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Measurements and modeling of temperature-strain rate dependent uniaxial and planar extensional viscosities for branched LDPE polymer melt

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A B S T R A C T

In this work, novel rectangle and circular orifice (zero-length) dies have been utilized for temperature-strain rate dependent planar and uniaxial extensional viscosity measurements for the LDPE polymer melt by using standard twin bore capillary rheometer and Cogswell model and the capability of five different constitutive equations (novel generalized Newtonian model, original Yao model, extended Yao model, modified White-Metzner model, modified Leonov model) to describe the measured experimental data has been tested. It has been shown that chain branching causes the strain hardening occurrence in both uniaxial and planar extensional viscosities and its maximum is shifted to the higher strain rates if the temperature is increased. The level of uniaxial extensional strain hardening for the branched LDPE sample has been found to be higher in comparison with the planar extensional viscosity within wide range of temperatures.

1. Introduction

The extensional viscosity represents key rheological parameter allowing understanding the molecular structure of the polymers as well as polymer processing at which the polymer melts are stretched [1–27]. Due to the fact that generation and control of the extensional flow is difficult, experimental determination of the extensional viscosity is a problem [28–32]. Probably the most challenging rheological task is experimental determination of planar extensional viscosity as one can see from very small numbers of experimental data available in the open literature [1,2,12–16,33]. With the aim to understand this important rheological parameter in more detail, in this work, novel rectangle and circular orifice dies [34,35] have been utilized for planar and uniaxial extensional viscosity measurements for branched LDPE by using standard twin bore capillary rheometer and Cogswell model [6,12] and the capability of five different constitutive equations [36–42] to describe the measured experimental data has been tested.

2. Experimental

In this work, extrusion coating, branched LDPE CA820 from Borealis Polyolefine was used. Linear viscoelastic properties (storage modulus, loss modulus, and complex viscosity) were measured on Advanced Rheometric Expansion System ARES 2000 (Rheometrics Scientific, USA) in parallel plates geometry mode (plates diameter of 25 mm) within 0.1 rad/s up to 100 rad/s frequency range at 1% shear strain to guarantee linear viscoelasticity regime only. In order to get storage and loss moduli for a large range of frequencies, the measurements were performed in a wide range of temperatures (130–250 °C). Possible degradation at high temperatures was suppressed by inert nitrogen atmosphere. The well-known time-temperature-superposition principle was used to generate master curves for particular reference temperature. Twin-bore capillary rheometer Rosand RH7-2 (Rosand Precision, United Kingdom) was used for experimental determination of uniaxial and planar extensional viscosities by using long as well as orifice dies having the abrupt contraction (i.e. 180° entrance angle) and circular/rectangular shape (see Figs. 1–2). The main advantage of both utilized orifice dies is the open downstream region design which eliminates any possibility for artificial pressure increase due to

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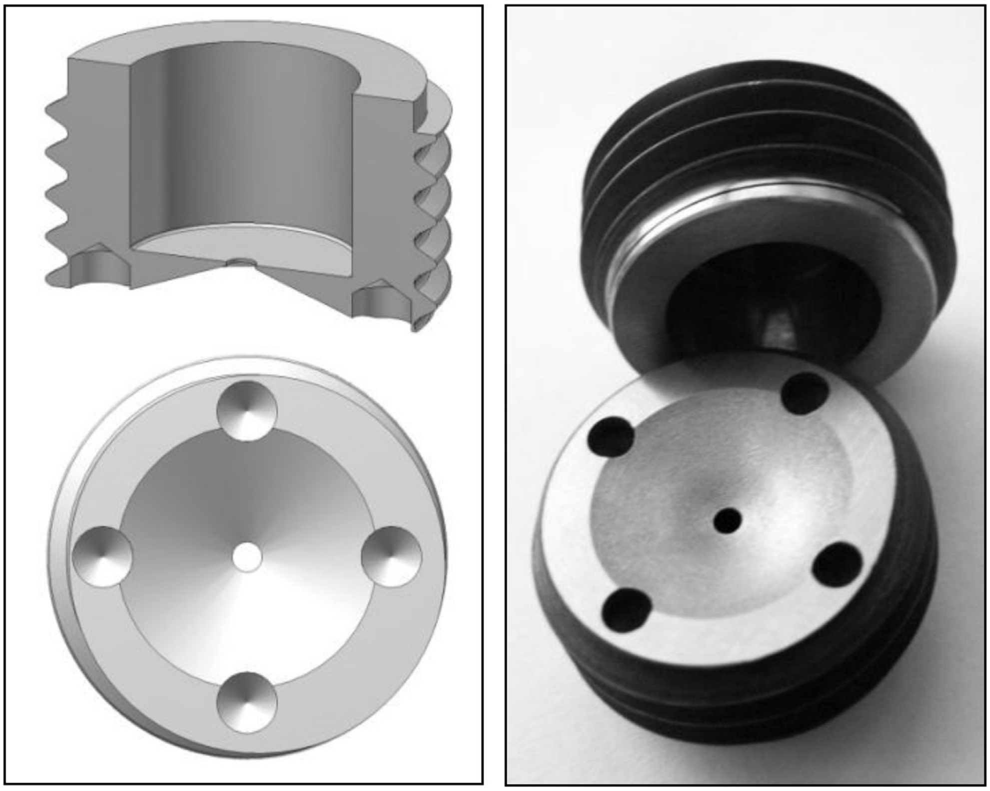

Fig. 1. Patented circular orifice die with abrupt entry utilized for uniaxial extensional viscosity measurements developed in Refs. [11,34].

polymer melt touching the downstream wall [34,35]. Temperature measurement and control was performed at three separate zones in the barrel by using platinum resistance thermometers (PRTs) via microprocessor based board contained within the electronics rack communicating with the computer over a serial interface. Software running on the temperature board implements a three term (PID) control algorithm allowing independent variation of power for the three cuff barrel heaters with different heights and power ratings (1000 W, 600 W and 1000 W).

The uniaxial and planar extensional viscosities have been determined through entrance pressure drop measurements by using the Cogswell model [6,12] (see Table 1). In this table, $P_{0,\mathrm{U}}$ and $P_{0,\mathrm{P}}$ represents the entrance pressure drop measured on circular and rectangular orifice die, respectively, Q is the volume flow rate, R is the capillary die radius, w and h is the width and the gap size of the rectangle die, respectively, $P_{\mathrm{L,U}}$ and $P_{\mathrm{L,P}}$ represents the pressure drop through a long die having circular and rectangular shape, respectively, L is the length of the long die. It should be mentioned that the long die has $L/(2R) = 16$ (or $L/h = 16$) whereas the orifice die has $L/(2R) = 0.1208$ (or $L/h = 0.1208$) as suggested in Ref. [10].

The maximum attainable extensional strain during abrupt contraction flow in the capillary rheometer can be calculated as

$$\varepsilon_{\max} = \ln\left(\frac{A_b}{A_d}\right) \tag{1}$$

where A_b and A_d is the cross-sectional area of the barrel and the die, respectively [43]. The barrel diameter of the rheometer is 15 mm, diameter of the circular die is 2 mm and gap size of the rectangular die is 0.5 mm. According to Eq. (1) ε_{max} is 4 and 3.6 for circular and rectangle orifice dies, respectively, and thus the average strain achievable in both dies can be considered to be comparable. Importance of comparing the extensional viscosity at the same strain level is discussed in Refs. [43–45]. Note that high shear rate rheology was evaluated via 1 mm diameter capillary dies.

3. Theoretical

3.1. Generalized Newtonian fluid model

In this work, recently proposed generalized Newtonian fluid model has been utilized [36,37]:

$$\tau = 2\eta\left(I_{|\mathrm{D}|}, II_{\mathrm{D}}, III_{\mathrm{D}}\right)D \tag{2}$$

where τ means the extra stress tensor, D represents the deformation rate tensor and η stands for the viscosity, which is not constant (as in the case of standard Newtonian law), but it is allowed to vary with the first invariant of the absolute value of deformation rate tensor $I_{|\mathrm{D}|} = tr(|D|)$, (where $|D|$ is defined as $\sqrt{D{\cdot}D}$) as well as on the second $II_{\mathrm{D}} = 2tr(D^2)$, and third, $III_{\mathrm{D}} = det(D)$, invariants of D according to Eq. (3)

$$\eta\left(I_{|\mathrm{D}|}, II_{\mathrm{D}}, III_{\mathrm{D}}\right) = A^{1-f\left(I_{|\mathrm{D}|}, II_{\mathrm{D}}, III_{\mathrm{D}}\right)}\eta(II_{\mathrm{D}})^{f\left(I_{|\mathrm{D}|}, II_{\mathrm{D}}, III_{\mathrm{D}}\right)} \tag{3}$$

where $\eta(II_D)$ is given by the well known Carreau-Yasuda model (with infinite viscosity η_∞ equal 0), Eq. (4) and $f(I_{|\mathrm{D}|}, II_{\mathrm{D}}, III_{\mathrm{D}})$ is given by Eq. (5)

$$\eta(II_{\mathrm{D}}) = \frac{\eta_0 a_{\mathrm{T}}}{\left[1 + \left(\lambda a_{\mathrm{T}}\sqrt{II_{\mathrm{D}}}\right)^a\right]^{\left(\frac{1-n}{a}\right)}} \tag{4}$$

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