EFFECT OF SWELLING IN THE EXTRUSION OF AN ELASTIC LIQUID FROM A CAPILLARY

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The article demonstrates that the profile of a jet extruded from a capillary can be constructed from the data of uniform stretching.

Under examination is inertialess isothermal extrusion with constant force F_1 of a jet of polymer liquid (Fig. 1a) (along the z axis) from a circular capillary. The jet is extruded with mean velocity $v_1 = q/\pi r_1^2$. The flow in the capillary is induced by pressure P in the preentry zone of the capillary and by the extrusion force F_1. Often, the effect of F_1 on the flow in the capillary is small, and the flow is induced only by P. As a special case we examine deformation with $F_1 = 0$; in that case the jet emerging from the capillary (Fig. 2a) swells because of the accumulated elastic energy (the so-called Barus effect). When $F_1 \neq 0$, the jet, after initially swelling, reduces its diameter (Fig. 1a). The larger F_1 is, the smaller is the swelling of the jet emerging from the capillary. When F_1 is sufficiently large, the liquid may become detached from the capillary walls; this was experimentally observed, e.g., in [1].

In examining the extrusion flow from a capillary, we can distinguish approximately three regions (Fig. 1a; 2a): the region of steady-state flow inside the capillary I, the region of swelling of the liquid III, and the intermediate region between them, the small region II. We want to point out that the swelling in the zone adjacent to the z axis also penetrates into region II [2].

The theoretical solution of the problem of extrusion of a jet from a capillary in the exact statement is complicated. Such a problem can be solved approximately by somehow combining the solution of the problem of steady-state flow inside a capillary (region I, Figs. 1a; 2a) with the solution of the problem of stretching a jet with uniform cross section outside the capillary (region III, Figs. 1a; 2a).

Leonov and Prokunin [3] obtained a good description of shear and extension of liquid polymer in a wide range of deformation rates on a rheological model consisting of two parallel nonlinear Maxwellian elements.

In examining the problem on the basis of the model [3] for combining the above-mentioned solutions, we need two equations for determining the elastic deformations λ_1 and λ_2 in each Maxwellian element at the edge of the capillary (boundary conditions of the second problem). The first equation is written on the assumption that the flux of elastic energy changes little in the transition zone II. In this case, if we calculate the energy flux in shear and tension for the classical potential of the network theory of high elasticity, we obtain

$$2\pi \int_0^{r_1} \rho v_z \left(\sum_1^2 x_{zz}^{(i)} + x_{rr}^{(i)} - 2 \right) d\rho = q \sum_1^2 (\lambda_i^2 + 2\lambda_i^{-1} - 3). \tag{1}$$

Here, $x_{zz}^{(i)}$ and $x_{rr}^{(i)}$ are the normal components of elastic deformations [4] obtained from the solution of the problem of flow in a capillary.

Because of the assumption that extension (along the section of the specimen perpendicular to the z axis) to the right of the edge of the capillary is uniform, the equation for determining λ_1 and λ_2 is written as follows [5]:

$$\frac{F_1}{\pi r_1^2} = \sigma_1 = \sum_1^2 \mu_i (\lambda_i^2 - \lambda_i^{-1}). \tag{2}$$

For the case $F_1 = 0$ it is not unusual to examine the narrower problem of determining the swelling coefficient γ without determining the profile of the jet:

$$\gamma = \frac{r(\infty)}{r_1}; \quad r(\infty) = \lim_{z \to \infty} r(z). \tag{3}$$

A method (retardation regime) is shown in [3] by which γ is determined if λ_1 and λ_2 are known. We want to point out that for combining the solutions, equations other than (1) and (2) may also be taken.

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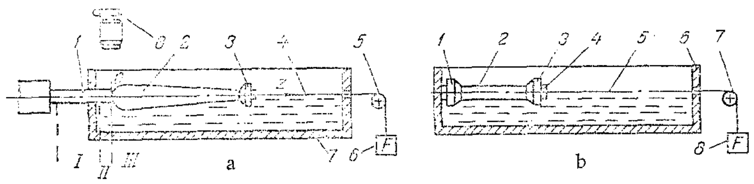

Fig. 1. Diagrams of the experimental setups: a) for extrusion of a jet from a capillary; b) for uniform stretching of a cylindrical specimen.

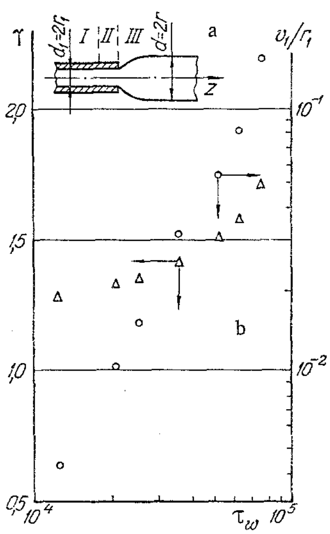

Fig. 2. Free extrusion from a capillary: a) swelling of a jet freely emerging from a capillary; b) experimental dependences of the ratio of the mean velocity of the jet emerging from the capillary to its radius v_1/r_1, $\sec^{-1}$ and of maximum swelling γ on the stress at the wall of the capillary τ_w, N/cm^2.

An attempt to combine two solutions of the inner and outer problem for determining γ was first made in [6, 7]. In [8] γ was determined from the condition of equality of the fluxes of elastic energy in shear and tension. The fluxes of elastic energy were calculated semiempirically for the steady-state flow in the capillary by the experimentally determined shear stress and elastic deformation. In calculating the energy flux in the region of emergence from the capillary it was assumed that here the polymer behaves purely elastically. The value γ thus calculated yielded a good description of the corresponding experiments.

In the present work we will model experimentally the free swelling by the process of uniform retardation, and extension of the jet emerging from the capillary by superimposing uniform extension on the retardation process.

We begin with the case $F_1 = 0$; the swelling process can be simulated by the process of uniform retardation. By retardation we mean the free shortening of the polymer specimen after uniform extension to the length l_H. During the retardation process the length of the specimen l is measured at time t for determining the deformation $\epsilon_1 = l/l_H$. At the instant of the beginning of retardation t = 0. This last premise is bound up with the assumption that the region of extension inside the capillary is small. Experiments show that for liquid polymers in the examined range of deformation rates, the retardation process is determined all in all by two parameters: elastic deformation α and deformation rate $\varkappa$ which the extended liquid had at the instant of beginning of retardation. The elastic deformation determined in the experiment is $\alpha = l_H/l_r\,(l_r = l(\infty))$ [9].

To construct the profile of the jet emerging from the capillary (the dependence of r on the longitudinal coordinate z) according to the data of uniform retardation, we use the correlation between z and t obtained in [10]:

$$z/v_1 = \int_0^t \varepsilon_1(t)\,dt;\ \ \varepsilon_1(t) = \frac{l(t)}{l_H} = \frac{r_1^2}{r^2(t)}\ . \tag{4}$$

Here, z is counted from the edge of the capillary (Fig. 2a). The dependences $\epsilon_1(\alpha, \varkappa, t)$ were selected by proceeding from the specified $\alpha = \gamma^2$ and the approximate fulfillment of the equality

$$L/v_1 = \int_0^{t_1} \varepsilon_1(t, \alpha, \varkappa)\,dt. \quad (5)$$

The dependences r(z) calculated from the experimental dependences $\epsilon_1(\alpha, \varkappa, t)$ and formulas (4) are compared below with the experimental profile of the jet obtained by photographing it as it emerges from the capillary.

We will now go over to the problem of a jet emerging from the capillary being stretched by the force F_1. With such deformation, the radius of the jet increases along z, and then it begins to decrease. We examine the case when there is no detachment of the liquid from the capillary walls (due to the force F_1) and the tensile stress $\sigma_1 = F_1/\pi r_1^2 \ll P$, i.e., the tensile force F_1 does not affect the flow rate q of the liquid. In that case we may use the previous method: to stretch in some way the cylindrical specimen to the specified α and $\varkappa$ (by which we simulate swelling), and then to measure $\varepsilon_1(t) = l(t)/l_H$ already at the specified load F_1. Then, from the dependence $\epsilon_1(\alpha, \varkappa, t)$ and formulas (3) and (4) we can calculate the profile r(z) with F_1 = const. We want to emphasize that to construct such a profile, we have to know γ and L/v_1 determined with $F_1 = 0$ for the specified pressure P, and the data for the corresponding uniform stretching under the effect of the force F_1. The profiles thus obtained are compared below with the experimental ones.

In the experiments we investigated liquid polyisobutylene P-20* (molecular weight $\sim 10^5$, density $9 \cdot 10^2$ kg/m³) with a viscosity of $\sim 10^6$ Pa·sec. All experiments were carried out at 22 ± 0.5°C.

The uniform stretching for simulating both the effect of deformation inside the capillary and of external stretching on the jet was carried out with constant force. A diagram of the setup is shown in Fig. 1b. The left-hand end of specimen 2 was fixed with the aid of bush 1 to the wall of bath 6 [11]. The right-hand end was fixed with the aid of the same kind of bush 3 to float 4 floating on the water. The water bath was intended for compensating the weight of the specimen and for thermostating. Attached to the float was thread 5 which ran over the pulley 7; suspended from the other end of the thread was weight 8.

The specimens for uniform stretching were made by spreading the polymer between two plates to the diameter d_0 = 0.6 to 0.7 cm, which was secured by spheres of specified size [12]. The length of the undeformed specimen l_0 was 3 to 4 cm.

To measure the dependence $\varepsilon_1(t) = l/l_H$ (4) after stretching the specimen to the length l_H by the weight F_0, part of the entire load was practically instantaneously removed, and the length of the specimen $l(t)$ was measured visually with a ruler placed alongside the bath. To determine the elastic deformation $\alpha = l_H/l_r$, the maximum length l_r ($l(t) \to l_r$ with $t \to \infty$) of the shortened specimen with completely removed load was measured. In practice, the time of shortening in measuring α was $\sim$30 min [9].

Extrusion of the jet emerging from the capillary and its free swelling were carried out on a setup whose diagram is shown in Fig. 1a. The jet of liquid 2 is extruded from the capillary 1 (length b = 10 cm, r_1 = 0.25 cm). When drawing was investigated, bush 3 was slipped over the jet emerging from the capillary (the pressure in the cylinder of the capillary viscometer was maintained); bush 3 was connected by thread 4 running over pulley 5 with weight 6. For thermostating and to eliminate the effect of the weight, the polymer was extruded into bath 7 filled with water. The number 8 denotes the photographic camera, which could be moved along the polymer jet 1. The profile of the specimen was photographed at the scale 1 : 1. A decoder ÉDI-452 was used for magnification when the jet profile was measured on the film. The length of the photographed profile depended on the time the movable end 3 required for passing through the entire bath. To specify the pressure, we used the equipment of a capillary constant-pressure viscometer designed by the special design bureau of the Institute of Petrochemical Synthesis, Academy of Sciences of the USSR.

To determine the maximum swelling $\gamma = r(\infty)/r_1$ upon free emergence of the jet from the capillary ($F_1 = 0$), we measured the diameter of the specimen at a place where it practically did not change along the coordinate z (the polymer was about 30 min outside the capillary at the place of measurement). The flow rate q, with pressure P specified, was determined by weighing cut-off pieces of the jet of polymer emerging from the capillary within a specified time.

Thus, we measured in the experiments with uniform stretching: $\varepsilon_1(t, \alpha, \varkappa)$, in extrusion from the capillary: r(z), γ(P), and q(P). The scatter of the entire set of experimental data did not exceed ±7%.

Figure 2 presents the dependences of maximum swelling γ and of the ratio of the mean velocity to the radius of the capillary $v_1/r_1 = q/\pi r_1^3$ on the stress at the capillary wall $\tau_w = Pr_1/2b$ (preliminary experiment). These dependences are invariant with respect to r_1 and b, and γ and v_1/r_1 increase monotonically with increasing τ_w.

*This polyisobutylene somewhat differs from the one investigated in [5].

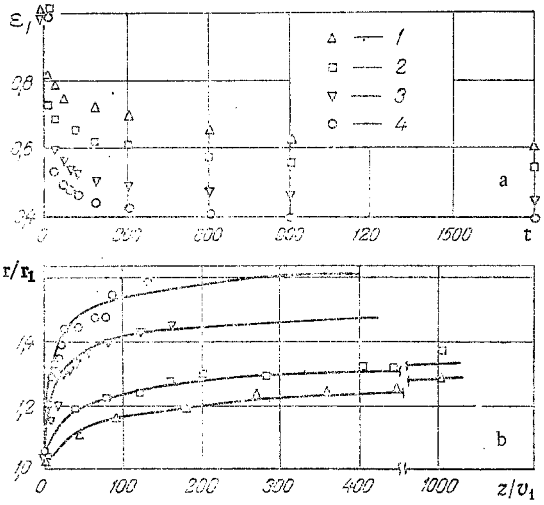

Fig. 3. Swelling of a jet of liquid polymer emerging from a capillary: a) dependence of ϵ_1 on t (sec) with different values of α: 1) α = 1.65; 2) 1.77; 3) 2.22; 4) 2.60; b) dependence of the relative jet radius r/r_1 on z/v_1, sec. Dots 1-4 correspond to the following values of $\gamma^2 = \alpha$: 1) 1.65; 2) 1.77; 3) 2.22; 4) 2.60. Lines represent the theoretical dependences obtained from experimental data of a and formula (4).

Extrusion of the jet from the capillary for measuring the profile r(z) was carried out by the pressure P. This pressure, which ensured the specified swelling γ and mean velocity v_1, was selected according to Fig. 2. The experimental stationary profiles obtained by photographing the jet freely emerging from the capillary for different τ_w are shown by dots in Fig. 3.

The dependences $\varepsilon_1(t) = l(t)/l_н$ for a freely shortening specimen after uniform stretching to deformation $\varepsilon_0 = l_н/l_0$, whose elastic part is $\alpha = \gamma^2$, are shown by dots in Fig. 3a. Relationship (5) also applies to the dependences ϵ_1(t). The solid lines in Fig. 3b represent the theoretical profiles of the swelling specimen, obtained from the experimental data in Fig. 3a and formulas (4) in which the value v_1 was determined from Fig. 2 by the specified τ_w and r_1.

In Fig. 4a, the dots represent the dependences ϵ_1(t) obtained in uniform stretching of specimens (preliminarily stretched to different deformations ϵ_0) by the constant force F_1. The values of ϵ_0 and of α corresponding to them are the same as above. In both cases stretching to deformation ϵ_0 was effected by the force F_0. It can be seen that when the force instantaneously decreases $(F_1 < F_0)$, the length of the specimen at first decreases, and then it increases. The larger α is, the longer it takes before the process of extension begins after F_0 has been replaced by F_1. We want to point out that variants are possible where with considerable decrease in the load ($\approx$10%), the recovery of the specimen amounts to less than 0.5% of the length (these data are not contained in Fig. 4), i.e., practically immediately after reduction of the force, extension begins.

The dots with index 4 in Fig. 4a show the curve obtained for fixed stress σ_1 without previous extension (α = 1 for t = 0). In Fig. 4b, the dots indicate the experimental dependences of r/r_1 on z/v_1 obtained on the basis of photographs of the polymer jet emerging from the capillary under the effect of pressure P and then of the constant tensile force F_1. Flow of this kind was steady. In Fig. 4b, $\sigma_1 = F_1/\pi r_1^2$ is fixed, and different is the deformation in the capillary preceding extension (different τ_w, γ, and v_1/r_1). The dependences r/r_1 on z pass through a maximum approximately at the same z/v_1. This maximum is the larger, the more intensive the deformation inside the capillary is (the larger τ_w is). We want to point out that with increasing τ_w, the maximum on the profiles r(z), with fixed σ_1, shifts toward larger z.

Extension of the jet emerging from the capillary within the limits of the error of measurement did not affect the flow rate. In extrusion from the capillary, the liquid may become detached from the wall, and this would be accompanied by a decrease in the diameter of the specimen inside the capillary. In the present experiments, no detachment was noticed because, as the photographs showed, the jet diameter with z = 0 was equal to the diameter of the capillary.

The curves in Fig. 4b represent the theoretical dependences obtained on the basis of the experimental data in Fig. 4a and formula (4). In formula (4) the value of v_1 was determined from Fig. 2 according to the specified τ_w and r_1. The dashed curve denotes the dependences calculated by ϵ_1(t) without previous extension (F_0 = 0, α = 1). The profile thus obtained corresponds to the extension of the polymer with constant force, where the effect of deformation in the capillary may be neglected. It can be seen from Fig. 4 (see the solid and dashed lines) that the effect of the deformation in the capillary on the profile of the jet may be considerable.

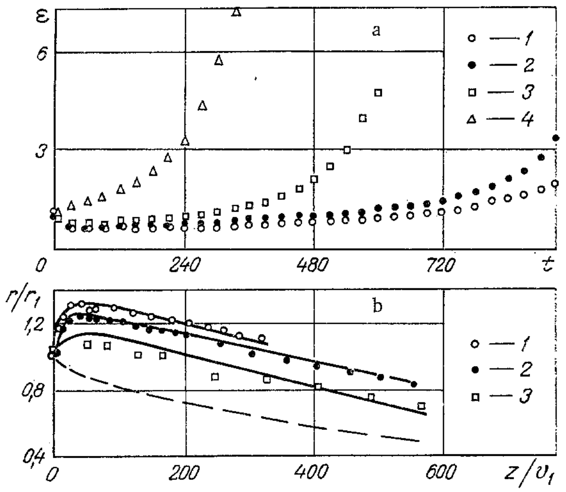

Fig. 4. Swelling of a polymer jet extruded by constant force upon emergence from a capillary: a) dependences of ϵ_1 on t, sec, for $\sigma_1 = 0.7 \cdot 10^4$ Pa [1) $\alpha = 2.6$; 2) 2.22; 3) 1.77; 4) 1.00]; b) dependences of r/r_1 on z/v_1, sec, for $\sigma_1 = 0.7 \cdot 10^4$ Pa. Dots 1, 2, 3 correspond to the square of maximum swelling $\gamma^2 = \alpha = 2.6, >2.22, 1.77$, respectively. Lines represent the theoretical dependences obtained from experimental data in a and formulas (3) and (4).

The dependences of r/r_1 on z/v_1, without the deformation in the capillary taken into account, do not depend on v_1 [see (4)]. When deformation in the capillary is taken into account, this does not apply, but the given representation of r/r_1 vs z/v_1 (Fig. 4b) makes the graphs more compact.

It follows from Figs. 3 and 4 that if the examined method is used, it is possible to describe quantitatively the profiles of the jet emerging from the capillary both with subsequent extension and without it, and there it is necessary to know the swelling coefficient of a freely emerging jet γ, the ratio L/v_1, and the data on uniform extension. The coincidence of the theoretical and experimental curves can be improved by a more accurate solution of Eq. (5).

The swelling during extrusion from a capillary indicates a process of desorientation (elastic deformation, which is a measure of the orientation, passes through its minimum). This conclusion follows directly from the fact that the specimen is shortened uniformly when the load is reduced (the region where $\epsilon_1 < 1$ in Fig. 4a). Upon shortening of the specimen, the elastic deformation α decreases with time.

The method of calculating the case when the jet extruded from a capillary is wound onto a drum is analogous to the one presented in [13].

The modeling explained above may be suitable in examining problems of the swelling of a polymer upon emerging from a capillary, both with subsequent extension and without it, even under anisothermal conditions.

NOTATION

v_1, mean velocity of the liquid in the capillary; r_1, radius of the capillary; q, flow rate; P, pressure in the preinlet zone of the capillary; F_1 and F_0, extrusion force; λ_1 and λ_2, elastic deformations in parallel Maxwellian elements upon stretching; ρ, z, radial and longitudinal coordinates, respectively; $v_z(\rho)$, velocity of steady-state flow in the capillary; μ_i, modulus of elasticity in the i-th Maxwellian element; γ, swelling coefficient; t, time; l, length of the specimen at instant t; α, elastic deformation; $\varkappa$, rate of deformation; ϵ_0, ϵ_1, deformation; L, length from which onward the radius of the jet extruded from the capillary practically does not change; r, radius of the jet; t_1, the time from which onward the radius of the specimen in retardation practically does not change; d_0, l_0, diameter and length, respectively, of the undeformed specimen; l_r, length to which the extended specimen tends after removal of the load; b, length of the capillary; σ_1, tensile stress (for uniform stretching at t = 0, for extrusion from the capillary at z = 0); l_H, length of the stretched specimen at the instant of beginning reading of t.

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