STRETCHING OF A PLANE ANOMALOUSLY VISCOUS FILM UNDER NONISOTHERMAL CONDITIONS

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A solution is obtained for the problem of the stretching of an anomalously viscous film under nonisothermal conditions. The solution is analyzed.

One of the principal processing operations in making thin films of thermoplastic resins is stretching. Stretching is generally done under nonisothermal conditions (particularly during forming) [1]. Also, the nonisothermality may significantly affect the accuracy of measurements of the rheological properties of the polymers in tension.

A detailed analysis was made in [2] of the isothermal stretching of a flat film. Thus, we have omitted questions related to the flow dynamics from the formulative part of the present work. For the force balance equations obtained in [2] to be applicable to nonisothermal stretching, they must be supplemented by a thermal balance equation, and viscosity must accounted for in the temperature functions.

Nonisothermal Stretching of Anomalously Viscous Film. A diagram of the stretching process is shown in Fig. 1. The following assumptions are made: 1) the thickness of the film is sufficiently small so that the nonuniformity of the temperature and velocity profiles in the transverse direction can be ignored; 2) the forces of surface tension, inertia, and friction of the film in air can be ignored in view of their smallness compared to the stress acting on the material in the lengthwise direction; 3) the heat of crystallization and dissipative heat liberation can be ignored; 4) the film is cooled mainly as a result of radiation and convection; 5) convective heat transfer is considerably greater in the longitudinal direction $\partial T/\partial x$ than in the transverse direction $\partial T/\partial b$; 6) the thermophysical properties of the material are constant; 7) the flat film is subjected to uniaxial tension (stretching); 8) the heat-transfer coefficient is constant along the film.

The heat-balance equation for an element of the film $b\delta dx$ has the form

$$\rho C \frac{Q}{b} \frac{dT}{dx} = q. \tag{1}$$

The quantity q is the sum of the convective and radiant heat fluxes from both sides of the film

$$q = -2\alpha_c (T - T_c) - 2\sigma^* \varepsilon (T^4 - T_c^4),$$

where $\alpha_c = (\alpha_1 + \alpha_2)/2$. We will also assume that cooling of the film occurs mainly in a sufficiently narrow temperature interval and that Newton's law can be applied to the radiation process [3]. Here, for q we may write

$$q = -2\alpha^* (T - T_c), \tag{2}$$

where $\alpha^* = \alpha_c + \varepsilon\sigma^* \varphi(T)$; $\varphi(T)$ is a coefficient [3].

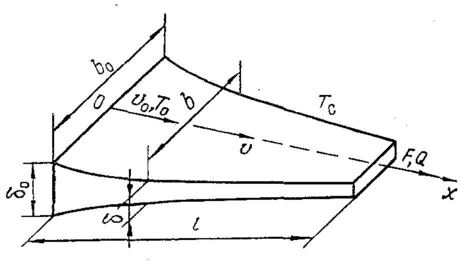

Fig. 1. Diagram of process of stretching a plane film.

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We will use the following semiempirical expression for the material function [4]:

$$\eta = \eta_0 \exp\left[\frac{E}{R}\left(\frac{1}{T} - \frac{1}{T_0}\right)\right]\left(\frac{I_2}{2}\right)^{\frac{n-1}{2}} \tag{3}$$

The equality $\sigma_{33} = 0$ is valid for the stress components across the film width under conditions of uniaxial tension [2]. Expressing σ_{33} through the strain rate, we obtain $d_{33} - d_{22} = 0$, or

$$b'/b = \delta'/\delta. \tag{4}$$

Integrating (4) with allowance for $x = 0$, $\delta = \delta_0$, and $b = b_0$, we obtain the condition of geometric similitude of the cross sections $b/b_0 = \delta/\delta_0$.

In accordance with [2], the expression for the second invariant of the strain-rate tensor has the form

$$I_2/2 = d_{11}^2 + d_{22}^2 + d_{33}^2,$$

where $d_{11} = -\dfrac{Q}{b\delta}\left(\dfrac{b'}{b} + \dfrac{\delta'}{\delta}\right)$; $d_{22} = \dfrac{Q\delta'}{b\delta^2}$; $d_{33} = \dfrac{Qb'}{b^2\delta}$.

We introduce the dimensionless parameters:

$$\bar{b} = \frac{b}{b_0};\ \bar{\delta} = \frac{\delta}{\delta_0};\ X = \frac{x}{l};\ \theta = \frac{T - T_c}{T_0 - T_c};$$

$$V = \frac{v}{v_0};\ Z(\theta) = \exp\left\{\frac{E}{R}\left[\frac{1}{T_0} - \frac{1}{\theta(T_0 - T_c) + T_c}\right]\right\}, \tag{5}$$

where $v_0 = Q/b_0\delta_0$ is the axial velocity in the initial section of the film. The quantity $Z(\theta)$ was introduced for brevity in writing the exponential multiplier in Eq. (3).

Allowing for condition (4) (and its integral result), we may represent the expression for the second invariant in (3) in the form of the following dependences on the kinematic parameters:

$$\left(\frac{I_2}{2}\right)^{\frac{1}{2}} = \frac{\sqrt{6}\, v_0 \bar{b}'}{l\bar{b}^3} = \frac{\sqrt{1.5}\, v_0 V'}{l}. \tag{6}$$

The system of equations describing the process of nonisothermal stretching of a flat film, with allowance for (1)-(6) and the force balance [2], has the form

$$\theta' = -2\,\mathrm{Mi}\,\bar{b}\theta, \tag{7}$$

$$\bar{b}' = -P\bar{b}Z(\theta)\left(\frac{\sqrt{6}\, v_0 \bar{b}'}{l\bar{b}^3}\right)^{n-1}, \tag{8}$$

$$V' = 2PVZ(\theta)\left(\frac{\sqrt{1.5}\, v_0 V'}{l}\right)^{n-1}, \tag{9}$$

where $\mathrm{Mi} = \alpha^* l b_0/\rho C Q$; $P = Fl/3\eta_0 Q$. The superimposed lines in (6)-(9) denote derivatives with respect to X. It follows from (4) that the thickness distribution in the stretching zone is similar to the width distribution ($\bar{\delta} = \bar{b}$). Thus, the equation for $\bar{\delta}$ has been omitted from system (7)-(9).

The initial condition for system (7)-(9) has the form:

$$\text{with } X = 0,\ \theta = \bar{b} = \bar{\delta} = V = 1. \tag{10}$$

We can obtain the following expression for the film width from Eq. (7):

$$\bar{b} = -\frac{\theta'}{2\,\mathrm{Mi}\theta},\ \bar{b}' = \frac{(\theta')^2 - \theta\theta''}{2\,\mathrm{Mi}\theta^2}.$$

Substituting the resulting expressions in (8) will give us an equation describing the temperature distribution along the stretching zone

$$\frac{\theta''}{\theta}-\left(\frac{\theta'}{\theta}\right)^2+\beta Z^{\frac{1}{n}}(\theta)\left(\frac{\theta'}{\theta}\right)^{\frac{3n-2}{n}}=0,$$

where $\beta=P^{\frac{1}{n}}(2\mathrm{Mi})^{\frac{2(1-n)}{n}}\left(\frac{\sqrt{6}\,v_0}{l}\right)^{\frac{1-n}{n}}$. Assuming that

$$Y=d\theta/dX, \tag{11}$$

we obtain Bernoulli's differential equation

$$\frac{dY}{d\theta}-\frac{Y}{\theta}+\beta Z^{\frac{1}{n}}(\theta)\left(\frac{Y}{\theta}\right)^{\frac{2(n-1)}{n}}=0,$$

which, through the substitution $U=Y^{\frac{2-n}{n}}$ [5], is changed into the linear equation

$$\frac{dU}{d\theta}-\left(\frac{2-n}{n}\right)\frac{U}{\theta}+\left(\frac{2-n}{n}\right)\beta Z^{\frac{1}{n}}(\theta)\,\theta^{\frac{2(1-n)}{n}}=0. \tag{12}$$

Allowing for (10), we obtain the following condition for Y from (7): with $\theta=1$, $Y=\theta'=-2\mathrm{Mi}$. Accordingly, for U we have the initial condition:

$$\text{with } \theta=1,\quad U=-(2\mathrm{Mi})^{\frac{2-n}{n}}. \tag{13}$$

Solving Eq. (12) with allowance for (13) and changing to the function Y, we have

$$Y(\zeta)=-\zeta\left[(2\,\mathrm{Mi})^{\frac{2-n}{n}}+\beta\left(\frac{2-n}{n}\right)J(\zeta)\right]^{\frac{n}{2-n}}\quad(n\neq 2), \tag{14}$$

where $J(\zeta)=\int\limits_1^{\zeta}\frac{Z^{\frac{1}{n}}(\xi)}{\xi}\,d\xi$. The variables ξ and ζ correspond to the dimensionless temperature and are introduced to explain the order of integration. With $n=2$, Eq. (14) is indeterminate. We find the function $Y(\zeta)$ for the case $n=2$ by direct integration of the above Bernoulli's differential equation, obtaining the following

$$Y(\zeta)=-2\,\mathrm{Mi}\,\zeta\exp\left[-\beta\int\limits_1^{\zeta}\frac{Z^{\frac{1}{2}}(\xi)}{\xi}\,d\xi\right].$$

In accordance with (11), the temperature distribution is determined by the expression

$$X=\int\limits_1^{\theta}\frac{1}{Y(\zeta)}\,d\zeta. \tag{15}$$

Substituting the expression for dX from (11) into (8) and integrating with allowance for (10) yields the following expression for the film width

$$\frac{n}{2(1-n)}\left[\bar{b}^{\frac{2(1-n)}{n}}-1\right]=-P^{\frac{1}{n}}\left(\frac{\sqrt{6}\,v_0}{l}\right)^{\frac{1-n}{n}}\int\limits_1^{\theta}\frac{Z^{\frac{1}{n}}(\zeta)}{Y(\zeta)}\,d\zeta\ (n\neq 1). \tag{16}$$

The velocity can be determined using the relation $\bar{b}=1/\sqrt{V}$, which was obtained from the continuity equation $\bar{b}\bar{\delta}V=1$ with allowance for (4).

Analysis of the models obtained requires a substantial amount of illustrative material, so we will examine special cases which are also of independent interest.

Nonisothermal Stretching of a Viscous Film. The solutions obtained are simplified somewhat for the case of a viscous fluid ($n=1$). We have the following expressions in this case: for temperature

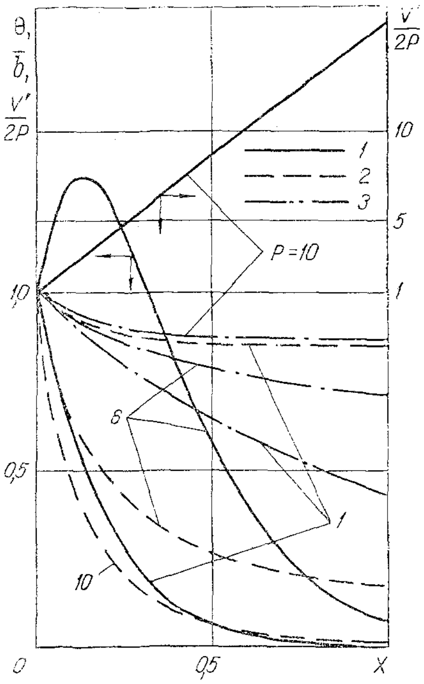

Fig. 2

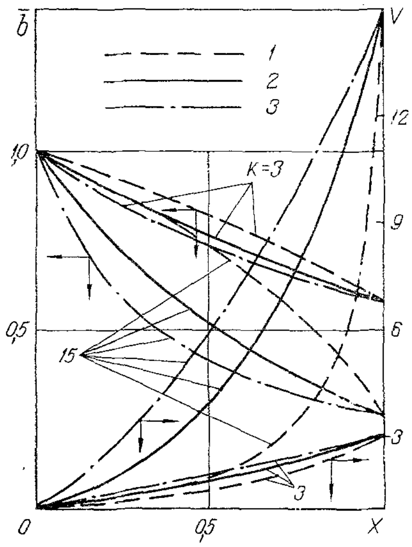

Fig. 3

Fig. 2. Distribution of dimensionless normalized gradient of axial velocity $V'/2P$ (1), width $\bar{b}$ (2), and dimensionless temperature θ (3) along the stretching zone with different stretching forces P and a constant Mikheev criterion (Mi = 0.5).

Fig. 3. Distribution of dimensionless width $\bar{b}$ and velocity V of a plane film in the stretching zone with different stretching ratios and flow indices: 1) n = 0.5; 2) 1.0; 3) 2.0.

$$X = \int_1^{\theta} \frac{1}{Y(\zeta)}\, d\zeta, \tag{17}$$

for film thickness and width

$$\ln \bar{\delta} = \ln \bar{b} = -P \int_1^{\theta} \frac{Z(\zeta)}{Y(\zeta)}\, d\zeta, \tag{18}$$

for velocity

$$\ln V = 2P \int_1^{\theta} \frac{Z(\zeta)}{Y(\zeta)}\, d\zeta,$$

for axial velocity gradient

$$V' = \frac{dV}{dX} = 2PZ(\theta)\exp\left[2P \int_1^{\theta} \frac{Z(\zeta)}{Y(\zeta)}\, d\zeta\right], \tag{19}$$

where $Y(\zeta) = -\zeta[2\,\mathrm{Mi} + PJ(\zeta)]$; $J(\zeta) = \int_1^{\zeta} \frac{Z(\xi)}{\xi}\, d\xi$.

Results of analysis of Eqs. (17)-(19) are shown in Fig. 2. The calculations were performed for $T_0 = 523°K$, $T_c = 293°K$, and E = 64.7 kJ/mole, which corresponds to the operation of forming a polypropylene film [6]. It follows from Fig. 2 that flow of and heat exchange with the film (with n = 1) is determined by two parameters: the Mikheev criterion Mi and the dimensionless stretching force P. Three regimes may be distinguished from the character of the axial velocity gradient (see Fig. 2).

1. The regime of "quasiisothermal" stretching. Values of P are relatively large and values of Mi relatively small. The axial velocity gradient increases monotonically and the condition $V' \geq 2P$ is satisfied (the curves corresponding to $P = 10$ in Fig. 2).

2. Intermediate regime. It is characterized by moderate values of P and Mi. The gradient of axial velocity has an extremum (maximum) (the curves corresponding to $P = 6$ in Fig. 2).

3) The polymer "setting" regime. It is characterized by high values of Mi and low values of P. The axial velocity gradient decreases monotonically and the condition $V' \leq 2P$ is satisfied (curves corresponding to $P = 1$ in Fig. 2).

In accordance with (6), the distribution of $\sqrt{I_2/2}$ along the stretching zone is similar to the distribution of V'.

The width (thickness) of the film decreases with an increase in the stretching force P, but the degree of film cooling decreases also, since it is in the stretching zone for a shorter period of time (Fig. 2).

The analysis shows that $Mi/P = \text{const}$ for a constant withdrawal rate and negligible change in P and Mi. In turn, this equation yields the relation $F \approx \alpha^* \eta_0$, which can be used to design automatic systems to regulate the stretching of plane films under nonisothermal conditions.

A check showed that Eq. (17) satisfactorily correlates with the experimental results in [1, 7].

Nonisothermal Stretching of an Anomalously Viscous Film. In the case $T = \text{const}$, the solution of Eq. (8) has the following form with allowance for (10)

$$\bar{b}^{\frac{2(1-n)}{n}} = 1 - \frac{2(1-n)}{n} P^{\frac{1}{n}} \left(\frac{\sqrt{6}\, v_0}{l} \right)^{\frac{1-n}{n}} X \quad (n \neq 1). \tag{20}$$

Using the withdrawal condition $X = 1$, $\bar{b} = 1/\sqrt{K}$, we can exclude P from (20). Here, we have for the film profile

$$\bar{\delta} = \bar{b} = [1 - (1 - K^{\frac{n-1}{n}}) X]^{\frac{n}{2(1-n)}} \quad (n \neq 1), \tag{21}$$

where $K = v_1/v_0$. The velocity distribution is determined from the relation $V = 1/\bar{b}^2$.

In stretching a film made of a material characterized by Newtonian properties ($n = 1$), the profile is described by the expression [2] $\bar{\delta} = \bar{b} = \exp(-0.5 X \ln K)$.

The expression for the dimensionless axial-velocity gradient in the case of an anomalously viscous fluid has the form

$$\frac{dV}{dX} = \frac{n}{n-1} [1 - (1 - K^{\frac{n-1}{n}}) X]^{\frac{1}{n-1}} (K^{\frac{n-1}{n}} - 1) \quad (n \neq 1).$$

Accordingly, for a Newtonian fluid, $V' = K^X \ln K$.

Figure 3 shows film velocity and width profiles with different stretching ratios and flow indices. The data were calculated with Eq. (21). It is apparent from the figure that the flow index has a significant effect on the form of the stream. Meanwhile, an increase in stretching ratio is accompanied by stronger manifestation of the anomalously viscous properties of the material. Thus, Eqs. (20) and (21) can be used to evaluate the rheological characteristics n and η_0. To determine n, it is sufficient to measure the initial and final width of the film, as well as its width in the middle part of the stream ($X = 0.5$). The stretching ratio may be defined as $K = (b_0/b_1)^2$.

It should be noted that, under stretching conditions, the flow properties of a polymeric material may differ significantly from the properties measured under conditions of shear strain [4].

It follows from Figs. 2 and 3 that an increase in parameter n is qualitatively equivalent to an increase in the Mikheev criterion or a decrease in the dimensionless stretching force under nonisothermal conditions.

General Remarks. The Mikheev criterion can be represented as the product of the Fourier and Biot numbers. It follows from [3] that the problem may be regarded as an external problem when $Bi \leq 0.1$. Thus, the solution obtained is acceptable if $\alpha^* \delta_0 / 2\lambda \leq 0.1$.

System (7)-(10) can be solved in finite differences (by the Runge-Kutta method) and in a more complete formulation (with allowance for the dependence of the thermophysical properties on temperature, dissipative heat release, and radiation in accordance with the Stefan—Boltzmann law, etc.).

To calculate the internal integral J in (14)-(19), it is convenient to use Gauss' formula for quadratures of the highest algebraic degree of accuracy [8]

$$\int_1^{\zeta} f(\xi)\, d\xi \approx (\zeta - 1)\left[0.25 f(1) + 0.75 f\left(\frac{1+2\zeta}{3}\right)\right].$$

In [2] was proposed a characteristic for the "widening" effect of the receiving beam K_1, representing the ratio of the coefficients of the second and first differences of the normal stresses. In the general case, K_1 depends not only on the dimensions of the stretching zone (l/b_0), but also on the Mikheev criterion. For example, in the "setting" regime (see above), there is always uniaxial tension and $K_1 = 0$. Thus, the method proposed in [2] for accounting for the "widening" effect of the receiving beam may be acceptable for stretching regimes which are close to isothermal.

The structure of the resulting film is determined by the nature of the polymer and its temperature-strain history in the manufacturing process. An important factor here is the time the polymer spends in the stretching zone. Under nonisothermal stretching conditions, it is determined from the relation

$$d\tau = \frac{l}{v_0} \frac{dX}{V},$$

or, allowing for (11),

$$\tau = \frac{l}{v_0} \int_1^{\theta} \frac{d\zeta}{Y(\zeta) V}.$$

NOTATION

b_0, δ_0, v_0, initial width, thickness, and velocity of film; l, length of stretching zone; b, δ, v, running width, thickness, and velocity of film; $\bar{b}$, $\bar{\delta}$, V, dimensionless width, thickness, and velocity of film; x, X, longitudinal dimensional and dimensionless coordinates; T_0, T_c, initial temperature of film and ambient temperature; T, running temperature of film; η_0, n, rheological constants; E, activation energy; R, universal gas constant; θ, dimensional temperature; I_2, second invariant of strain-rate tensor; Q, volumetric flow rate; ρ, C, λ, density, heat capacity, and thermal conductivity of polymer; α_c, arithmetic-mean heat-transfer coefficient due to natural convection; α_1, α_2, convective heat-transfer coefficients for transfer from free surfaces of film; α^*, heat-transfer coefficient accounting for convection and radiation; η, viscosity; σ_{33}, component of normal stress across film width; d_{11}, d_{22}, d_{33}, diagonal components of strain-rate tensor; Mi, Mikheev's criterion; P, dimensionless stretching force; F, tensile force; β, parameter; Y, U, auxiliary functions; ζ, ξ, auxiliary variables; J, integral, a function of temperature; K, stretching ratio; v_1, withdrawal speed; K_1, ratio of coefficients of second and first differences in normal stresses; f, arbitrary smooth function; ε, emissivity; σ^*, Stefan—Boltzmann constant; Bi, Biot number; b_1, film width at end of stretching zone; $\varphi(T)$, coefficient dependent on film temperature and ambient temperature; q, total of convective and radiant heat fluxes from both sides of film; τ, time.

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NONISOTHERMAL RETARDATION OF ELASTIC FLUIDS

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It is shown that nonisothermy during the retardation of a polymer fluid after extension can result in diminution in specimen reduction.

The nonisothermal effect under consideration is characteristic for elastic fluids. After homogeneous isothermal extension [1] of the cylindrical specimen of elastic fluid to a length l and diameter d ($l \gg d$), the stress can drop instantaneously to zero and afford the possibility of reducing the specimen in time because of the elastic energy accumulated during extension (retardation process). There is no stress in the inertialess approximation for this process. The time of specimen reduction is $t_1 \sim \theta_2$. Here θ_2 is the retardation time, which is a constant of the fluid. As the temperature changes, $\theta_2 \approx \exp(E/RT)$ for an elastic polymeric fluid. Under isothermal conditions, when the temperature during retardation is identical as during extension, the retardation process will be homogeneous. If the temperature of the environment changes by a jump,* say, after extension, then the retardation process will not be homogeneous if the specimen heating time is $t_2 \sim d^2/\xi \sim \theta_2$. In this case, tangential stresses, say, will occur during retardation because of the variable temperature along the specimen radius and the dependence $\theta_2(T)$. Hence, part of the elastic energy will be expended in relaxing the nonzero stresses, and specimen reduction will consequently diminish as compared to the isothermal case. A theoretical consideration of the problem of nonisothermal retardation because of its inhomogeneity is complex even in linear rheology. The effect discussed above is found experimentally in this paper.

The experiment was performed on a polyisobutylene P-20 melt with the greatest Newtonian viscosity $\eta = 1.3 \cdot 10^6$ Pa·sec, relaxation time $\theta_1 \sim 4 \cdot 10^2$ sec, and retardation time $\theta_2 \approx 80$ sec. This polymer had been investigated earlier under homogeneous extension and retardation in [2, 3]. Preliminary extension of the cylindrical specimen was carried out in the constant strain rate mode at T = 22 and 70°C. The extension and retardation were performed in a water bath to compensate for the specimen weight and thermostatting. To accomplish the retardation the extended specimen was cut by knives. The retardation process was performed at both the extension temperatures (isothermal case) and at 22°C after extension at 70°C (nonisothermal case). In the latter case, not more than 5 sec elapsed in the last case in the cutting and transferring of the specimen from one bath (70°C) to the other (22°C). The specimen diameter changed from ≈ 1.5 to ≈ 3 mm in the retardation. The time for the temperature change after the changeover was $t_2 \sim d^2/\xi \sim 10^2$ sec. The change in specimen length l_r with time was observed visually on a ruler during retardation.

The dependences of l_r/l on the time t obtained during retardation in the isothermal case for 22 and 70°C are presented in Fig. 1 (points 1, 2, respectively), and for specimen transferral from 70 to 22°C in the nonisothermal case. In all three cases the specimen was

*At a lower temperature the specimen is in the running state.

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