Journal of Rheology

Elongation properties of a polymer melt: How to measure its rheological response with a tensile machine?

Alexandru Tudor Boborodea[1], Naïma Sallem-Idrissi[1], Freek Janssens[2], Evelyne van Ruymbeke[1]

1. *Université catholique de Louvain (UCLouvain), Institute of Condensed Matter and Nanosciences (IMCN), Bio- and Soft Matter (BSMA), Croix du Sud 1, Louvain-la-Neuve B-1348 Louvain-la-Neuve, Belgium*
2. *Instron Benelux, Sales and Service department, Hovesesteenweg 6, 2530 Boechout, Belgium*

Corresponding authors:

Alexandru Tudor Boborodea (alexandru.boborodea@uclouvain.be)

Evelyne van Ruymbeke (evelyne.vanruymbeke@uclouvain.be)

ABSTRACT

Extensional rheology is a technique complementary to shear rheology, enhancing comprehension of a material's deformation and flow behavior under different stress conditions. Although shear tests are relatively straightforward with rotational rheometers, instruments for extensional rheology have undergone numerous improvements over the last six decades to address challenges of testing molten polymers at high temperatures and difficulties in predicting sample geometry under large uniaxial extension. A powerful method for accounting for specific geometry modifications under high normal stresses is to perform extensional tests in which the crosshead speed is controlled by a feedback-loop program throughout the entire duration

Journal of Rheology

of the test. This method is implemented on the Versatile Accurate Deformation Extensional Rheometer (VADER 1000). However, this device is limited to relatively soft samples. Moreover, there is, today, no clear method capable of bridging the viscoelastic elongation properties of a polymer in its molten state to its properties in the solid state. We report here a methodology using an Instron tensile machine for extensional rheological tests, highlighting its capability to operate over a broad temperature range and to characterize samples that may not readily adhere to metal surfaces. We evaluate the validity of the method by analyzing elongation properties of a polydisperse polystyrene sample in its molten state. Results obtained using a constant Hencky strain rate profile on an Instron 68TM-10 instrument were found to be in very good agreement with the measurements from a VADER 1000 device and from an Extensional Viscosity Fixture (EVF) on an Advanced Rheometric Expansion System.

KEYWORDS

Extensional rheology, polystyrene, Hencky strain rate

I. INTRODUCTION

Extensional rheology is a technique complementary to shear rheology, providing a more complete understanding of the deformation of a material and of its viscoelastic behavior under different stress conditions[1]. While shear rheology focuses on the material's response to tangential forces, causing layers to slide past each other, extensional rheology examines the response to normal stresses that stretch or elongate the material. This distinction is crucial because materials can exhibit very different properties under extensional flow compared to shear flow. For instance, extensional viscosity can be significantly higher than shear viscosity, which is particularly important for processes involving stretching, such as fiber spinning[2] or blow molding[3]. Therefore, both types of tests are needed to provide a comprehensive view of the viscoelastic properties and processability of a polymer.

Non-linear shear tests are now better established and more widely understood, making them relatively straightforward to perform. The development of advanced geometries, such as the cone-partitioned plate geometry (CPP), has enabled reliable measurements even at high shear rates, demonstrating the maturity of shear rheology methodologies and instrumentation[4-6]. In contrast, extensional rheology requires specialized instrumentation to generate a homogeneous uniaxial or biaxial extensional flow, making such experiments more challenging to conduct. Additionally, the interpretation of extensional rheological data is less intuitive due to the complex nature of the deformation involved.

Extensional rheological measurements are based on the *Hencky* strain, denoted by ε_h. It is defined as the natural logarithm of the ratio between the instantaneous length (L) and the

initial length (L_0) of the sample, $\varepsilon_h = \ln(L/L_0)$. In solid mechanics literature, this measure is also commonly referred to as "true strain". The Hencky strain rate, $\dot{\varepsilon}_h$, is defined as the time derivative of the Hencky strain. In rheology, working at a constant Hencky strain rate is advantageous as it ensures a constant Weissenberg number, defined as $Wi = \dot{\varepsilon}_h \cdot \tau$, where τ is a characteristic relaxation time of the material. Maintaining a constant Wi is therefore essential for probing the material's response under specific dynamic conditions, particularly when studying strain-rate-dependent viscoelastic behavior.

In the present work, emphasis is on *uniaxial* extensional flow as it represents the most common flow used to study the properties of polymer melts. Under the assumption of incompressibility, it is described by the following velocity field[7]:

$$v_x = -1/2 \cdot \dot{\varepsilon}_h \cdot x \qquad v_y = -1/2 \cdot \dot{\varepsilon}_h \cdot y \qquad v_z = \dot{\varepsilon}_h \cdot z \tag{1}$$

One may reformulate these equations in terms of the position of a fluid particle as:

$$x(t) = x_0 \cdot \exp(-1/2 \cdot \dot{\varepsilon}_h \cdot t) \qquad y(t) = y_0 \cdot \exp(-1/2 \cdot \dot{\varepsilon}_h \cdot t) \qquad z(t) = z_0 \cdot \exp(\dot{\varepsilon}_h \cdot t) \tag{2}$$

where t denotes time, and (x_0, y_0, z_0) denotes the initial position of the particle.

From these equations, it follows directly that maintaining a constant Hencky strain rate results in an exponential increase in the sample length over time:

$$L(t) = L_0 \cdot \exp(\dot{\varepsilon}_h \cdot t) \tag{3}$$

It is important to note that, in practice, the applied strain does not necessarily result in a homogeneously distributed constant strain along the sample length or cross-sectional dimensions[8]. These inhomogeneities in the flow field along the elongation axis are often due to

necking phenomena or edge effects. Therefore, maintaining a constant Hencky strain rate within the cross-section remains a fundamental challenge in extensional rheometry. Deviations from ideal deformation conditions may introduce significant measurement errors, emphasizing the need for precise control over the deformation profile and optimal sample geometry to ensure accurate and reliable material characterization.

Although the first elongational studies of polymer melts were performed by Jenkel and Ueberreiter as early as 1938[9], to our knowledge, the first instrument for conducting homogeneous extensional flow tests was introduced in 1969 by Meissner[10]. This was achieved by utilizing a rotary clamp elongational rheometer which allowed reaching large elongations. Subsequent advancements in instrumentation have been developed to overcome the limitations of Meissner's original design[11]. These advancements included (1) the use of two rotational clamps to stretch homogeneously a molten polymer rod floating on a silicone oil bath[12], (2) the use of eight rotary clamps in a circular arrangement to obtain a biaxial extensional rheometer[13], (3) the replacement of the rotary clamps with metal conveyor belts as well as the replacement of the supporting liquid with a cushion of inert gas[11] to analyze polymers up to 300°C.

In 2002, Sentmanat proposed an alternative solution by developing a device compatible with rotational rheometers: the Sentmanat Extension Rheometer (SER). This system uses two counter-rotating drums with intermeshing gears to apply uniform stretching to a small rectangular sample placed between them[14,15]. Comparative studies have demonstrated that the SER design produces reliable and reproducible data, showing strong agreement with benchmark measurements from the traditional Meissner rheometer[16]. The commercial adoption of this technology was further accelerated as elongation data demonstrated to be highly sensitive to

the presence of long chain branching (LCB). This has been well illustrated in ref. 17 which showed that blends of linear low-density polyethylene (LLDPE) and low-density polyethylene (LDPE) exhibit strain hardening at high rates of deformation even at LDPE concentrations as low as 1%. Apart from the SER device, accessories to perform extensional rheology on rotational rheometers were also patented by Waters (TA Instruments, Extensional Viscosity Fixture, EVF, US 7,096,728 B2) and Anton Paar (Universal Extensional Fixture, UXF-TD, US 9,766,172 B2). While these devices provide valuable capabilities for rheological characterization, several practical considerations merit attention. The maximum achievable Hencky strain in standard SER operation is typically limited to approximately 3.5 due to mechanical constraints imposed by a single drum rotation[18]. However, recent methodological advances have demonstrated pathways to circumvent this limit for suitable materials. For instance, Parisi et al.[19] achieved Hencky strains up to 7 by using an oblique sample positioning and robust adhesion protocols (*e.g.*, double-sided tape), enabling the study of uncured rubbers at higher strain, albeit requiring a small correction to the Hencky strain rate. Similarly, innovations involving a vertical drum motion have been developed to further extend the achievable strain range[20]. Control of the temperature presents additional challenges, as thermal equilibration requires extended durations and often exhibits significant deviations between setpoint and measured values[21]. Furthermore, the horizontal placement of the film sample using two pins cannot prevent sample flow during high-temperature equilibration, particularly for low-viscosity systems[22], thereby limiting the range of testable temperatures and materials. Sample fixation at lower temperatures also proves challenging, though this has been successfully addressed in reference[15] by using a double-sided adhesive tape.

The challenges to be solved for a more general application of the elongation rheometer were reviewed by Meissner[23]. Keys goals for improving experimental accuracy include (1) refining sample clamping mechanisms to ensure reliable grip, (2) achieving better temperature homogeneity across the sample during testing, and (3) enhancing the precision and control of test parameters, particularly in terms of true deformation and deformation homogeneity. Beyond these priorities, Meissner also emphasized miniaturization of test devices as a long-term objective, to be able to analyze small quantities of samples.

In a parallel development, Münstedt[24] developed a rheometer specifically designed to address challenges such as maintaining controlled stress or strain rates across a wide range of sample deformations, ensuring sample geometry homogeneity during measurements, securely clamping the sample, and minimizing friction in the loading system. This innovative rheometer was used for conducting tensile creep experiments, with the cylindrical sample being vertically positioned within an oil bath. The vertical orientation of the sample closely mirrors the traditional position of samples in conventional tensile test instruments.

Keeping a similar concept, the device needed to measure the elongation properties of relatively low viscosity and sticky materials can be simplified by stretching a low quantity of sample placed between two coaxial discs of several millimeter diameter. This is the concept behind the filament stretching rheometer (FSR), which was first applied for measuring the extensional viscosity of polymer solutions at a constant Hencky strain rate. The filament stretching rheometer was introduced by Sridhar in 1991[25] and was further developed by Hassager et al.[26,27]. Later, a commercial version of this instrument was proposed, under the name of Versatile Accurate Deformation Extensional Rheometer (VADER 1000, Rheo Filament). The filament

obtained by moving the top plate upwards with a speed that increases exponentially with time has a geometry of a long cylinder. Its diameter can be monitored using a beam of light passed through it, the amount of light absorbed by the filament being directly proportional to the diameter of the filament. To ensure a constant Hencky strain rate for polymer melts, the upper plate is moving based on a velocity algorithm consisting of a feed-back loop and a feed-forward contribution[28]. The feed-forward contribution is derived from an empirical function and is referred to as the "kinematic master curve". It relates the Hencky strain along the filament length to the Hencky strain at the mid-filament plane[28].

Knowing the Hencky strain ε_h, the elongation properties of a sample can be determined by accurately measuring the true stress – denoted by σ_E^+ and alternatively termed the elongation stress growth *function* – in function of the instantaneous cross-sectional area (A) during deformation. This fundamental requirement follows from the definition $\sigma_E^+ = F/A$ where F represents the measured tensile force. From the true stress, the elongation stress growth *coefficient* – denoted by η_E^+ and derived as $\eta_E^+ = \sigma_E^+/\dot{\varepsilon}_h$ – determines the increase in extensional viscosity with time during uniaxial stretching. An important aspect to consider in this context is strain hardening – the progressive increase in η_E^+ beyond what would be predicted from linear viscoelastic theory. For some materials, it eventually reaches a "steady state" value, denoted by η_E. The steady state value is an intrinsic material property and is very sensitive to molecular architecture[29-33].

Although this work focuses on uniaxial extensional rheometry, the historical development of biaxial and multi-axial testing methods is also significant, particularly for investigations into the effects of compressibility. Seminal contributions include the biaxial stretching devices

developed and utilized by Kawabata and coworkers[34,35] and Urayama and coworkers[36,37], which employed square sheet specimens clamped on four sides for independently controlled orthogonal deformations.

Besides *uniaxial* rheological tests, the elongation properties of a polymeric sample can also be determined in its solid state, with the help of a tensile machine. In a standard tensile test, a dogbone-shaped specimen is clamped at both ends and elongated at a constant crosshead speed. The characteristic geometry of the sample ensures that deformation – and eventual fracture – occur mainly within the gauge length. As a tensile test is often conducted on materials in their solid state, it falls within the domain of solid mechanics. In this case, the *engineering* strain (denoted by ε_e) is typically used for analysis and is defined as $\varepsilon_e = (L - L_0)/L_0$. Under a constant crosshead speed condition, this results in a constant value for the engineering strain rate, denoted by $\dot{\varepsilon}_e$. Therefore, the length of the sample increases linearly with time as:

$$L(t) = L_0 + L_0 \cdot \dot{\varepsilon}_e \cdot t = L_0 + v \cdot t \tag{4}$$

where v denotes the crosshead speed.

The measurement of elongation rheological properties in polymer melts using tensile-test-based methods presents a significant scientific interest. Indeed, while the filament stretching rheometer has been successfully used to bridge the gap between polymer melts and concentrated solutions[38-40], there is, today, no clear method capable of bridging the viscoelastic elongation properties of a polymer in its molten state to its properties in the solid state. A tensile-test-based approach capable of operating across a wide temperature range could provide this missing connection.

However, such measurements require Hencky strain rate control, as constant engineering strain rates (or constant crosshead speeds) are inadequate for characterizing nonlinear viscoelastic behavior. This work builds upon the same core idea explored in earlier studies: leveraging a standard Instron tensile tester to perform extensional rheometry at a constant Hencky strain rate. A precedent is the work of Cotten and Thiele[41], who achieved this for filled elastomers at room temperature using a roller-based system. More recent advances have demonstrated two viable approaches to achieve this control. The first approach uses exponentially increasing crosshead speeds to have constant Hencky strain rate conditions. This approach has been successfully applied to bread dough through end-gluing sample fixation[42]. The observed agreement between calculated and experimental stress-strain data confirmed uniform gauge-length deformation for this viscoelastic material. This was further confirmed by direct visual observation of the samples during experiments. The second involves video-monitored tensile testing, where real-time sample geometry tracking enables constant Hencky strain rate along the *length* of the sample. While effective, this method has only been implemented below T_g[43], where "diffuse" necking phenomena remain governed by solid mechanics principles. One should also keep in mind that the technique itself requires the application of optical markers on the samples for reliable video tracking.

Extending these methods to molten polymers presents distinct challenges, as thermoplastic flows differ fundamentally. While preliminary studies suggest feasibility near T_g[44], high-temperature implementation requires addressing two critical aspects: (1) developing appropriate sample fixation techniques that prevent slip while minimizing edge effects, and (2) ensuring Hencky strain rate uniformity not just axially but throughout the cross-sectional dimensions.

This work specifically investigates these challenges, aiming to adapt tensile-based characterization for molten polymers while maintaining strain rate control. Successful implementation can provide the missing experimental link between solid-state and melt-state elongational behavior, which would complement the existing rheometer capabilities with enhanced temperature-range versatility. While controlled tensile tests are often performed below the glass transition temperature T_g, this requires an instrument capable of performing extensional rheometry tests above T_g (usually, $T_{\text{exp}} \approx T_g + 30$°C).

As a first step, we use the new loop-controlled feature of Instron's Bluehill Program to program and control the crosshead velocity profile with an appropriate and simple mathematical function of the *engineering* strain rate to achieve a constant *Hencky* strain rate. This feature operates by dynamically updating the velocity command at a defined time interval (set to one second in this work). At each step, the software calculates and applies a new crosshead speed based on the user-defined function. We also optimize the sample geometry and experimental set-up for ensuring a homogeneous deformation. Our method is validated on a commercial polystyrene sample by comparing the results obtained with the Instron instrument to those measured with the two well-established extensional rheometry systems: VADER 1000 filament-stretching rheometer (Rheo Filament) and ARES with Extensional Viscosity Fixture (EVF) (TA Instruments). We then discuss the possibility of extending the measurements to lower temperatures.

II. EXPERIMENTAL SECTION

1. Material

All tests were performed on a commercial general-purpose polystyrene, PS 1160 acquired from TotalEnergies, with a melt flow index (MFI) of 2.4 g/min (ISO 1133 H) and a Vicat softening point of 101°C (ISO 306B50). The weight-average molar mass is $M_w = 130{,}000$ g/mol and the polydispersity index is $Đ = 2.5$ (measured in tetrahydrofuran, using Agilent Technologies PLgel Mixed B columns and a calibration of polystyrene standards).

2. Differential Scanning Calorimetry

The glass transition temperature (T_g) of the material was determined using Differential Scanning Calorimetry (DSC) on a Mettler Toledo DSC STARe System. Samples of 5 to 10 mg were sealed in aluminium pans and heated from 30°C to 180°C at a rate of 10°C/min under a nitrogen atmosphere (flow rate: 50 mL/min). The midpoint of the heat flow change in the second heating cycle was taken as T_g.

3. Linear viscoelastic measurements

Linear viscoelastic measurements were conducted on an Advanced Rheometric Expansion System (ARES) (*i.e.*, a strain-controlled rheometer) equipped with 25 mm parallel plates and a transducer with a maximum torque of 200 g · cm (0.0196 N · m) and a minimum torque of 0.02 g · cm (0.00000196 N · m). The sample was loaded at a temperature of 200°C. Strain amplitude sweeps were performed to determine the linear regime of deformation. The frequency sweep tests were performed between 100 rad/s and 0.1 rad/s, at the following temperatures: 140°C $\pm$ 0.1°C, 160°C $\pm$ 0.1°C, 200°C $\pm$ 0.1°C, under an inert nitrogen atmosphere. Linear viscoelastic measurements serve as a baseline for validating experimental data obtained in the non-linear deformation regime.

4. Nonlinear viscoelastic measurements on VADER 1000

Sample preparation

Preparation involved compression molding at 200°C where polymer pellets were directly placed between the coaxial discs of the rheometer. Following molding, a controlled pre-stretch was performed at 170°C to generate the target "hour-glass" specimen geometry for extensional testing. Then, the sample was allowed a short relaxation period to dissipate stresses generated by the pre-stretch, and the oven temperature was reduced to the test temperature of 130°C with a minimum 15-minute equilibration period. Performing a pre-stretch followed by relaxation reduces shear components and edge effects near the end-plates and is common practice in extensional rheometry[45,46].

Experimental set-up and conditions

The Hencky strain rates used for experiments were 0.01 s^{-1}, 0.03 s^{-1}, and 0.1 s^{-1}.

Experimental data post-processing

No post-processing was performed on the acquired experimental data *by the user*.

5. Nonlinear viscoelastic measurements on ARES-EVF

Sample preparation

Preparation involved compression molding at 200°C under 50kN of applied force, performed in three consecutive 8-minute cycles to reduce the presence of air bubbles. The resulting rectangular specimens measured 18 mm × 10 mm × 1 mm.

Experimental set-up and conditions

The Hencky strain rates used for experiments were 0.01 s^{-1}, 0.03 s^{-1}, and 0.1 s^{-1}.

Experimental data post-processing

No post-processing was performed on the acquired experimental data *by the user*.

6. Nonlinear viscoelastic measurements on Instron

Sample preparation

Preparation involved compression molding at 200°C under 50kN of applied force, performed in three consecutive 8-minute cycles to reduce the presence of air bubbles. The resulting square specimens measured 25 mm × 25 mm × 2 mm.

Experimental set-up and conditions

The approach is a modification of a standard tensile test. The square specimens were installed between the clamps lined with a 100-grit sandpaper to ensure a firm grip and prevent slippage. Due to the clamped portions at both ends of the sample, the effective geometry becomes rectangular, with an aspect ratio $\Lambda_0 = L_0/W_0 \approx 0.4$ for the free zone. This value represents the smallest achievable ratio within the current clamp design, which serves to minimize the free length and thereby reduce sample sagging in the molten state. Furthermore, a small aspect

Journal of Rheology

ratio is important for two main reasons: it enables reaching larger Hencky strain values within the limited physical displacement inside the oven, and it ensures that the crosshead speeds required to achieve the target Hencky strain rates remain low, thereby minimizing normal forces and the risk of sample slippage or detachment. This specific geometry was also found to mitigate edge effects and minimize sample sagging in the molten state prior to testing. The final sample geometry was the product of an iterative optimization process, initiated from a standard ASTM D638 Type 1BA dogbone configuration. All experiments were conducted on an Instron 68TM-10 instrument. The oven and fixture were preheated for one hour without the sample to ensure thermal equilibrium. The sample was then inserted and allowed to equilibrate for 15 minutes at the test temperature. When working near the glass transition temperature, particular care must be taken to ensure a correct temperature stabilization, as small deviations in this regime can significantly impact the measured properties. To better illustrate the experimental methodology, Figure 1 shows the sample during an experiment performed at a Hencky strain rate of 0.03 s^{-1} at 130°C. The small vertical black line was originally a dot placed at the center of sample.

Figure 1. Experimental setup for tensile extensional rheometry. The image, captured through the oven window (note reflections), shows the sample at 130°C during a test at a Hencky strain rate of 0.03 s^{-1}, at a crosshead displacement of 40 mm ($\varepsilon_h \approx 1.4$). The fixture, clamps, and the deformation of a central (black) marker are visible.

Method implementation

The Bluehill Universal Software allows to impose a user-defined *engineering* strain rate function. We therefore used this software to program the desired strain rate profile. To achieve a constant *Hencky* strain rate, the engineering strain rate must be derived from the sample's time-dependent length:

$$L(t) = L_0 \cdot \exp(\dot{\varepsilon}_t \cdot t) \tag{5}$$

The required *engineering* strain rate is determined by differentiation:

$$\dot{\varepsilon}_e = 1/L_0 \cdot dL/dt = \dot{\varepsilon}_t \cdot \exp(\dot{\varepsilon}_t \cdot t) \tag{6}$$

The function derived above was programmed into the software and the strain rate was updated accordingly every 1 s. This protocol was found to be an optimal compromise between the accuracy of the displacement profile and the constraints on the instrument (*e.g.*, to avoid excessive mechanical command updates). One can observe that our approach imposes an a-priori known profile of deformation, which contrasts with the dual-loop system of the VADER 1000 rheometer. The validity of Eq. 6 relies on the assumption of a homogeneous deformation along the sample length, a condition that is material-specific and not universally guaranteed (*e.g.*, certain materials may exhibit local necking). Therefore, to preliminarily assess the applicability of this method for a new material, we recommend confirming deformation homogeneity. This could be achieved experimentally by using a video-extensometer to track markers on the sample surface, ensuring the strain is uniform.

The Hencky strain rates implemented were 0.01 s^{-1}, 0.03 s^{-1}, and 0.1 s^{-1}.

Implementation details of this methodology within the Bluehill Universal software environment, including screenshots and step-by-step explanations, are provided in the Supporting Information.

Experimental data post-processing

The testing software records time, displacement, and force measurements at regular intervals. The tensile stress growth function (*i.e.*, the true stress) is then computed. The calculation assumes a homogeneous deformation, where the cross-sectional area decreases exponentially with time according to $A(t) = A_0 \cdot \exp(-\dot{\varepsilon}_t \cdot t)$, with A_0 as the initial cross-sectional area. From this, the elongational stress growth coefficient can be determined.

Furthermore, using a tensile machine as an elongation rheometer also requires considering that at small strains ($\varepsilon_t < 1$) in the start-up, the measured force has contributions from both the *extensional* and the *shear* deformation components (*i.e.*, no-slip boundary condition), for samples with a non-uniform width along the extension direction (see Figure 1). Fluid elements near the rigid endplates experience significant shear displacements initially, although these shear rates decay exponentially with time. Spiegelberg et al. first quantified these end effects through lubrication analysis[47], revealing an exponential decay of radial pressure gradients with strain for a cylindrical sample, with an initial length denoted by L_0 and an initial radius denoted by R_0. The correction factor for the transient elongation viscosity is given by:

$$\eta^+_{\mathrm{E}\ \mathrm{corrected}} = \eta^+_{\mathrm{E}\ \mathrm{uncorrected}} \cdot \left[1 + \frac{1}{3 \cdot \Lambda_0^2} \cdot \exp\left(-\frac{7}{3} \cdot \varepsilon_t\right)\right]^{-1} \tag{7}$$

where Λ_0 is the initial aspect ratio, defined as $\Lambda_0 = L_0/R_0$.

While this was never done with a tensile machine, such issue was accounted for in the analysis of the data obtained with the filament stretching rheometer (*e.g.*, using the VADER 1000 instrument), for which a correction factor is also used to suppress the influence of this shear-dominated end-effects near the rigid plates. Rasmussen et al.[48] refined the correction in 2010 to:

$$\eta^{+}_{\mathrm{E\ corrected}} = \eta^{+}_{\mathrm{E\ uncorrected}} \cdot \left[1 + \left(\frac{R}{R_0}\right)^{10/3} \cdot \exp\left(-\Lambda_0^3\right) \cdot \frac{1}{3 \cdot \Lambda_0^2}\right]^{-1} \tag{8}$$

This correction factor was further modified by the same authors in 2013[49] to:

$$\eta^{+}_{\mathrm{E\ corrected}} = \eta^{+}_{\mathrm{E\ uncorrected}} \cdot \left[1 + \exp\left(-\frac{5}{3} \cdot \varepsilon_t - \Lambda_0^3\right) \cdot \frac{1}{3 \cdot \Lambda_0^2}\right]^{-1} \tag{9}$$

In 2016, Huang et al.[50] reference the expression denoted by equation (8) for the correction factor implemented in the VADER 1000 system. To our knowledge, this version represents the current correction protocol in the VADER 1000 system.

From these expressions, one can also conclude that a smaller initial aspect ratio is preferable as it ensures a reduced end-effect on the measured viscosity. Moreover, these corrections become negligeable at larger strains ($\varepsilon_t > 1$) and do not significantly impact the value of the steady state (often reached at $\varepsilon_t \approx 3$).

Following the established protocol used for the VADER 1000 system, we apply a correction to account for the edge effects during start-up (see equation 8). This correction function was adapted to account for the rectangular geometry of our sample, by introducing a modified

aspect ratio $\Lambda_0 = L_0/W_0 \approx 0.4$. It is important to note that this correction becomes negligible for $\varepsilon_t > 1$, which ensures a reliable steady-state viscosity value. A complete discussion of the correction function is beyond the scope of this work; the interested reader is directed to the work by Cardinaels et al.[46] for a more detailed study and recent advancements in this area.

Finally, it is of prime importance to control the speed at which the sample is stretched in the tensile machine. Recent software advancements now allow for precise control of the cross-head speed, with the ability to adjust speed, step by step, during testing with intervals as low as 0.1 seconds. Instron's Bluehill Program, starting from version 2.4 and available since December 2020, has incorporated this feature.

III. RESULTS AND DISCUSSION

1. Thermal Analysis

The glass transition temperature (T_g) of the material was determined to be 104.4 $\pm$ 0.3°C.

2. Linear Viscoelastic Properties

Using the Williams-Landel-Ferry (WLF) model, a master curve can be constructed at a reference temperature of 130°C to match with the experiments conducted in the non-linear regime. The shift factors were derived following the procedure detailed in ref. 51 with $C_1 = 8.99$ and $C_2 = 81.53$°C. As expected, the master curves exhibit consistent superposition across the tested temperature range (Figures 2a and 2b). This observation aligns with expectations for thermo-rheologically simple materials.

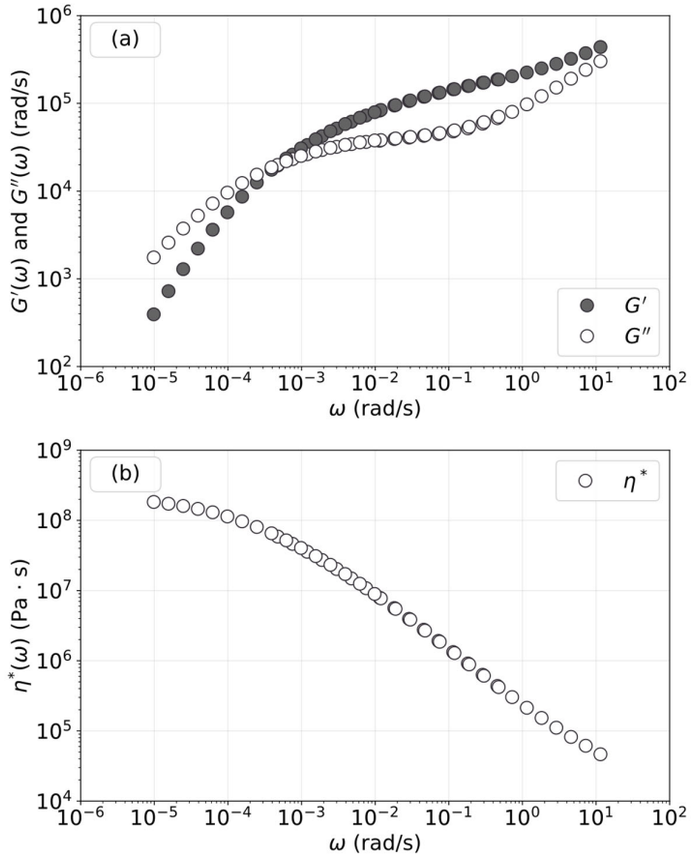

Figure 2. (a) Master curve of the storage (G') and loss (G'') moduli as a function of angular frequency (ω) at the reference temperature of 130°C. The storage modulus is represented by filled symbols and the loss modulus by empty symbols. (b) Master curve of the complex viscosity (η^*) as a function of angular frequency (ω) at the reference temperature of 130°C.

The linear viscoelastic envelope of the elongation stress growth coefficient $\eta^+_{LVE}(t)$ is then determined from the master curve by inverting the angular frequency (ω) to the time domain (t) for the complex viscosity η^* and using the Trouton ratio of 3[7,52]. The curve is shown in Figure 3 and provides a basis for comparison with the transient elongation viscosity $\eta^+_E(t)$ measured in nonlinear experiments. This approach follows established rheological principles where the LVE data serve as a benchmark for the non-linear data.

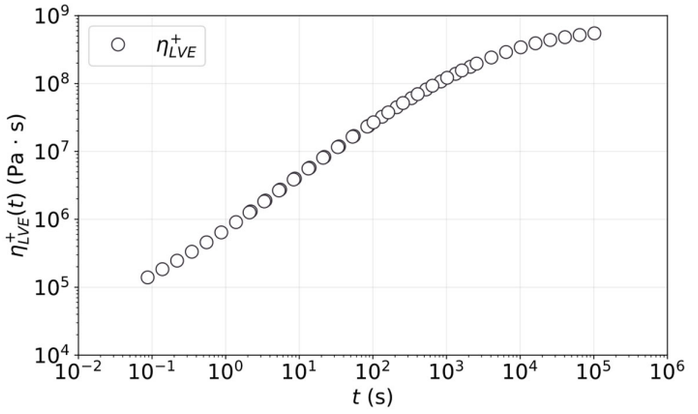

Figure 3. Linear viscoelastic envelope of as a function of time (t) at a reference temperature of 130°C.

3. Nonlinear Elongation Properties in the Melt State

Figure 4 presents the elongation stress growth coefficient (η_E^+) as a function of time at 130°C, determined with the three experimental devices. While the protocols to measure the elongation properties with the VADER 1000 and the ARES-EVF are well established, we apply the new protocol proposed in section II. 6. to measure the viscoelastic response of our sample with our Instron tensile machine. A good agreement across instruments is found for the three Hencky strain rates that we tested. This validates both the preparation/measurement protocols, and the edge-effect corrections applied to the Instron data at early times (or low Hencky strain values).

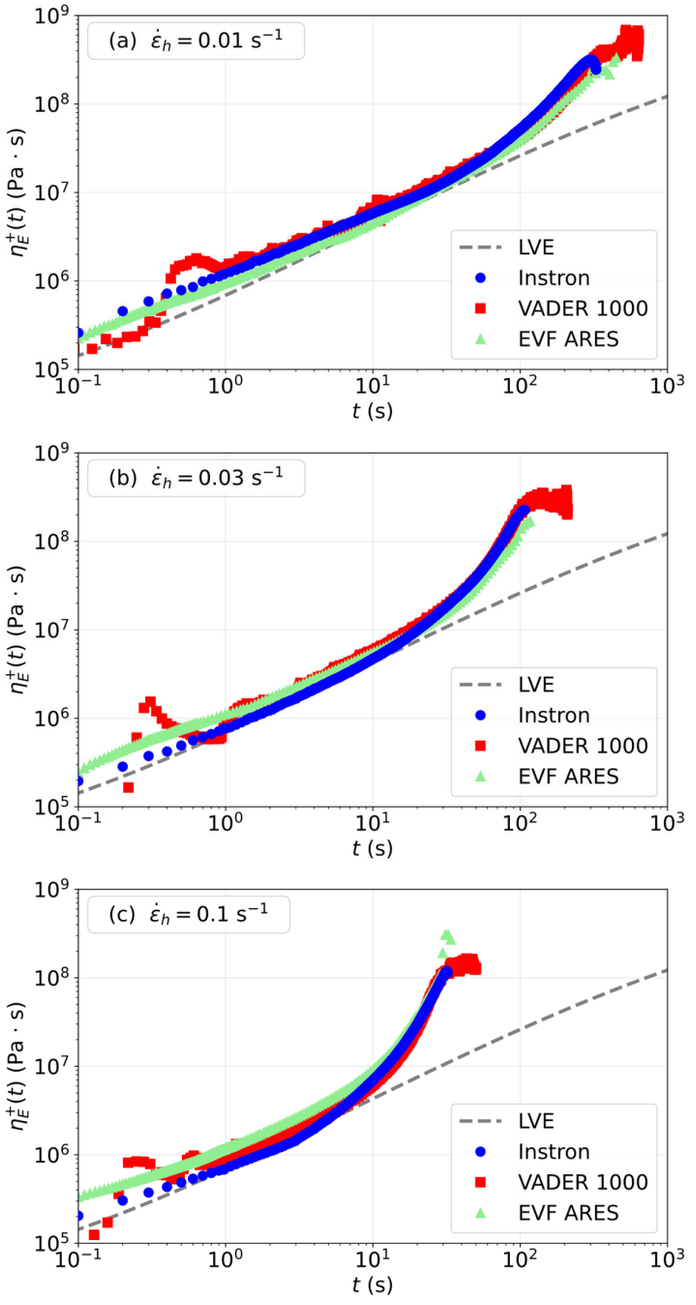

Figure 4. Extensional stress growth coefficient as a function of time at 130°C for the material considered at strain rates of (a) 0.01 s^{-1}, (b) 0.03 s^{-1}, and (c) 0.1 s^{-1} for the Instron (blue), VADER 1000 (red) and EVF (green). The dashed grey line is the linear viscoelastic envelope.

Complementary data in Figure 5 show the extensional stress growth function (true stress, σ_t) versus Hencky strain. It is observed that the VADER 1000 achieves the highest strain values ($\varepsilon_h \approx 6$), while the EVF and Instron systems are limited to $\varepsilon_h \approx 3.5$ and $\varepsilon_h \approx 3$, respectively. One should mention that these limitations arise from distinct causes:

1. The mechanical constraint for the EVF (*i.e.*, one drum rotation) causes the sample to overlap at Hencky strain values beyond $\varepsilon_h \approx 3.5$
2. The oven dimensions in our Instron setup restrict the maximum displacement to 200 mm, corresponding to $\varepsilon_h \approx 3$. However, this is not an intrinsic instrument limitation because with a larger oven, or with a material with a low T_g that does not require the use of the oven, the achievable Hencky strain increases to $\varepsilon_h \approx 4.5$ (950 mm displacement).

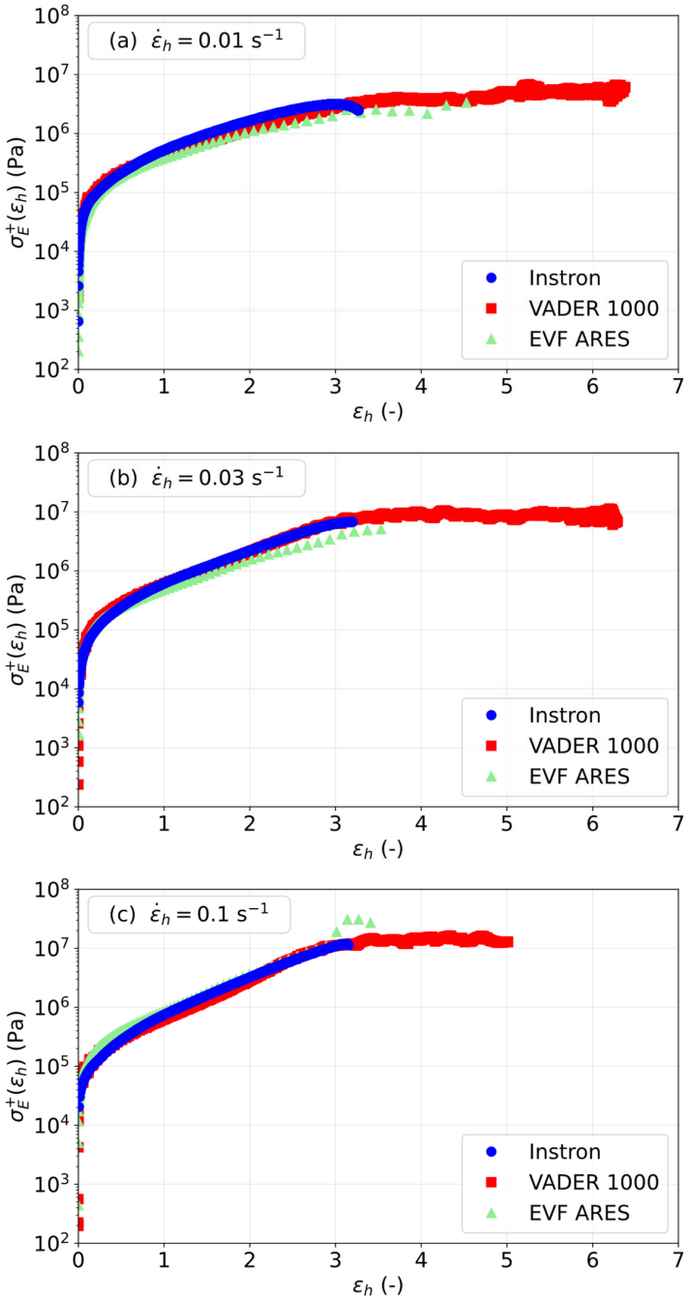

Figure 5. Extensional stress growth function as a function of Hencky strain at 130°C for the material considered at strain rates of (a) 0.01 s^{-1}, (b) 0.03 s^{-1}, and (c) 0.1 s^{-1} for the Instron (blue), VADER 1000 (red) and EVF (green).

An essential aspect of polymer characterization in extensional flow is the determination of the steady-state value. Indeed, the steady-state elongation viscosity is increasingly important in rheology, as nonlinear behavior under elongational deformations provides insights into molecular structural features that are not accessible through shear measurements. However, as observed in Figures 4 and 5, the experimental data from the VADER 1000 exhibit noise, particularly at low Hencky strain rates. In this regard, the Instron enables more accurate determination of the steady-state value as the signal-to-noise ratio is higher.

An additional advantage of the Instron system compared to the VADER 1000 system is its capability to clamp the sample. Unlike traditional systems, sample adhesion to the plates is not required, therefore it may significantly expand the range of materials that can be studied (*e.g.*, covalent adaptable networks).

Finally, the main interest in using the Instron device is its potential to characterize materials both above and below their T_g. As an example, first experiments were conducted at 110°C and 120°C, *i.e.*, at $T_{\text{exp}} \approx T_g + 5.6$°C and $T_{\text{exp}} \approx T_g + 15.6$°C respectively, on the same polystyrene sample. Hencky strain rates of 0.0002 s^{-1} at 110°C and of 0.0075 s^{-1} at 120°C were applied. These rates were selected to ensure an equivalent dynamic state between the two temperatures, based on the shift factors $C_1 = 8.99$ and $C_2 = 81.53$°C. A subsequent measurement was also conducted at 130°C and with a Hencky strain rate of 0.12 s^{-1} to further confirm the sensitivity of the method and to maintain thermo-rheologically equivalent conditions with the experiments performed at 110°C and 120°C. Results are shown in Figure 6, while Figure 7 compares the data at a reference temperature of 130°C, together with the data previously shown.

Our ongoing work is also focused on adapting the methodology for the solid state ($T < T_g$), where accounting for a variable Poisson's ratio, within a 3-dimensional framework, will be essential, as studied by Urayama and coworkers[36,37].

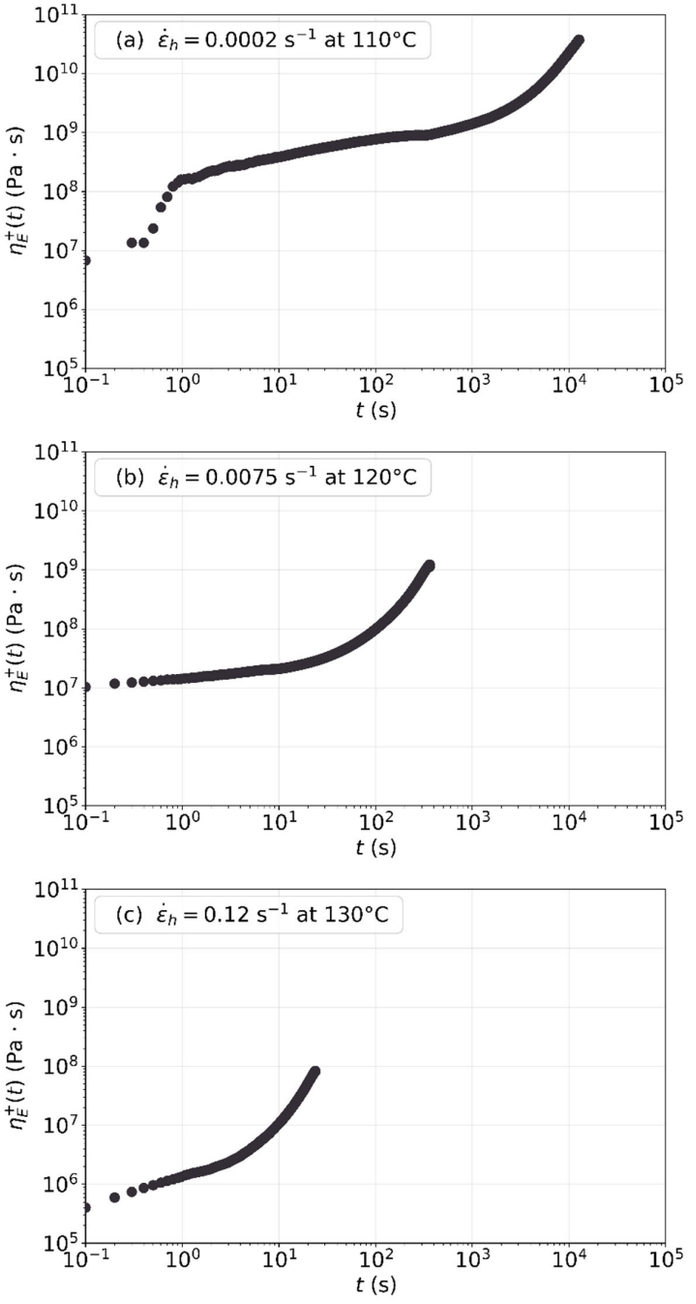

Figure 6. Extensional stress growth coefficient as a function of time for the material considered at a Hencky strain rate of (a) 0.0002 s^{-1} at 110°C, (b) 0.0075 s^{-1} at 120°C, and (c) 0.12 s^{-1} at 130°C.

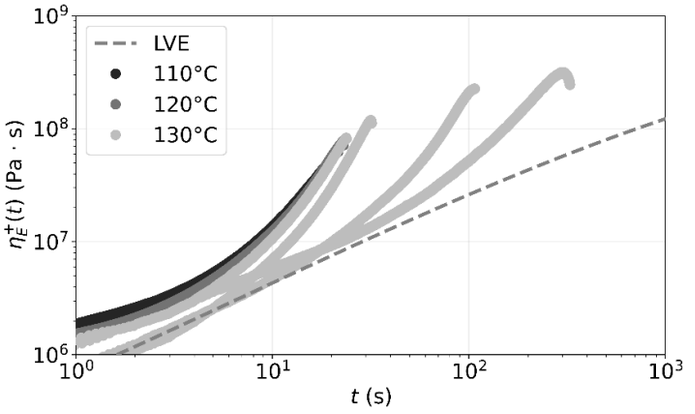

Figure 7. Overlay of the extensional stress growth coefficients acquired with the proposed protocol at 110°C, 120°C and 130°C. The data at 110°C and 120°C have been shifted to the reference temperature of 130°C.

It is observed that the shifted data obtained at 110°C (0.0002 s^{-1}) and 120°C (0.0075 s^{-1}) superimpose well onto the reference curve at 130°C (0.12 s^{-1}), as expected for thermo-rheologically simple samples. This successful superposition further validates the measurement method and establishes an interesting framework for connecting the elongational viscoelastic response of a polymer from the molten state to the solid state. Further analysis of the 110°C and 120°C data reveals a deviation from the linear viscoelastic (LVE) envelope at short times. We hypothesize that this stems from the increasing influence of solid-state mechanics near the glass transition. Furthermore, temperature control becomes critically important in this regime. As the sample deforms, its increasing surface-to-volume ratio enhances heat exchange with the oven environment, which can induce non-isothermal conditions and even temperature gradients within the sample. Such effects have been documented in extensional flows; for instance, Hassager et al.[53] estimated temperature gradients on the order of 2°C for polystyrene of similar molar mass when it is stretched at comparable strain rates to the one used

in this study, though the effect is less pronounced for the lower rates used in this study. Future work will aim to directly monitor sample temperature, for example using thermal imaging, to quantitatively account for these non-isothermal effects as described by Lin and Wang[54].

CONCLUSIONS

We have successfully implemented a method using the TestProfiler option in Instron's Bluehill software to control the engineering strain rate and have a constant Hencky strain rate over time. While only preliminary tests have been performed, we hope that "converting" a tensile machine into a rheometer will help to connect the mechanical characterization typically conducted at low temperatures with rheological characterization at high temperatures. Although we have only performed preliminary tests at temperatures of 110°C, 120°C and 130°C so far, it is evident that the Instron setup enables us to conduct experiments near the glass transition temperature, and potentially even below that threshold. Moreover, the availability of clamps and other accessories can enhance our ability to securely hold the sample in place during measurements. The Instron system also provides additional experimental capabilities by eliminating the requirement for sample adhesion to the rheometer plates. This unique feature not only enables characterization of non-adhesive materials but also extends the achievable range of strain rates compared to conventional systems.

The sensitivity of extensional rheology to molecular architecture and topology suggests this method could provide significant advantages for the detection and the quantification of long-chain branching in polyolefins (*e.g.*, LDPE and LLDPE), through the analysis of the strain-hardening behavior in the η_E^+ curves. Additionally, beyond conventional commercial polymers, this method shows a potential to characterize challenging polymers such as dynamic covalent networks (*i.e.*, vitrimers) by overcoming adhesion issues present in conventional instruments.

SUPPLEMENTARY MATERIAL

Supplementary information is available for this article. It includes the implementation of the loop-control feature with screenshots to show how the constant Hencky strain rate protocol was programmed and executed within the Instron Bluehill software environment.

ACKNOWEDGEMENTS

Alexandru Tudor Boborodea is a FRIA [Fonds pour la Formation à la Recherche dans l'Industrie et dans l'Agriculture] grantee of the Fonds de la Recherche Scientifique – FNRS (grant numbers: 40029831 and 40037308). Evelyne van Ruymbeke is Senior Researcher of the FNRS. Financial support by Communauté française de Belgique (Fédération Wallonie-Bruxelles) and Université catholique de Louvain (UCLouvain) for the Coordinated Research Project INVITE is gratefully acknowledged (grant number: 22/27-125).

DATA AVAILABILITY

The data that supports the findings of this study are available within the article and its supplementary material.

AUTHOR DECLARATIONS

The authors have no conflicts to disclose.

REFERENCES

[1] S. Costanzo, Q. Huang, G. Ianniruberto, G. Marrucci, O. Hassager, and D. Vlassopoulos, "Shear and Extensional Rheology of Polystyrene Melts and Solutions with the Same Number of Entanglements", Macromolecules, vol. 49, no. 10. American Chemical Society (ACS), pp. 3925–3935, 2016.

[2] T. Sridhar, R. K. Gupta, D. V. Boger, and R. Binnington, "Steady spinning of the Oldroyd fluid B II: Experimental results", Journal of Non-Newtonian Fluid Mechanics, vol. 21, no. 1. Elsevier BV, pp. 115–126, 1986.

[3] H. Münstedt, S. Kurzbeck, and J. Stange, "Importance of elongational properties of polymer melts for film blowing and thermoforming", Polymer Engineering & Science, vol. 46, no. 9. Wiley, pp. 1190–1195, 2006.

[4] T. Schweizer, "Measurement of the first and second normal stress differences in a polystyrene melt with a cone and partitioned plate tool," Rheologica Acta, vol. 41, no. 4, pp. 337–344, 2002.

[5] T. Schweizer, "Comparing cone-partitioned plate and cone-standard plate shear rheometry of a polystyrene melt," Journal of Rheology, vol. 47, no. 4, pp. 1071–1085, 2003.

[6] F. Snijkers and D. Vlassopoulos, "Cone-partitioned-plate geometry for the ARES rheometer with temperature control," Journal of Rheology, vol. 55, no. 6, pp. 1167–1186, 2011.

[7] R. B. Bird, R. C. Armstrong, and O. Hassager, "Dynamics of polymeric liquids, volume 1: Fluid mechanics", 2nd ed. Nashville, TN: John Wiley & Sons, 1987.

[8] Q. Huang, "When polymer chains are highly aligned: A perspective on extensional rheology," Macromolecules, vol. 55, no. 3, pp. 715–727, 2022.

[9] E. Jenckel and K. Ueberreiter, "Über Polystyrolgläser verschiedener Kettenlänge", Zeitschrift für Physikalische Chemie, vol. 182A, no. 1. Walter de Gruyter GmbH, pp. 361–383, May 01, 1938.

[10] J. Meißner, "Rheometer zur Untersuchung der deformationsmechanischen Eigenschaften von Kunststoff-Schmelzen unter definierter Zugbeanspruchung", Rheologica Acta, vol. 8, no. 1. Springer Science and Business Media LLC, pp. 78–88, 1969.

[11] J. Meissner and J. Hostettler, "A new elongational rheometer for polymer melts and other highly viscoelastic liquids", Rheologica Acta, vol. 33, no. 1. Springer Science and Business Media LLC, pp. 1–21, 1994.

[12] J. Meissner, "Development of a Universal Extensional Rheometer for the Uniaxial Extension of Polymer Melts", Transactions of the Society of Rheology, vol. 16, no. 3. Society of Rheology, pp. 405–420, Sep. 01, 1972.

[13] J. Meissner, T. Raible, and S. E. Stephenson, "Rotary Clamp in Uniaxial and Biaxial Extensional Rheometry of Polymer Melts", Journal of Rheology, vol. 25, no. 1. Society of Rheology, pp. 1–28, Feb. 1981.

[14] M. L. Sentmanat, "Dual windup drum extensional rheometer", US6691569, Jul. 31, 2002 (priority date).

[15] M. L. Sentmanat, "Miniature universal testing platform: from extensional melt rheology to solid-state deformation behavior", Rheologica Acta, vol. 43, no. 6. Springer Science and Business Media LLC, pp. 657–669, Aug. 12, 2004.

[16] M. Sentmanat, B. N. Wang, and G. H. McKinley, "Measuring the transient extensional rheology of polyethylene melts using the SER universal testing platform," Journal of Rheology, vol. 49, no. 3, pp. 585–606, 2005.

[17] O. Delgadillo-Velázquez, S. G. Hatzikiriakos, and M. L. Sentmanat, "Thermorheological properties of LLDPE/LDPE blends", Rheologica Acta, vol. 47, no. 1. Springer Science and Business Media LLC, pp. 19–31, May 07, 2007.

[18] C. Hadinata et al., "Elongation-induced crystallization of a high molecular weight isotactic polybutene-1 melt compared to shear-induced crystallization," Journal of Rheology, vol. 51, no. 2, pp. 195–215, 2007.

[19] Parisi et al., "Alternative use of the Sentmanat Extensional Rheometer to investigate the rheological behavior of industrial rubbers at very large deformations," Rubber Chemistry and Technology, vol. 95, no. 2, pp. 241–276, 2022.

[20] M. Khabazian Esfahani, M. G. Schußmann, J. A. Rodríguez Agudo, J. Haeberle, J. Läuger, C. Georgantopoulos, V. Hirschberg, L. Faust, Q. Huang, M. Wilhelm, "Measuring tensile stress growth coefficient $\eta_E^+(t)$ up to a Hencky Strain of $\varepsilon = 8.7$, introducing the 2D-UXF method" currently under revision (Rheologica Acta)

[21] J. Kaschta, H. Münstedt, A. Co, G. L. Leal, R. H. Colby, and A. J. Giacomin, "Measuring the elongational properties of polymer melts-a simple task?," in AIP Conference Proceedings, 2008.

[22] B. Li, W. Yu, X. Cao, and Q. Chen, "Horizontal extensional rheometry (HER) for low viscosity polymer melts," J. Rheol., vol. 64, no. 1, pp. 177–190, 2020.

[23] J. Meissner, "Experimental aspects in polymer melt elongation rheometry", Chemical Engineering Communications, vol. 33, no. 1–4. Informa UK Limited, pp. 159–180, Feb. 1985.

[24] H. Münstedt, "Viscoelasticity of polystyrene melts in tensile creep experiments", Rheologica Acta, vol. 14, no. 12. Springer Science and Business Media LLC, pp. 1077–1088, Dec. 1975.

[25] T. Sridhar, V. Tirtaatmadja, D. A. Nguyen, and R. K. Gupta, "Measurement of extensional viscosity of polymer solutions", Journal of Non-Newtonian Fluid Mechanics, vol. 40, no. 3. Elsevier BV, pp. 271–280, Nov. 1991.

[26] A. Bach, H. K. Rasmussen, P.-Y. Longin, and O. Hassager, "Growth of non-axisymmetric disturbances of the free surface in the filament stretching rheometer: experiments and simulation", Journal of Non-Newtonian Fluid Mechanics, vol. 108, no. 1–3. Elsevier BV, pp. 163–186, Dec. 2002.

[27] A. Bach, H. K. Rasmussen, and O. Hassager, "Extensional viscosity for polymer melts measured in the filament stretching rheometer", Journal of Rheology, vol. 47, no. 2. Society of Rheology, pp. 429–441, Mar. 2003.

[28] J. M. Román Marín, J. Kjøbsted Huusom, N. J. Alvarez, Q. Huang, H. K. Rasmussen b, A. Bach, A. L. Skov, O. Hassager, "A control scheme for filament stretching rheometers with application to polymer melts", Journal of Non-Newtonian Fluid Mechanics, vol. 194. Elsevier BV, pp. 14–22, Apr. 2013.

[29] J. M. Dealy, Structure and rheology of molten polymers: From structure to flow behavior and back again. Hanser Gardner Publications, 2018.

[30] Gabriel, C., Münstedt, H. Strain hardening of various polyolefins in uniaxial elongational flow. J. Rheol. 47, pp. 619–630, 2003.

[31] Torres, E., Si-Wan, L., Costeux, S., Dealy, J.M. "Branching structure and strain hardening of branched metallocene polyethylenes". J. Rheol. 59, pp. 1151–1172, 2015.

[32] E. van Ruymbeke, E. B. Muliawan, S. G. Hatzikiriakos, T. Watanabe, A. Hirao, and D. Vlassopoulos, "Viscoelasticity and extensional rheology of model Cayley-tree polymers of different generations," J. Rheol. (N. Y. N. Y.), vol. 54, no. 3, pp. 643–662, 2010.

[33] H. Lentzakis et al., "Uniaxial extensional rheology of well-characterized comb polymers," J. Rheol. (N. Y. N. Y.), vol. 57, no. 2, pp. 605–625, 2013.

[34] Y. Obata, S. Kawabata, and H. Kawai, "Mechanical properties of natural rubber vulcanizates in finite deformation," J. Polym. Sci. A-2 Polym. Phys., vol. 8, no. 6, pp. 903–919, 1970.

[35] S. Kawabata, M. Matsuda, K. Tei, and H. Kawai, "Experimental survey of the strain energy density function of isoprene rubber vulcanizate," Macromolecules, vol. 14, no. 1, pp. 154–162, 1981.

[36] T. Kawamura, K. Urayama, and S. Kohjiya, "Multiaxial deformations of end-linked poly(dimethylsiloxane) networks. 1. Phenomenological approach to strain energy density function," Macromolecules, vol. 34, no. 23, pp. 8252–8260, 2001.

[37] K. Urayama, T. Kawamura, and S. Kohjiya, "Multiaxial deformations of end-linked poly(dimethylsiloxane) networks. 2. Experimental tests of molecular entanglement models of rubber elasticity," Macromolecules, vol. 34, no. 23, pp. 8261–8269, 2001.

[38] Q. Huang et al., "Concentrated polymer solutions are different from melts: Role of entanglement molecular weight," Macromolecules, vol. 46, no. 12, pp. 5026–5035, 2013.

[39] Q. Huang, N. J. Alvarez, Y. Matsumiya, H. K. Rasmussen, H. Watanabe, and O. Hassager, "Extensional rheology of entangled polystyrene solutions suggests importance of nematic interactions," ACS Macro Lett., vol. 2, no. 8, pp. 741–744, 2013.

[40] Q. Huang, L. Hengeller, N. J. Alvarez, and O. Hassager, "Bridging the gap between polymer melts and solutions in extensional rheology," Macromolecules, vol. 48, no. 12, pp. 4158–4163, 2015.

[41] G. R. Cotten, and J. L. Thiele, "Influence of carbon black on processability of rubber stocks III. Extensional viscosity, Rubber Chem. Technol., vol. 51, no. 4, pp. 749-763, 1978.

[42] M. N. Charalambides, L. Wanigasooriya, J. G. Williams, S. M. Goh, and S. Chakrabarti, "Large deformation extensional rheology of bread dough", Rheologica Acta, vol. 46, no. 2. Springer Science and Business Media LLC, pp. 239–248, Apr. 29, 2006.

[43] C. G'Sell, J. M. Hiver, and A. Dahoun, "Experimental characterization of deformation damage in solid polymers under tension, and its interrelation with necking", International Journal of Solids and Structures, vol. 39, no. 13–14. Elsevier BV, pp. 3857–3872, Jun. 2002.

[44] P. J. Hine, A. Duckett, and D. J. Read, "Influence of molecular orientation and melt relaxation processes on glassy Stress−Strain behavior in polystyrene," Macromolecules, vol. 40, no. 8, pp. 2782–2790, 2007.

[45] Y. W. Ooi and T. Sridhar, "Extensional rheometry of fluid S1," J. Nonnewton. Fluid Mech., vol. 52, no. 2, pp. 153–162, 1994.

[46] F. P. A. van Berlo, R. Cardinaels, G. W. M. Peters, and P. D. Anderson, "Towards a universal shear correction factor in filament stretching rheometry," Rheol. Acta, vol. 60, no. 11, pp. 691–709, 2021.

[47] S. H. Spiegelberg, D. C. Ables, and G. H. McKinley, "The role of end-effects on measurements of extensional viscosity in filament stretching rheometers," J. Nonnewton. Fluid Mech., vol. 64, no. 2–3, pp. 229–267, 1996.

[48] H. K. Rasmussen, A. G. Bejenariu, O. Hassager, and D. Auhl, "Experimental evaluation of the pure configurational stress assumption in the flow dynamics of entangled polymer melts," Journal of Rheology, vol. 54, no. 6, pp. 1325–1336, 2010.

[49] D. M. Hoyle et al., "Transient overshoot extensional rheology of long-chain branched polyethylenes: Experimental and numerical comparisons between filament stretching and cross-slot flow," Journal of Rheology, vol. 57, no. 1, pp. 293–313, 2013.

[50] Q. Huang, M. Mangnus, N. J. Alvarez, R. Koopmans, and O. Hassager, "A new look at extensional rheology of low-density polyethylene," Rheol. Acta, vol. 55, no. 5, pp. 343–350, 2016.

[51] T. Shahid, C. Clasen, F. Oosterlinck, and E. van Ruymbeke, "Diluting entangled polymers affects transient hardening but not their steady elongational viscosity", Macromolecules, vol. 52, no. 6, pp. 2521–2530, 2019.

[52] G. H. McKinley and T. Sridhar, "Filament-stretching rheometry of complex fluids," Annual Review of Fluid Mechanics, vol. 34, no. 1, pp. 375–415, 2002.

[53] A. Bach, K. Almdal, H. K. Rasmussen, and O. Hassager, "Elongational viscosity of narrow molar mass distribution polystyrene," Macromolecules, vol. 36, no. 14, pp. 5174–5179, 2003

[54] P. Lin and S.-Q. Wang, "Erratum: Nonisothermal condition in past melt extension experiments," J. Rheol., vol. 59, no. 5, pp. 1329–1334, 2015.