Constitutive Model Parameters Estimation from Rheotens Steady State and Resonance Characteristics

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ABSTRACT: This paper presents a method to determine the parameters in a polymer constitutive model using data obtained from a Rheotens experiment. The novelty of the suggested approach is the simultaneous fitting of model parameters to different types of data given by Rheotens, i.e., force-velocity curve, onset of draw resonance and frequency of oscillations. To determine the onset and frequency of oscillations, a stability analysis is exploited and the spectrum of a quasi-hyperbolic differential operator is calculated. The proposed approach is efficient and accurate. To demonstrate its applicability and consistency, a modified Giesekus constitutive model is chosen and model parameters are fitted to the Rheotens data for three different types of polymer (LLDPE, PP, PS).

KEYWORDS: Rheology, Modeling, Parameter Estimation, Rheotens, Draw Resonance

1 INTRODUCTION

Polymer process simulations aim to discover the best processing options for a polymer material without running the actual processes. In practice, the simulations are based on simple rheological models that do not fully describe the complexity of the real polymer behavior. Complex polymer constitutive models are under-used, though being implemented, because their parameters are unknown a priori and hard to estimate. A way to estimate them is to apply the model in question to simulate certain well-chosen experimental data.

This paper addresses the Rheotens experiment as an appropriate candidate for model parameter estimation. The experiment is based on drawing a molten polymer filament by two rotating wheels, thereby inducing an elongational deformation. There are two procedures for running Rheotens: the ”acceleration mode”, where the velocity of the wheels is increased according to a specified constant acceleration and the force is measured as a function of velocity; the ”constant mode”, where the velocity of the wheels is kept piece-wise constant and the force is measured as a function of time. The graphs depicting measurement data show force oscillations after a certain drawing velocity is reached. These oscillations represent stretching-related instability, called ”draw resonance”. Thus, Rheotens establishes two different polymer operational regimes: stable and unstable.

Up to now, in literature only the acceleration mode is considered, and from the resulting curve only the part related to stable operation is fitted. This paper studies the constant mode, demonstrating its applicability and the importance of its results for parameter estimation. The paper describes a method to determine model parameters by using the results of a stable operating regime from the acceleration mode and the instability related results from the constant mode. Simulations of a linear low density polyethylene (LLDPE), polypropylene (PP), and polystyrene (PS) with a modified Giesekus constitutive model show accuracy and efficiency of the method.

2 RHEOTENS EXPERIMENT

2.1 Description

In Rheotens, a polymer is extruded through a circularly shaped small die and drawn down by take-up wheels. The dimensions of the die hole are fixed. Although usually fixed at 100 mm, the spin line length

may vary experiment by experiment. At the take-up point the drawing force is measured. The measurement frequency can vary from 5 Hz to 100 Hz. The total number of measurements can go up to 10000. The mass throughput Q, the extrusion temperature of the polymer T_{extr}, and the temperature of the surrounding air T_{air} are process parameters. The final velocity of the filament u_f is directly related to the wheels velocity.

2.2 Experimental Results

In Figures 1 and 2, the graphs corresponding to the acceleration and the constant mode of operating Rheotens are given for an LLDPE type of polymer at the same conditions.

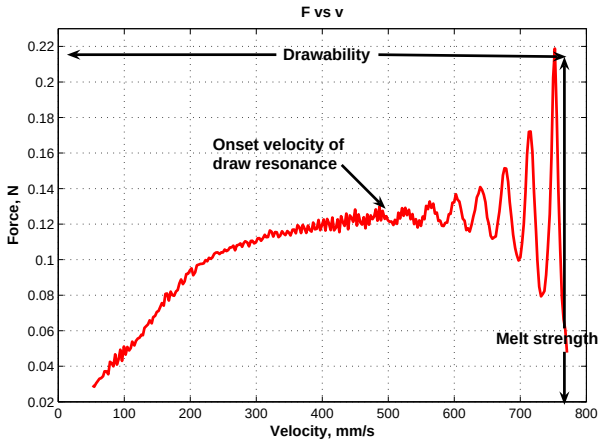

Figure 1: *Acceleration mode,* $a = 24$ *mm/s*^{2}, $L = 0.1$*m. LLDPE type of polymer:* $Q = 600$ *g/h,* $T_{extr} = 190^oC$

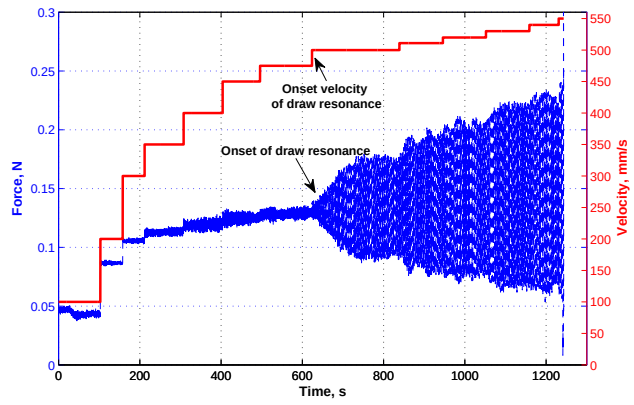

Figure 2: *Constant mode: draw velocity is piece-wise constant,* $L = 0.1$ *m. LLDPE type of polymer:* $Q = 600$ *g/h,* $T_{extr} = 190^oC$

The measurements depicted in Figure 1 are performed at a sampling frequency of 100 Hz and the ones in Figure 2 at a sampling frequency of 10 Hz. Figure 2 shows that at low velocities the variation in force level is small, in the range of measurement error $\pm 10^{-3}$ N. As the velocity reaches the critical value of 500 mm/s, the amplitude of force oscillations increases significantly, indicating that draw resonance is present. Increase of the velocity above the critical value yields further increase of the amplitude of oscillations. We consider the data presented in Figure 2 as a discrete-time signal with a sampling frequency of 10 Hz. By applying the Discrete Fourier Transformation to the data taken from the segments related to the velocity values of 500 mm/s (critical velocity), 510 mm/s and 520 mm/s, we obtain the amplitude spectra depicted in Figure 3 in dB. The scale is such that value of 0 dB denotes the amplitude equal to 0.001 N. In all spectra we observe a harmonic complex with the basic frequency of 0.58 Hz, which corresponds to the frequency of the force oscillations in draw resonance.

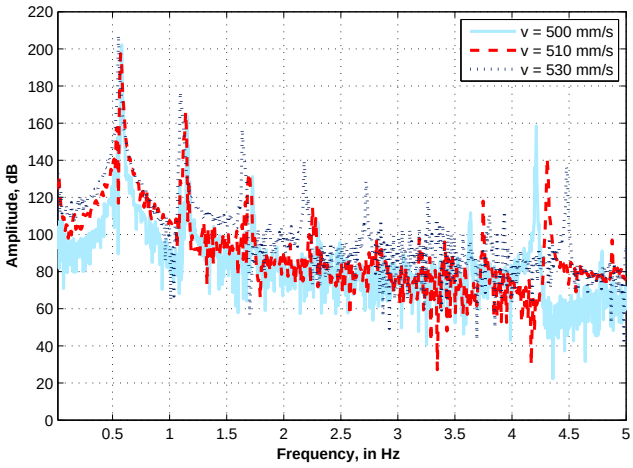

Figure 3: *Amplitude spectra of force versus frequency for the draw velocities of 500, 510 and 530 mm/s.*

Furthermore, similar analysis is performed for the rest of the data shown in Figure 2. We conclude that the harmonic complex has disappeared from the spectra related to the non-critical velocities.

Thus, data obtained from the constant mode of operating Rheotens brings additional information that can be used in future studies and numerical simulations. We combine the constant mode data with the acceleration mode data to estimate constitutive model parameters.

3 MATHEMATICAL MODELING

3.1 Modeling Assumptions

The flow domain of the polymer is described by cylindrical coordinates $(r,\, \theta,\, z)$. The origin of the coordinate system is taken at the point of maximal swell. The flow is assumed to be rotationally symmetric, i.e., independent of θ. The thickness of the fiber, given by its radius, is in the order of 10^{-2} smaller than its length. Thus, we approximate the fiber by a slender jet and consider the normal stress at the fiber surface to be zero, $\sigma_{rr} = 0$. The flow field of the polymer is mainly elongational. The velocity component in the extension direction is uniform over the cross-section. Further, the polymer is incompressible. Forces acting during the experiment are drawing force, gravity, inertia, and air drag. The surface tension is neglected as being relatively small with respect to the drawing force.

3.2 Equations

The governing system of equations consists of conservation of mass (1), equation of motion (2), conservation of energy (3), and polymer constitutive equations (4, 5),

$$2\frac{\partial R}{\partial t} + 2u_z\frac{\partial R}{\partial z} + R\frac{\partial u_z}{\partial z} = 0 \tag{1}$$

$$\rho R u_z\frac{\partial u_z}{\partial z} = R\frac{\partial \sigma_{zz}}{\partial z} + 2\sigma_{zz}\frac{\partial R}{\partial z} - 2C_f\rho_a u_z^2 + \rho g R \tag{2}$$

$$\rho C_p u_z\frac{\partial T}{\partial z} = -\frac{2h_c}{R}(T - T_{air}) + \sigma_{zz}\frac{\partial u_z}{\partial z} \tag{3}$$

$$\lambda\left(\frac{\partial c_{rr}}{\partial t} + u_z\frac{\partial c_{rr}}{\partial z} + c_{rr}\frac{\partial u_z}{\partial z}\right) = \frac{k_bT}{K_0}\left((1-\alpha) + \alpha\frac{K_0}{k_bT}Ec_{rr}\right)\left(1 - \frac{K_0}{k_bT}Ec_{rr}\right), \tag{4}$$

$$\lambda\left(\frac{\partial c_{zz}}{\partial t} + u_z\frac{\partial c_{zz}}{\partial x} - 2c_{zz}\frac{\partial u_z}{\partial z}\right) = \frac{k_bT}{K_0}\left((1-\alpha) + \alpha\frac{K_0}{k_bT}Ec_{zz}\right)\left(1 - \frac{K_0}{k_bT}Ec_{zz}\right). \tag{5}$$

The time t and the height z are the independent variables. Dependent variables are the radius R, velocity in elongation direction u_z, normal stress σ_{zz}, and temperature of the filament T. In (2), C_f is a dimensionless air drag coefficient, taken according to [1]; in (3), C_p is heat capacity of the polymer, that we assume to depend linearly on temperature, [2]; h_c is the convective heat transfer coefficient. Modeling the constitutive behavior of the polymer, we consider a modified Giesekus model, [3]. In (4, 5), λ is the polymer relaxation time; it is assumed to depend on temperature according to Arrhenius law. Variables c_{rr} and c_{zz} are the non-zero conformation tensor components. The relation between the conformation tensor $\mathcal{C}$ and the stress tensor σ is defined as

$$\sigma = -p\mathcal{I} + \mathcal{T} \tag{6}$$

$$\text{with} \quad \mathcal{T} = n(K_0E\mathcal{C} - k_bT\mathcal{I})\,. \tag{7}$$

In (7), n is the number of molecules per unit volume; E is the non-linear spring force factor that accounts for a finite chain extensibility, taken according to [4]; k_b is the Boltzmann constant; K_0 is the spring constant,

$$K_0 = \frac{3k_bT_0}{N_0l^2}\,. \tag{8}$$

Here N_0 is a model parameter that represents the average number of flexible links of length l contained in a molecular chain. Also, in (4, 5), α is the model parameter representing molecular mobility. Values of α and N_0 need to be estimated.

3.3 Boundary conditions

From the process conditions, we deduce the boundary conditions at $z = 0$ and $z = L$:

$$u_z(0,t) = u_0, \quad R(0,t) = R_0, \quad T(0,t) = T_{extr}, \tag{9}$$

$$c_{rr}(0,t) = c_{rr}^{eq}, \quad u_z(L,t) = u_f. \tag{10}$$

The boundary value of the conformation tensor component c_{rr}^{eq} is taken at equilibrium; it is the value found from the condition that all extra stresses are relaxed at equilibrium. The radius R_0 is corrected for extrudate swell. In our study, extrudate swell is measured experimentally according to the following method: the polymer extruded through the die was allowed to fall freely; the radius of the resulting filament was measured at the end of the spin line.

4 SIMULATIONS

For numerical simulations of the presented model, we use the technique described in [5]. The simulations are performed for an LLDPE, PP, and PS. The input values for the simulations are given in Table 1. In our simulations, we use two values of the relaxation time: λ_1 is used for the Rheotens curve calculation and λ_2 is used for calculation of the onset velocity of draw resonance and frequency of force oscillations at draw resonance. The value of the viscosity is kept the same in both cases. Concluding from our previous research, where Newtonian model simulations were carried out, we take viscosity corresponding to the shear rate values in the vicinity of 1 s^{-1}. The value of λ_1 is taken

accordingly. The value of λ_2 is fitted together with the modified Giesekus model parameters α and N_0.

Table 1: *Process and polymer experimental conditions and resulting values of modified Giesekus model parameters*

	LLDPE	PP	PS	
T_{extr} (^{o}C)	190	220	220	
T_{air} (^{o}C)	20	20	20	
Q (kg/h)	0.6	0.6	0.6	
ρ (kg/m^3)	760	748.5	957	
E_a (kJ/mol)	34.5	41.4	45	
L (mm)	100	100	50	
R_{die} (mm)	1.25	1.25	1.25	
R_0 (mm)	1.8	2.0	1.9	
λ_1 (s)	1	2.8	1.4	
λ_2 (s)	0.024	0.060	0.009	
η $(Pa \cdot s)$	6000	33600	7600	
α	0.19	0.31	0.29	
N_0	5	3	14	

The estimated values of the model parameters are also specified in Table 1. Those values are result of optimization giving the best match between the simulated and the experimental Rheotens data. Figure 4 presents the best fit between the simulated and the experimental Rheotens curves for all three materials. All experimental curves are corrected for the influence of gravity according to [3]. Table 2 presents the best fit between the simulated and the experimental data obtained from the Rheotens constant mode. Experimental onset velocity is determined by the measurement resolution, which was different in every case. For instance, for LLDPE at a velocity of 475 mm/s the process is stable, and at a velocity of 500 mm/s, the next taken, the process is unstable. Thus, the critical velocity is reported in range of $475 - 500$ mm/s.

5 CONCLUSIONS

The results presented in the previous section show the accuracy and efficiency of the suggested methodology for model parameters estimation. Analysis of the data obtained by the Rheotens constant mode, when the velocity of the pulling wheels is piece-wise constant, proves the importance of those experimental data not only for the parameter determination problem, but for many other possible applications.

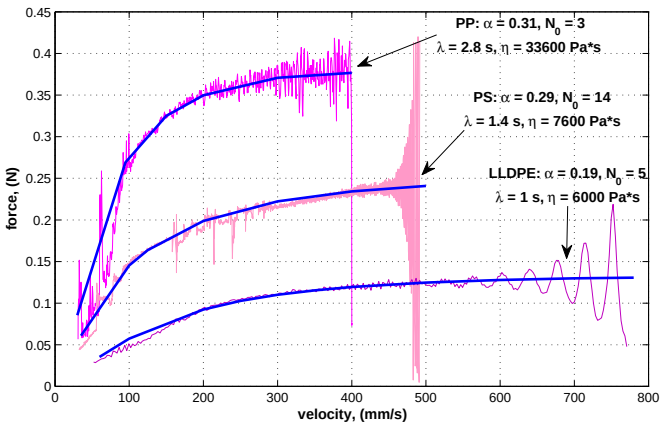

Figure 4: *Demonstration of the Rheotens curves fitted under the steady state conditions for LLDPE, PP, and PS*

Table 2: *Critical velocity at onset of draw resonance and frequency of force oscillations: comparison of experimentally measured values (exp) with the simulated ones (sim)*

	Onset velocity (mm/s)		Frequency (Hz)	
	exp	sim	exp	sim
LLDPE	475 - 500	480	0.58	0.59
PP	290 - 300	310	0.44	0.44
PS	400 - 425	430	1.15	1.17

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