

Ref.	Author date	Apparatus description	Deformation mode	Material	Temperature °C	Strain	Strain rate s ⁻¹ , maximum value or range where quoted (or stress values)	Clamping	Media
84	Han et al 1975	Blown film extrusion force balance Killion extruder Requires sectioning of film to determine thicknesses and thus strains, strain rates, viscosities.	uniaxial biaxial	HDPE LDPE PP	200	≈ 5	0.01 - 2	film blowing pilot plant	
45	Hingmann et al 1994	type 3, vertical (see ref 9)	constant $\dot{\epsilon}$ constant σ	PP LDPE	175	2.3 (3)	0.002 - 1 ($\sigma = 1.5 \times 10^2 - 2 \times 10^3 \text{ Nm}^{-2}$)	glued to metal clamps	silicone oil
85	Hingmann et al 1994	type 3, vertical (see ref 9)	constant $\dot{\epsilon}$	PP	175	2.5	0.002 - 1	etched and glued to metal clamps	silicone oil
43	Ide et al 1978	Extensional rheometer, type 2, horizontal, load cell	constant $\dot{\epsilon}$ stress relaxation	LDPE HDPE PS PP PMMA	160 - 180	3.5	0.005 - 1.5	take-up roller and fixed clamp	silicone oil
49	Ishizuka et al 1980	type 2, horizontal	constant $\dot{\epsilon}$	PP	180 - 220	≈ 4	0.002 - 0.6	single pair of rotating clamps	silicone oil
56	Kalyon et al 1990	Rheometrics Extensional Rheometer, model 9000 type 3, vertical	constant $\dot{\epsilon}$	LLDPE LDPE HDPE VLDPE	160 - 200	3.3	0.001 - 0.1	epoxy adhesive on etched ends	silicone oil Dow Corning 200 poly(dimethylsiloxane)
42	Kamal et al 1984	Rheometrics Elongational Rheometer, model 9000 type 3, vertical	constant $\dot{\epsilon}$	Glass fibre filled PP (0 - 40% of fibres by weight)	180	1.6	0.003 - 0.6	epoxy tie clips	silicone oils: Dow-1265 Dow-210H
64	Kamei et al 1975	Instron type tensile tester Tensilon UTM 4L type 3 film specimens	constant clamp speed, variable strain rate	PS	110 - 170 (25-250)	≈ 2	-	conventional clamps for film specimens	oven
81	Karian et al 1984	Centrifugal pellitizer stretching times 0.02 - 1 s	constant tensile force	HDPE LDPE PP PVC PS	146 - 250	≈ 2	1 - 47	extrusion of pellets through holes -	measurement of dimensions of quenched, deformed pellets.

Ref.	Author date	Apparatus description	Deformation mode	Material	Temperature °C	Strain	Strain rate s ⁻¹ , maximum value or range where quoted (or stress values)	Clamping	Media
86	Kobayashi et al 1995	type 2, horizontal (see ref 49)	constant $\dot{\epsilon}$	fibre-filled PS	160	≈ 2	0.003 - 0.38	single pair of rotating clamps	floating on silicone oil KF50-100cs, Shinetu Chemical Co.
87	Koyama et al 1983	type 2, horizontal	constant $\dot{\epsilon}$	LDPE	120 - 185	≈ 4	0.002 - 0.6	rotating clamps	silicone oil
76	Lacaze et al 1988	Metravib elongational rheometer type 3, vertical, 2 moving clamps load cell	constant $\dot{\epsilon}$	LLDPE HDPE	190	2.3	0.5 - 2	vertical guides with contra-rotating screws	silicone oil
70	Lanfray et al 1990	Metravib VGE elongational rheometer type 3 vertical, 2 moving clamps (see ref. 76)	constant $\dot{\epsilon}$	PS PP	140-160 190-210	2	0.005 - 2		blend of polysiloxane oils
10	Laun et al 1976	Universal extensional rheometer, rotating clamp type 1, horizontal (see ref. 7) Tensile creep type 3, vertical, twin cam loading (see ref. 44)	constant $\dot{\epsilon}$ recoil constant σ	LDPE	150 (190)	6 3.6	0.03 - 0.1 0.03 - 0.1	two pairs of toothed rotating clamps glued to metal sheets then fixed to clamps	silicone oil
37	Laun et al 1978	horizontal, type 2	constant $\dot{\epsilon}$	LDPE stabilised LDPE	125 - 150	6	10 ⁻⁵ - 10	single pair of toothed rotating clamps fixed end-plate	silicone oil
11	Laun et al 1989	Meissner type type 1, horizontal (see ref. 7) Gottfert Rheotens spinning type 4 Cogswell - Münstedt type 3, (see ref. 9)	constant $\dot{\epsilon}$ constant draw-down speed constant σ	LDPE PS	170 230* 170	> 3.5 - 4.5 3.2	10 ⁻⁴ - 10 0.001 - 1	toothed rotating gears toothed gear wheel *non-isothermal, inhomogeneous, glued to metal plates	silicone oil atmosphere silicone oil
88	Leblans et al 1986	Gottfert Rheotrain	constant $\dot{\epsilon}$	LDPE	150	3	0.005 - 0.97		

Ref.	Author date	Apparatus description	Deformation mode	Material	Temperature °C	Strain	Strain rate s ⁻¹ , maximum value or range where quoted (or stress values)	Clamping	Media
39	Leitlands 1980	Uniaxial extension, Vibrorheometer, type 1, horizontal	unsteady extensional flow (oscillatory superimposed on constant $\dot{\epsilon}$)	Polyisobutylene	19	3	0.001 - 0.14	two pairs of roller clamps	thermostatic bath
47	Li et al 1990	Meissner type type 1, horizontal	constant $\dot{\epsilon}$	ABS	130 - 180		0.017 - 0.084 (8.3 × 10 ⁻⁴ - 8.4)	rotating grooved roller clamps	silicone oil
74	Macosko et al 1973	Modified Rheometrics Mechanical Spectrometer using offset drive and torque transducer system, type 2, horizontal	constant $\dot{\epsilon}$	HDPE	180	≈ 3.3	0.011 - 1.2	slit clamp 2.4 cm specimen length	
89	Matsumoto et al 1977	Simultaneous extensional-birefringence measurement, type 2, isothermal or controlled temperature ramp conditions	constant $\dot{\epsilon}$	PS	120 - 160 also non-isothermal testing	≈ 2	0.075 - 2	fixed clamp and twin roller clamp	air circulating oven
17	Meissner 1969 (in German)	Uniaxial extensional rheometer, rotating clamp type 1, horizontal	constant $\dot{\epsilon}$ constant σ	LDPE	150	4	0.001 - 0.01	two pair of gears as rotational clamp	silicone oil
7	Meissner 1972	Universal extensional rheometer, rotating clamp type 1, horizontal Uniaxial haul-off tester type 4, vertical	constant $\dot{\epsilon}$ constant σ recoil constant down-haul speed	LDPE	150	4.5	0.001 - 1	two pairs of gears as rotational clamps (recoil of samples using scissors) rotating haul-off device from extrusion rheometer	silicone oil
36	Meissner et al 1981	Uniaxial type 1, horizontal pneumatic cutting of specimens	constant $\dot{\epsilon}$ constant σ	LDPE	150	7	0.001 - 1	rotating clamp	silicone oil
48	Minoshima et al 1986	Elongational rheometer type 2, horizontal (see ref. 50)	constant $\dot{\epsilon}$	LDPE HDPE LLDPE	180	4	0.001 - 0.1	twin take-up rollers + fixed clamp	silicone oil

Ref.	Author date	Apparatus description	Deformation mode	Material	Temperature °C	Strain	Strain rate s ⁻¹ , maximum value or range where quoted (or stress values)	Clamping	Media
44	Münstedt 1975	Tensile creep, cam loading type 3, vertical	constant σ $3 \times 10^4 - 4.5 \times 10^5 \text{ Nm}^{-2}$ recoil	PS	140 (160)	2.3	estimated < 0.001 ($\sigma = 3 \times 10^4 - 4.5 \times 10^5 \text{ Nm}^{-2}$)	etched and glued to metal clamps	silicone oil
9	Münstedt 1979	Uniaxial extensional rheometer, translating clamp, maximum displacement 500 mm, type 3, vertical	constant $\dot{\epsilon}$ constant σ recoil & relaxation	PS	130 (150)	3 (3.9)	0.2 - 0.4 (0.0005 - 0.5)	etched and glued to metal clamps. (fixed clamp, moving clamp pulled by rotor)	silicone oil
34	Münstedt 1981	1. Universal extensional rheometer type 3, vertical (see ref. 9) 2. Modified Meissner design type 1, horizontal (see ref. 37)	constant σ constant $\dot{\epsilon}$	LDPE HDPE	150	4 6	$(\sigma = 5 \times 10^2 - 2 \times 10^5 \text{ Pa})$ 0.001 - 30	translating clamps two pairs of rotating clamp	silicone oil
62	Muller et al 1985	Extensional rheometer / flow birefringence, type 3, vertical but with two moving clamps, 450 mm total travel	constant $\dot{\epsilon}$ constant σ	PS	150	3.2	0.01 - 1 ($2 \times 10^{-4} - 2 \times 10^{-2}$ or $2 \times 10^{-4} - 2$)	self locking clamps	silicone oil
77	Raible et al 1982	Rotary clamp rheometer type 1, horizontal (see ref. 36)	constant $\dot{\epsilon}$ constant σ constant force	LDPE	150	7	0.001 - 1	rotating clamps	silicone oil
41	Revenu et al 1993	Isothermal melt spinning type 4 Elongational rheometer type 3, vertical (see ref. 62)	constant take-up speed constant $\dot{\epsilon}$	LDPE LLDPE	160	4.8 2	0.05 - 0.35 0.1 - 0.5	rotating haul-off two translating clamps	Heated air chamber $\pm 1^\circ \text{C}$ silicone oil
33	Meissner et al 1994	tank track clamps with N ₂ supporting cushion, 1 g test specimens type 1, horizontal	constant $\dot{\epsilon}$	PS, PA6, PES	170 - 350	7	0.1 - 1 (0.001 - 1)	two pairs of tank tracks	N ₂ cushion
32	Rhi-Sausi et al 1976	Elongational rheometer type 3, horizontal, profiled specimens 8 mm in diameter in central region, 10 mm at clamps	constant $\dot{\epsilon}$ constant σ	Alathon (LDPE)	180	2.5	0.02 - 0.1	single moving oil cooled clamp fitted with compressible bushing	silicone oil
52	Rhia et al 1994	Unsteady extensional rheometer: superimposed steady and dynamic extension, type 2, horizontal (see ref. 40)	constant $\dot{\epsilon}$ with superimposed sinusoidal oscillations	LDPE	160	0.8	0.01 - 0.08	single pair of rotating clamps	silicone oil

Ref.	Author date	Apparatus description	Deformation mode	Material	Temperature °C	Strain	Strain rate s ⁻¹ , maximum value or range where quoted (or stress values)	Clamping	Media
54	Schlund et al 1987	Rheometrics Elongational Rheometer, model 605, type 3, vertical	constant $\dot{\epsilon}$	LLDPE LDPE	150	3	0.0005 - 1	glued using epoxy to Al ties	silicone oil: Dow 200
57	Schlund et al 1987	Rheometrics Extensional Rheometer, model 9000 type 3, vertical	constant $\dot{\epsilon}$	blends of LLDPE/ LLDPE/ LLDPE & LLDPE/ LDPE	150	3.1	10 ⁻⁴ - 1	epoxy glued to metal	silicone oil
72	Sebastian et al 1983	Extensional rheometer, type 2, horizontal, load cell (following design of ref. 43)	constant $\dot{\epsilon}$ constant σ constant load	HDPE LLDPE LDPE PP	190	≈ 2	0.002 - 1.1	take up roller and fixed clamp	silicone oil
50	Suetsugu et al 1983	Modified version of ref. 43. type 2, horizontal	constant $\dot{\epsilon}$	CaCO ₃ filled PS	180	2.5	0.005 - 0.15	two-gear wheel clamp, other fixed clamp	silicone oil
69	Takahashi et al 1994	type 2, horizontal (see ref. 49)	constant $\dot{\epsilon}$	ethylene based ionomer melts E/MMA	140	≈ 3	0.004 - 0.51	single pair of rotating clamps	floating on silicone oil
46	Takahashi et al 1993	Meissner type rheometer uniaxial & biaxial type 1 (see ref. 7)	constant $\dot{\epsilon}$	PS PP	145-160 180	5	0.015 - 0.9	two pair of gears as rotational clamp	silicone oil KF54 & KF968 (Shin-etu Chemical Co.)
90	Takahashi et al 1994	type 2, horizontal (see ref. 49)	constant $\dot{\epsilon}$	LDPE LDPE-g-PS	140 - 200	≈ 2.5	0.003 - 0.4	single pair of rotating clamps	floating on silicone oil
51	Takahashi et al 1995	type 2, horizontal (see ref. 49)	constant $\dot{\epsilon}$	PS, SBS copolymer	120 - 160	≈ 2	0.003 - 0.46	rotating pair of gears	floating on silicone oil
80	Takai et al 1975	Drop weight extensometer, type 3, vertical	$\approx$ constant force	PE	150 - 200	3	≈ 0.5	special clamps	oil
40	Tanaka et al 1993	Unsteady extension rheometer: superimposed steady and dynamic extension type 2, horizontal	constant $\dot{\epsilon}$ with superimposed sinusoidal oscillations	LDPE HDPE	160	3.2	0.08 (0.0008 - 0.5)	single pair of rotating clamps	silicone oil

Ref.	Author date	Apparatus description	Deformation mode	Material	Temper- ature °C	Strain	Strain rate s^{-1} , maximum value or range where quoted (or stress values)	Clamping	Media
58	Utracki et al 1984	Rheometrics Extensional Rheometer, model 9000 type 3, vertical	constant $\dot{\epsilon}$	LLDPE HDPE/LDP E blend	190 - 220		0.001 - 1		silicone oil DOW 210H
53	Utracki et al 1985	Rheometrics Elongational Rheometer, type 3, vertical	constant $\dot{\epsilon}$ constant σ	LDPE LLDPE HDPE	150 - 190	≈ 3	0.001 - 3	fixed to Al ties	silicone oil
55	Utracki et al 1990	Rheometrics Extensional Rheometer, model 9000 type 3, vertical maximum displacement 600 mm	constant $\dot{\epsilon}$	PS/LDPE blend	150	3.1	0.0005 - 1	glued to aluminium ties using epoxy	dimethylsiloxane oil Dow 200
61	Vinogradov et al 1970	horizontal, type 3	constant $\dot{\epsilon}$	PS	130	≈ 2.3	$3 \times 10^{-3} - 6$	artist's pencil holder type clamps	glycerine

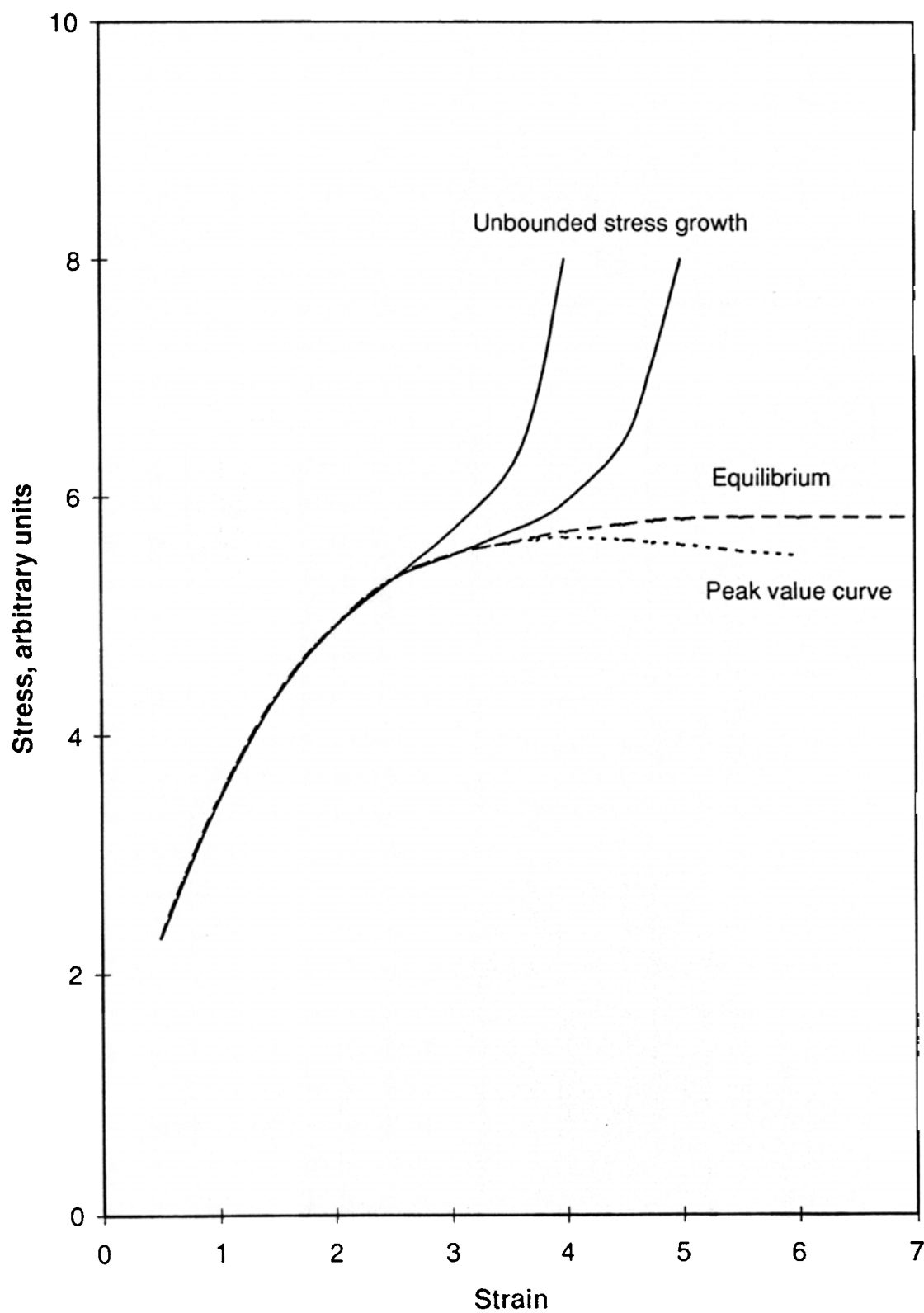


Figure 1 Schematic of stress growth behaviour of polymer melts illustrating the three principle types of behaviour.

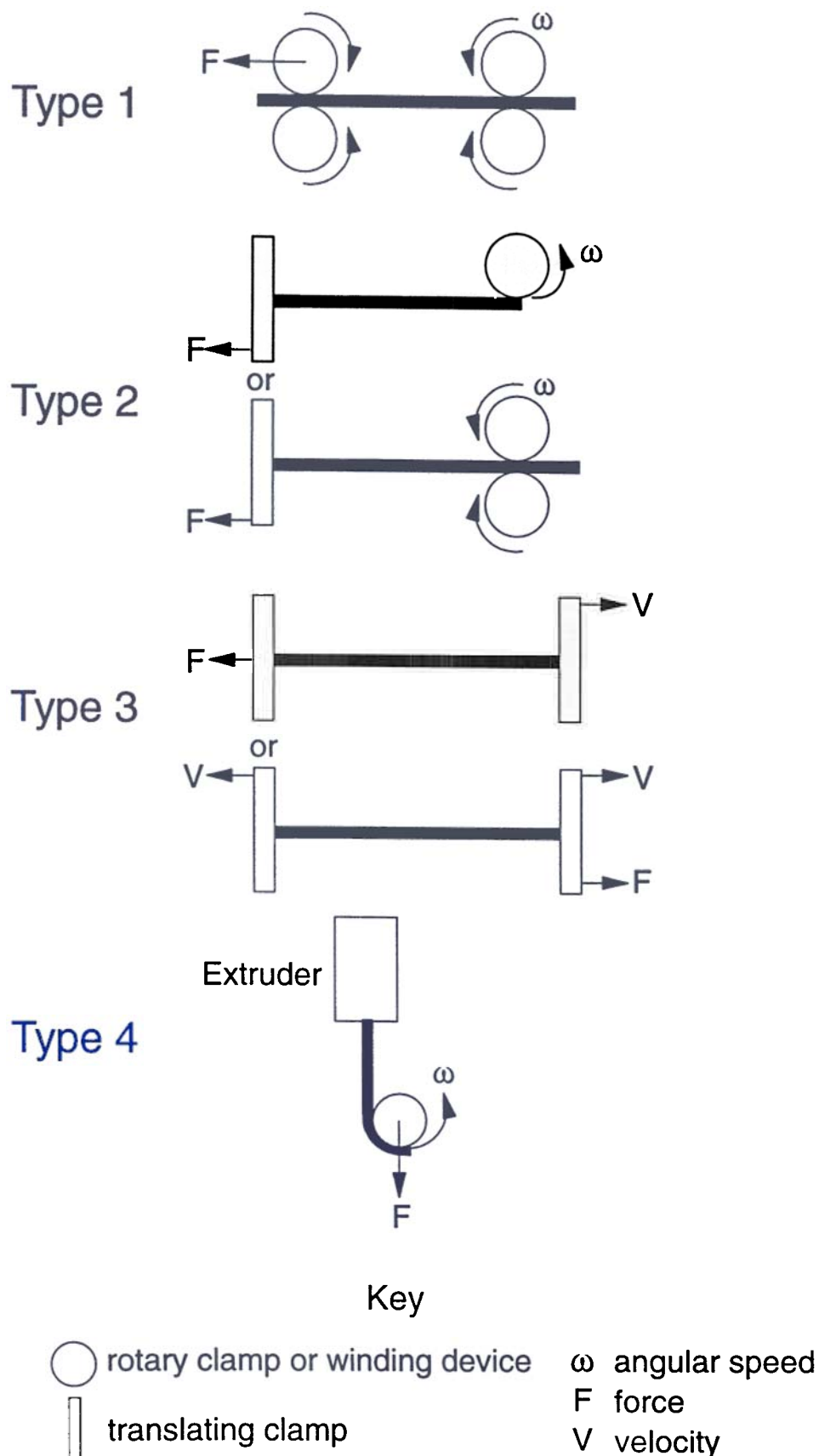


Figure 2 Schematic of test instrument types 1 - 4.

APPENDIX A1 DEFINITIONS OF STRAIN, STRAIN RATE, STRESS AND MATERIAL PROPERTIES FUNCTIONS IN TENSILE (SIMPLE) EXTENSION

The following definitions are given by Whorlow (21) for strains and strain rates. Further descriptions are given by, for example, Gupta et al (1), Petrie (70), Dealy (18, 19).

A1.1 Elongation ratio

The *Elongation ratio* (ER) is the ratio of the current length l to the initial length l_0 of the specimen:

$$ER = l/l_0.$$

A1.2 Cauchy ϵ_c and Hencky ϵ strains

The *Cauchy strain* ϵ_c is given by the ratio of the change in length δl to the initial length l_0 of the specimen:

$$\epsilon_c = \delta l/l_0.$$

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

$$\epsilon = \ln(l/l_0).$$

Thus the elongation ratio is related to the Cauchy strain by

$$ER = 1 + \epsilon_c$$

ie $l/l_0 = 1 + \delta l/l_0.$

To illustrate the difference between these values 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.

A1.3 Cauchy $\dot{\epsilon}_c$ or Hencky strain rates

The *Cauchy strain rate* is given by

$$\dot{\epsilon}_c = 1/l_0 \times \partial l / \partial t.$$

and the *Hencky strain rate* by

$$\dot{\epsilon} = 1/l \times \partial l / \partial t.$$

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.

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 (82).

Mathematical descriptions of extensional flows were presented, for example, by Meissner et al (24) and Walters (22).

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

$$\epsilon = \ln(l/l_0)$$

then in rectangular coordinates:

$$v_1 = \dot{\epsilon} x_1$$

$$v_2 = -\frac{1}{2} \dot{\epsilon} x_2$$

$$v_3 = -\frac{1}{2} \dot{\epsilon} x_3$$

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

Alternatively, in cylindrical coordinates:

$$v_z = \dot{\epsilon} z$$

$$v_r = -\frac{1}{2} \dot{\epsilon} r$$

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

A1.5 Material properties

Following the notation presented by Dealy (82) and prepared by the Nomenclature Committee of the Society of Rheology, for start-up flow in tensile (simple) extension at constant (Hencky) strain rate $\dot{\epsilon}$ 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 (82).

A1.5.1: The *net tensile stress* σ_E is defined by

$$\sigma_E = \sigma_{11} - \sigma_{22} = \sigma_{11} - \sigma_{33} = \sigma_{zz} - \sigma_r$$

σ_{ii} is a stress tensor in either rectangular or axisymmetric co-ordinates. The tensile stress growth function is indicated by σ_E^+ where the + indicates start-up rather than cessation of flow.

A1.5.2 The *tensile stress growth coefficient* η_E^+ is defined by:

$$\eta_E^+(t, \dot{\epsilon}) = \sigma_E / \dot{\epsilon}$$

where t is time.

A1.5.3 The *tensile viscosity* η_E is defined by:

$$\eta_E(t, \dot{\epsilon}) = \lim_{t \rightarrow \infty} [\eta_E^+(t, \dot{\epsilon})]$$

