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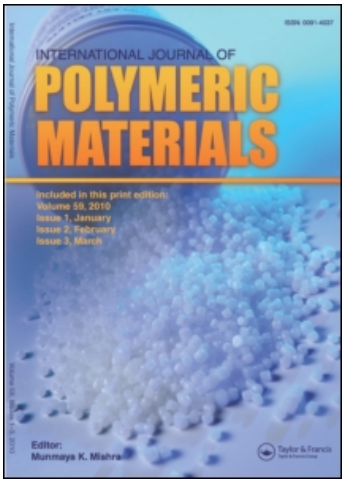

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Investigation of Unsteady Elongational Flow of Polymer Melts

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Investigation of Unsteady Elongational Flow of Polymer Melts

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A method for estimation of the viscoelastic characteristics of polymer melts in the prestationary elongational flow is given. The experimental data show that at specific strain rates the polymer starts to respond to dynamic deformation as a highly elastic material. The viscoelastic characteristics of the polymer in the prestationary extensional flow can be described by a modified Maxwell equation.

In this work the viscoelastic properties of polymer melts in prestationary elongational flow are estimated after their reaction to periodic deformation. The periodic deformation with small amplitudes is applied together with uniaxial extension of a polymer sample at constant rate of deformation ($\dot{\varepsilon} = \text{const}$).

A special experimental device, a vibrorheometer, and an automatic control system herein[1] have been created. The block-diagram is given in Figure 1.

The elongational flow of sample 1 is carried out according to the scheme given by J. Meisner,[2] with two pairs of spaced gear rollers revolving in opposite directions. To eliminate the gravitational force and to keep constant the temperature of the sample the latter is submerged into a thermostatic bath 2. Gear rollers 3 and 4 are driven by motor 5 and reduction gears 6. The rollers of each pair pull the sample between them. The length of the sample tension zone l_0 is kept constant throughout the whole test. The distance between the pairs of rollers (grips) corresponding to the length of the sample tension zone is predetermined by the displacement of carriage 7 with grip 3. The kinematic scheme of the originated vibrorheometer allows the rate of

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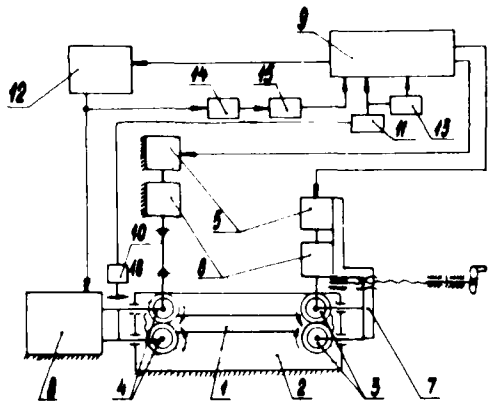

FIGURE 1 Block-diagram of vibrorheometer (E' and E'').

uniaxial deformation to change by factor of 10^5. The periodic strain of the sample is provided by electrodynamic oscillator 8 connected to grip 4.

The relation between the periodic constituents σ_{11g} of the total stress and periodic deformation ε_g is called a complex modulus. The components E' and E'' are calculated from the difference of the vibrorheometer mechanical impedances with the sample z_n and without the sample z_0. The equations for the calculation[1] of E' and E'' are as follows:

$$E' = (z_n \cos \delta_n - z_0 \cos \delta_0) l_0 / \pi r^2;$$
$$E'' = (z_n \sin \delta_n - z_0 \sin \delta_0) l_0 / \pi r^2,$$

where r is the value of the sample radius and δ is the phase angle between force and deformation. It is essential that in the course of the experiment the vibrorheometer mechanic impedance should be constant at the same frequency of the periodic deformation[3] and should be less than the sample impedance $z_0 < z$. The impedance z_0 in the range of subresonance frequencies depends mainly on the suspension rigidity. However, for the purpose of measuring the material resistance to the uniaxial extension at $\dot{\varepsilon} = \text{const}$, grips with constant and small rigidity are not suitable. The position of the grip depends on the stretching force which causes a nonlinear change of the mechanic impedance of the system. The sample suspension must be rigid, while the material is being stretched at constant strain rate, and must yield at superposition of the periodic elongational deformation. The above features of the grip are provided by motion control of the grip connected to the movable coil of the electrodynamic oscillator 8 employing computer 9. The grip rigidity is determined by the mechanic suspension rigidity and the effect of the electrodynamic force which is directly proportional to the current intensity in the movable coil of the oscillator. The control circuit comprises contactless displacement transducer 10, control unit 9, converters 11, 13, 14,

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15, amplifier 12 and electrodynamic oscillator 8. The stretching force is compensated by comparing the actual grip position x_i (displacement) with its predetermined constant position x_0. With the displacement present, the control unit acts on the electrodynamic oscillator until the stretching force effect P_{11} is eliminated. The control circuit includes an integrator and, therefore, the precision of the compensation system is independent of the force value. When the predetermined position x_0 of the grip is replaced by pulses of predetermined shape and frequency, the periodic oscillations are superimposed on the sample extension at constant rate of deformation.

The vibrorheometer has two operating modes. In the preparatory mode the mechanical impedance z_0 is being defined, while in the working mode two stages are distinguished: stage A during which $\dot{\varepsilon} = \text{const}$ and stage B where apart from uniaxial extension several periods of dynamic deformation are in progress. The least number of the periods of oscillations is limited by termination of the transient processes in the material caused by superposition of the oscillations. To determine the mechanic impedance of the vibrorheometer together with the sample z_n, a periodic force component must be singled out in stage B. The force component appearing in uniaxial extension is defined by interpolation of the stretching force dependence from adjacent stages A to the respective stage B. The periodic force component is a difference of the total force in stage B and the force determined by interpolation. The first harmonics of periodic force and deformation are derived from the last period of stage B with Fourier analysis and, subsequently, the components of the complex modulus are calculated.

The stress $\sigma_{11} = P_{11}/\pi r^2$ at $\dot{\varepsilon} = \text{const}$ (data of stage A) as a function of ε (elongational deformation according to Hencky) is calculated according to the method given in Ref. 2. In the case of noncompressibility of material the equation for the deformation rate and the actual sample radius takes the form:

$$\dot{\varepsilon} = \frac{V_1 + V_2 - (V_1/R_1 + V_2/R_2) r_0 e^{-\varepsilon/2}}{l_0 - \sqrt{r_0 e^{-\varepsilon/2}(2R_1 - r_0 e^{-\varepsilon/2})} - \sqrt{r_0 e^{-\varepsilon/2}(2R_2 - r_0 e^{-\varepsilon/2})}}; \quad r = r_0 e^{-\varepsilon/2},$$

where V_1 and V_2 are the tangential rate of the roller circle, while R_1 and R_2 are the roller radii and r_0 is the initial sample radius. To find the correspondence of the calculated radius to the actual one the latter is determined as follows: after the test is finished the extended sample is fixed by special clamps and is cut at both ends outside the clamping zone. The clamped sample is removed from the thermostatic bath and cooled, and the sample diameter is measured in different cross-sections.

The experiments were carried out at 19°C and were performed with polyisobutylene PIB-20. The material possessed the following characteristics: the zero-shear viscosity η_0 was 8×10^5 Pa × s, the characteristic relaxation

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time $\tau_r - 26.3$ s and the decrease rate coefficient of the shearing dynamic viscosity $(d\eta'/d\omega)\alpha - 0.83$. The values of η_0, τ_r and α were determined by approximation of the shearing dynamic viscosity[4] to the equation:

$$\eta' = \frac{\eta_0}{1+(\tau_r \omega)^\alpha},$$

where ω is the angular frequency of periodic deformation. Now the viscoelastic characteristics allow us to calculate the relaxation frequency spectrum of the material.

The experimentally determined values of the complex modulus components given in Figure 2 (a and b) show that E' and E'' of the superimposed

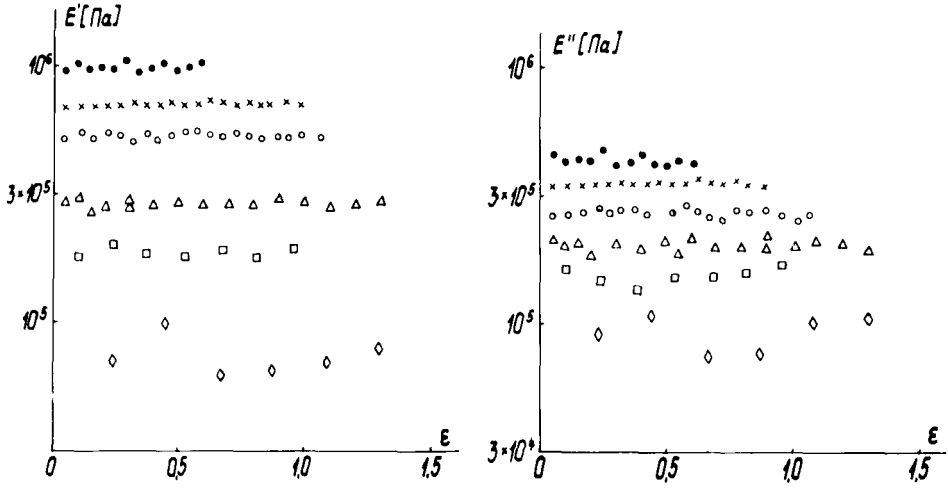

FIGURE 2a,b Dependence of the real complex modulus component on deformation at strain rate $\dot{\varepsilon} \leqslant 10^{-3}\,s^{-1}$. The frequency of periodic deformation $\omega\,s^{-1}$: ● = 31.4; × = 9.4; ○ = 3.14; △ = 0.628; □ = 0.314; ◇ = 0.0628.

periodic deformation are constant values independent of the deformation of uniaxial extension at all used angular frequencies ω. The values of E' and E'' determined during the test at various levels of uniaxial extension of the samples lead to the conclusion that deformation with $\dot{\varepsilon} = \text{const}$ in the Trauton flow region does not affect the viscoelastic characteristics.

Consequently, it enables us to correlate the dependence of E' and E'' on the angular frequency with the analogous dependence of the complex shearing modulus components G' and G''. As seen from Figure 3, the frequency dependences of E' and E'' measured by the uniaxial flow with $\dot{\varepsilon} \leqslant 10^{-3}\,s^{-1}$ correspond to triple values of the complex shearing modulus component determined by Weissenberg rheogoniometer. In this case the relations $E' = 3G'$ and $E'' = 3G''$ are valid both for the frequencies corresponding to the

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flow region ($tg\delta > 1$) and for higher frequencies at which the flowing polymer responds to the periodic deformation as a highly elastic material.

At shear deformation with frequency quantitatively equal to the shear rate the dynamic viscosity correlates with the effective viscosity of the polymer, since the mechanisms of shearing flow and periodic deformation[5] are alike. The experimental data of uniaxial extension and periodic deformation of polyisobutylene PIB-20 allow to apply such similarities also in uniaxial deformation. In the case of periodic deformation of PIB-20 and slow elongational flow, as given in Figure 3, regardless of the growth of E'' with the

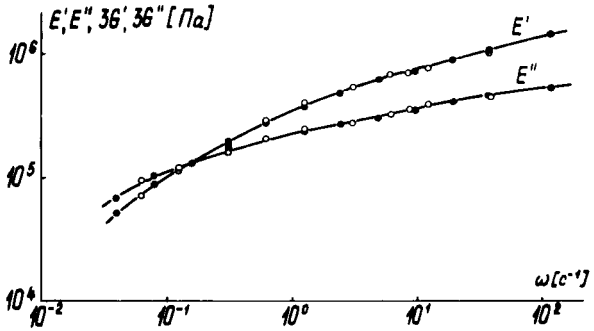

FIGURE 3 Frequency dependence of the complex modulus components ● = $3G'$, $3G''$; ○ = E', E''.

frequency increasing, the dynamic viscosity $\lambda' = E''/\omega$ is a falling function of the angular frequency ω. As seen from Figure 4, the correlation between elongational and triple shearing viscosity of PIB-20 occurs at small rates of

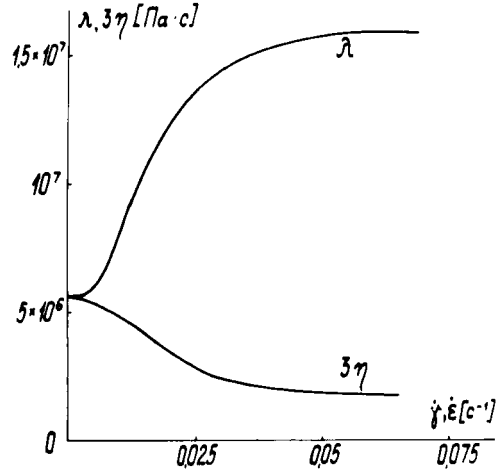

FIGURE 4 Elongational and triple shear viscosity correlation at various values of strain rate.

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deformation. Increasing the rate of deformation the elongational viscosity λ grows, while the shearing viscosity falls. Thus, in the case of shearing deformation $\eta(\dot{\gamma})$ and $\eta'(\omega)$ are of similar, and in the case of uniaxial deformation $\lambda(\dot{\varepsilon})$ and $\lambda'(\omega)$ are of opposite character. Therefore, in the case of longitudinal deformation there is no similarity between $\lambda(\dot{\varepsilon})$ and $\lambda'(\omega)$. Such phenomenon is likely to be attributed to the different polymer flow mechanisms in shearing and uniaxial flows.

The experimental data of superposition of periodic deformation on uniaxial extension of polyisobutylene PIB-20 at different rates are given in Figure 5. In the range of deformations from $\varepsilon = 0$ to $\varepsilon = 3$, E'' is independent of the growth of the elongational deformation. For the deformation rate

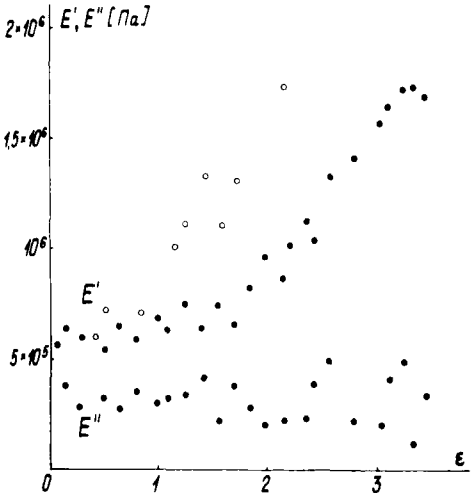

FIGURE 5 E' and E'' dependence on the extension deformation. The periodic deformation frequency $\omega = 3.14\ s^{-1}$. Uniaxial extension rate $\dot{\varepsilon}\ s^{-1}$: ● = 0.012; ○ = 0.136.

$\dot{\varepsilon} = 0.012\ s^{-1}$ and the angular frequency of periodic deformation $\omega = 3.14\ s^{-1}$ the mean statistic value of E''_m can be obtained which correlates with the value of E'' determined in uniaxial tension of PIB-20 at small strain rates in the Trauton flow region. The difference between the two values of the complex modulus is not greater than 7%. At deformation rates close to the inverse characteristic relaxation time $\dot{\varepsilon} \approx 1/\tau_r$, E' has a constant value in the domain of small values of tension deformation ($\varepsilon \to 0$), as seen from Figure 5. The initial value of E' equals to the triple value of the corresponding component of the complex shearing modulus. Therefore, at the initial stage of uniaxial extension at any given strain rate the sample possesses the viscoelastic characteristics of linearly deformed material.

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At a certain value of deformation, depending on the rate of $\dot{\varepsilon}$ a sharp increase in E' as the function of ε is observed. Now the following regularity is evident: with the increase in strain rates the limit of the constant values of E' tends towards small deformations (Figure 5).

The growth of the real component of complex modulus is observed on reaching the stretching strain which corresponds to a sharp increase of the tensile stress. In this case, according to Ref. 6, the loss of the flowing abilities of individual fractions of the polymer melts results in hardening of the material in the course of deformation.

The experimentally derived dependences of E' on ω in the periodic deformation frequency range $\omega = 0.125 \div 6.28\ s^{-1}$ at high deformation rate $\dot{\varepsilon} > 1/\tau_r$ are given in Figure 6.

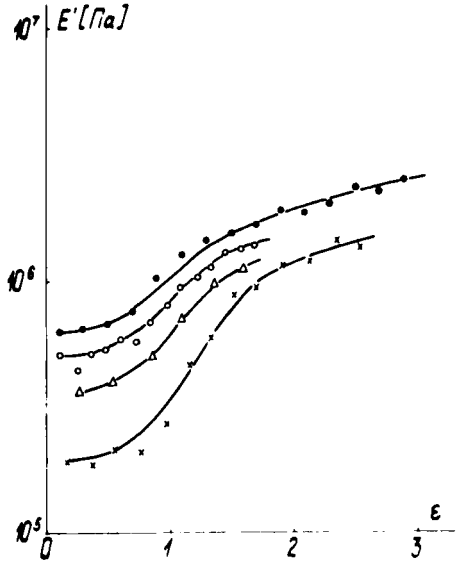

FIGURE 6 The dependence of the real complex modulus component on extension deformation at $\dot{\varepsilon} = 0.03\ s^{-1}$. The periodic deformation frequency $\omega\ s^{-1}$: ● = 6.28; ○ = 3.14; △ = 0.314; × = 0.125.

In the region of small values of elongational deformation, as mentioned above, the given dependences exhibit a domain of constant E' values. The further development of the sample tension involves an increase in the real component of complex modulus.

The frequency dependence of E' and E'' at given value $\varepsilon = \text{const}$ is plotted in Figure 7, which shows that starting from the specific deformation rates the polymer melt begins to respond to the superimposed periodic deformation like a highly elastic material. Then the flowing zone limit characterized by $tg\delta = E''/E' = 1$ shifts towards the small frequency domain.

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Figures 7–9 show a correlation between the experimentally determined values E' and E'' and those calculated after the Maxwell equation modified for the large deformations and applied to the continuous relaxation frequency

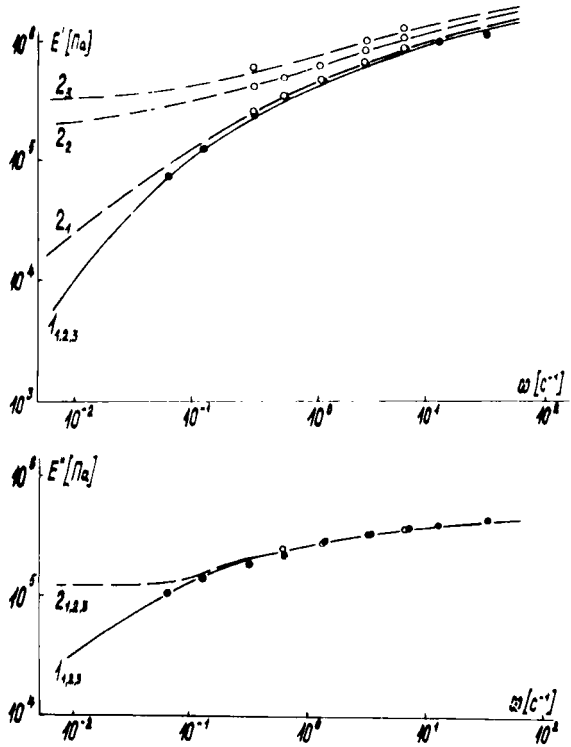

FIGURE 7 Frequency dependence E' and E'' at strain rates $\dot{\varepsilon}$ s^{-1}; $1-10^{-3}$; $2-3\times10^{-1}$. Extension deformations ε: 1_1 and $2_1-0.1$; 1_2 and $2_2-1.1$; 1_3 and $2_3-1.5$. The points on the curves are experimental data, the lines are calculated values.

spectrum. The complex modulus components[7] can be written as follows:

$$E' = \int_0^\infty \left[2\frac{\omega^2 - 2\dot{\varepsilon}(s-2\dot{\varepsilon})}{(s-2\dot{\varepsilon})^2+\omega^2} + \frac{\omega^2+\dot{\varepsilon}(s+\dot{\varepsilon})}{(s+\dot{\varepsilon})^2+\omega^2} + 4\dot{\varepsilon}\frac{1-e^{-(s-\dot{\varepsilon})t}}{s-2\dot{\varepsilon}} - \dot{\varepsilon}\frac{1-e^{-(s+\dot{\varepsilon})t}}{s+\dot{\varepsilon}} \right] N(s)\,ds;$$

$$E'' = \omega\int_0^\infty s\left[\frac{2}{(s-2\dot{\varepsilon})^2+\omega^2} + \frac{1}{(s+\dot{\varepsilon})^2+\omega^2}\right] N(s)\,ds,$$

where $N(s)$ is a relaxation frequency spectrum and s is a relaxation frequency. The calculated E' and E'' are in good agreement with the experimentally determined values of the prestationary extensional flow (Figure 8). At the uni-

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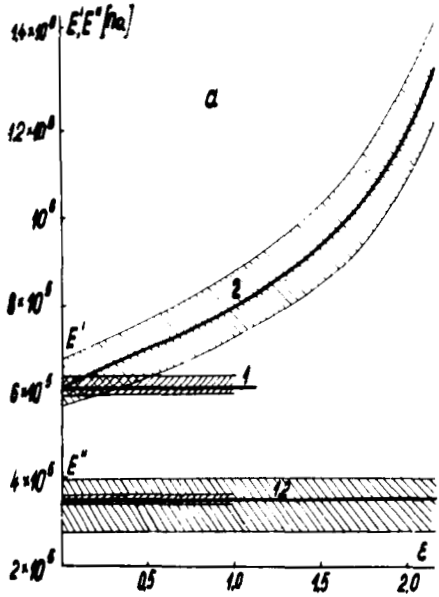

FIGURE 8 Correlation of the experimentally defined and calculated dependences of E' and E'' on deformation at $\omega = 3.14\ s^{-1}$. Strain rate $\dot{\varepsilon}\ s^{-1}$: $1 - 10^{-3}$; $2 - 3 \times 10^{-2}$.

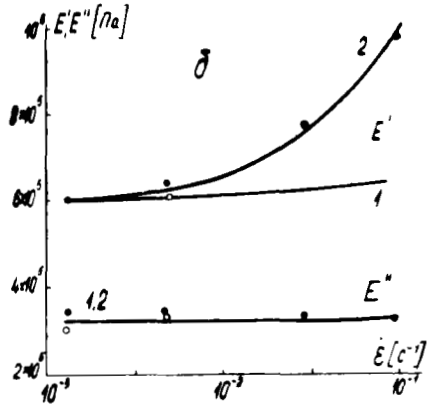

FIGURE 9 Dependence of E' and E'' on strain rate at various extension deformation values ε: $1 - 0.1$; $2 - 0.9$. The points on the curves are experimental data, the lines are calculated values.

axial extension rate $\dot{\varepsilon} = 10^{-3}\ s^{-1}$ (corresponding to the Trauton flow region) the experimental data for E' and E'' differ from the calculated values not more than 4%. At deformation rate $\dot{\varepsilon} = 0.03\ s^{-1}$ the experimental data for E'' differ from the calculated values not more than 19% in the whole range of elongational deformations, while the difference between the experimental

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data for E' in the domain of small deformations ε and the corresponding calculated values is not greater than 10%, and on reaching the deformation $\varepsilon \approx 2$ the difference is 18–20%. A correlation between the calculated and experimentally determined values of E' and E'', given in Figure 8, was based on the data of three independent tests for each deformation rate.

The components of complex modulus, as illustrated in Figure 9, are determined by the given deformation rate $\dot{\varepsilon}$ and the value of ε in the course of the prestationary extensional flow. Figure 9 shows a correlation between the calculated and experimental frequence dependences of E' and E'' on the deformation rate $\dot{\varepsilon}$ at the same values of ε. The complex modulus component E'' obtains a constant value and the calculated data are in good agreement with the experimental results. The deformation rate growth gives a slight increase in E' at $\varepsilon = 0.1$ and a rather sharp increase in E' at $\varepsilon = 0.9$. The correlation allows to consider the modified Maxwell equation to be suitable for the description of E' and E'' in the prestationary extensional flow of the polymer.

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