MATHEMATICAL DESCRIPTION OF THE RHEODYNAMICS AND HEAT TRANSFER ASSOCIATED WITH THE BLOWN-FILM PROCESS

A. V. Baranov, N. V. Tyabin,
V. A. Gerasimenko, and L. G. Raiz

UDC 678:532.135

A mathematical model is presented for nonuniform biaxial stretching of a nonlinear viscoelastic crystallizing medium under nonisothermal conditions.

The production of thin thermoplastic films by the blown-film (inflated-tube) extrusion process has enjoyed wide applications in recent years for polymer processing. This method can be used to obtain thinner biaxially oriented films than is possible in flat (sheet) extrusion, and they have higher tensile strength, frost resistance, tear strength, and transparency, making them useful not only for packaging, but also for electrical engineering, instrument design, medicine, furniture-making, etc. The tubular bubble that emerges from the die is drawn upward by the drawing mechanism, and air is injected into the die, inflating the bubble. External blowing is used to cool the hot bubble at a set height. The axial force F required on the rollers in order to draw the film upward is regulated, along with the pressure p inside the bubble.

Several serious attempts have been made previously to describe the blown-film extrusion process. The most complete coverage of this effort is found in the work of Han and Park [1, 2] on the nonisothermal stretching of an inelastic fluid with power-law rheology and an exponential temperature dependence of the viscosity in the entire inflation zone up to the solidification line. Below the melting point, however, the same degree of crystallinity corresponds to different temperatures and vice versa under nonisothermal crystallization conditions at different cooling rates. The structural state of the polymer is therefore determined not only by the temperature, but also by the degree of crystallinity. Consequently, even for approximate solutions to characterize the physical nature of the process correctly it is necessary to allow for the dependence of the rheological and thermophysical properties of the polymer both on the temperature and on the crystallinity. The density and specific heat are knwn not to depend as strongly on the temperature as, e.g., the viscosity. However, when the polymer cools down by 100-200°C, going through a phase transition in the process, the variation of these physical properties can be significant, and the calculations must reflect this fact.

Many researchers have noted [1, 3] that effects associated with a rubber-elastic melt must be taken into account in the theoretical analysis of the production of blown films. Such effects are even more essential than the possible dependence of the longitudinal viscosity on the shear rate [3]. This consideration is particularly timely for the production of oriented thermoplastic films by extrusion below the melting point [5]. However, only Petrie [4] has attempted to describe the isothermal blown-film process for a viscoelastic medium on the basis of the Maxwell model.

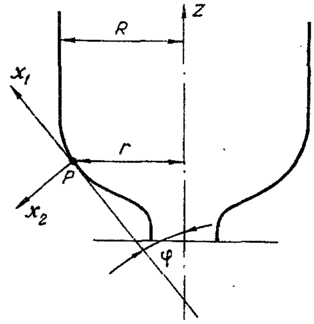

Fig. 1. Schematic view of the formation of a blown film.

Polytechnic Institute, Volgograd. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 54, No. 6, pp. 956-962, June, 1988. Original article submitted March 5, 1987.

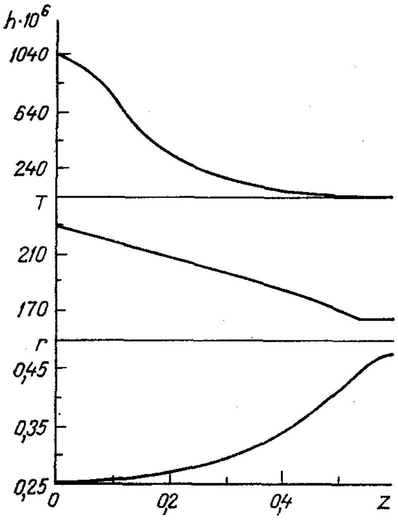

Fig. 2

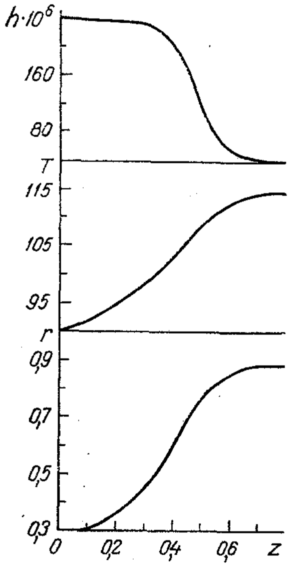

Fig. 3

Fig. 2. Distributions of the tube radius r (m) and the film thickness h (m) and temperature T (°C) along the height z (m) in formation from the melt; $\beta_1 = 1.5$ J/kg K^2; $\beta_2 = 6.0$ J/kg K^2; $\gamma_1 = 0.5$ kg/m^2 K; $\gamma_2 = 0.8$ kg/m^3 K; m = 3; $\eta_{02} = -8000$ Pa·sec^2; $\eta_{03} = 0$.

Fig. 3. Oriented film; distributions of the tube radius r and the film thickness h and temperature T along the height z.

In the present article we investigate the nonisothermal nonuniform biaxial stretching of a nonlinearly viscoelastic film in the zone where the tube radius r and the film thickness vary in the direction of motion of the film z. Since $h \ll r$, we use the approximation of the theory of thin shells. The process of formation of the blown film is shown schematically in Fig. 1. The rectangular coordinates at a certain point P on the surface of the film are related to the cylindrical coordinates in order to facilitate the solution. In this case x_1 corresponds to the direction of flow, x_2 is directed along the normal to the film, and the x_3 axis is perpendicular to the plane of the figure. The following assumptions are made in the formulation and solution of the problem: The film thickness is small enough that the nonuniformity of the velocity profile in the transverse direction can be ignored; the forces of surface tension, inertia, and air friction of the film tube are disregarded; heat transfer does not take place between the inner surface of the tube and the air contained in it; heat conduction in the thin film itself can be neglected (the Biot number is of the order of 10^{-1} to 10^{-3}).

We assume that the rheological properties of the polymer are described by a differential model, according to which the stress tensor is determined by the kinematic tensors of strain rates B_1 and strain accelerations B_2 [6, 7]:

$$\hat{\sigma} = -p\hat{\delta} + \eta_1 B_1 + \eta_2 B_2 + \eta_3 B_1^2. \tag{1}$$

The use of higher-order kinematic tensors does not have any practical value, since this would require knowledge of more initial conditions than can be established on the basis of physical considerations.

The strain-rate tensor for the given situation is known to have the form [1]

$$B_1 = 2\begin{bmatrix} -\left(\frac{1}{r}\frac{dr}{dt} + \frac{1}{h}\frac{dh}{dt}\right) & 0 & 0 \\ 0 & \frac{1}{h}\frac{dh}{dt} & 0 \\ 0 & 0 & \frac{1}{r}\frac{dr}{dt} \end{bmatrix}. \tag{2}$$

Then

$$B_1^2 = 4 \begin{bmatrix} \left(\frac{1}{r}\frac{dr}{dt} + \frac{1}{h}\frac{dh}{dt}\right)^2 & 0 & 0 \\ 0 & \left(\frac{1}{h}\frac{dh}{dt}\right)^2 & 0 \\ 0 & 0 & \left(\frac{1}{r}\frac{dr}{dt}\right)^2 \end{bmatrix}. \tag{3}$$

The strain-acceleration tensor of an incompressible medium is determined by means of the Oldroyd contravariant derivative [6, 7]:

$$B_2^{(ij)} = \frac{D}{Dt} B_1^{(ij)} - \frac{\partial v_i}{\partial x_\beta} B_1^{(\beta j)} - \frac{\partial v_j}{\partial x_\beta} B_1^{(i\beta)},$$

$$B_2 = \begin{bmatrix} W_{11} & 0 & 0 \\ 0 & W_{22} & 0 \\ 0 & 0 & W_{33} \end{bmatrix}, \tag{4}$$

where

$$W_{11} = -2\left(\frac{1}{h}\frac{dh}{dt} + \frac{1}{r}\frac{dr}{dt}\right)^2 - \frac{2}{h}\frac{d^2h}{dt^2} - \frac{2}{r}\frac{d^2r}{dt^2} - 4\left(\frac{1}{h}\frac{dh}{dt}\right)\left(\frac{1}{r}\frac{dr}{dt}\right),$$

$$W_{22} = -\frac{6}{h^2}\left(\frac{dh}{dt}\right)^2 + \frac{2}{h}\frac{d^2h}{dt^2}, \quad W_{33} = -\frac{6}{r^2}\left(\frac{dr}{dt}\right)^2 + \frac{2}{r}\frac{d^2r}{dt^2}.$$

Substituting Eqs. (2)-(4) in (1) and determining the value of p from the condition $\sigma_{22} = 0$, we obtain

$$\begin{aligned} \sigma_{11} = {} & 2\eta_1\left(\frac{1}{r}\frac{dr}{dt} + \frac{2}{h}\frac{dh}{dt}\right) - 2\eta_2\left(\frac{1}{r}\frac{d^2r}{dt^2} + \frac{2}{h}\frac{d^2h}{dt^2}\right) + \\ & + 2(2\eta_3 - \eta_2)\left(\frac{1}{r}\frac{dr}{dt} + \frac{1}{h}\frac{dh}{dt}\right)^2 - 2(2\eta_3 - 3\eta_2)\frac{1}{h^2}\left(\frac{dh}{dt}\right)^2 - \\ & - 4\eta_2\left(\frac{1}{r}\frac{dr}{dt}\right)\left(\frac{1}{h}\frac{dh}{dt}\right), \end{aligned} \tag{5}$$

$$\begin{aligned} \sigma_{33} = {} & 2\eta_1\left(\frac{1}{r}\frac{dr}{dt} - \frac{1}{h}\frac{dh}{dt}\right) + 2\eta_2\left(\frac{1}{r}\frac{d^2r}{dt^2} - \frac{1}{h}\frac{d^2h}{dt^2}\right) + \\ & + 2(2\eta_3 - 3\eta_2)\left[\frac{1}{r^2}\left(\frac{dr}{dt}\right)^2 - \frac{1}{h^2}\left(\frac{dh}{dt}\right)^2\right], \end{aligned} \tag{6}$$

where the material functions are assumed to have the form

$$\eta_1 = \eta_{01}\exp\left[\frac{E}{R^*}\left(\frac{1}{T} - \frac{1}{T_0}\right) + s_1\Theta\right]\left(\frac{J_2}{2}\right)^{\frac{n-1}{2}}, \tag{7}$$

$$\eta_2 = \eta_{02}\exp\left[b_2\left(\frac{1}{T} - \frac{1}{T_0}\right) + s_2\Theta\right], \tag{8}$$

$$\eta_3 = \eta_{03}\exp\left[b_3\left(\frac{1}{T} - \frac{1}{T_0}\right) + s_3\Theta\right]. \tag{9}$$

The second invariant of the strain-rate tensor J_2 is given by the equation

$$\frac{J_2}{2} = \left(\frac{1}{r}\frac{dr}{dt}\right)^2 + \left(\frac{1}{r}\frac{dr}{dt}\right)\left(\frac{1}{h}\frac{dh}{dt}\right) + \left(\frac{1}{h}\frac{dh}{dt}\right)^2. \tag{10}$$

In Eq. (7) the longitudinal viscosity η_1 does not tend to infinity in the limit $\Theta \to 1$, since many polymers can be drawn at temperatures far below the equilibrium melting point if the film thickness is sufficiently small.

In order to determine $r(t)$, it is necessary to form a system of equilibrium equations for the forces acting on the tube as it is inflated. We use the classical equations of the couple-free theory of thin films for this purpose. The condition for equilibrium of the shell zone in the direction of its axis has the following form with allowance for the stated assumptions [1]:

$$2\pi r P_1 \cos\varphi + \pi\Delta p\,(R^2 - r^2) + 2\pi\rho g \int_z^Z rh \sec\varphi\, dz = F. \tag{11}$$

It must be noted that the quantity Z is not known beforehand; it must first be specified approximately and then refined. The force P_3 acting in the perpendicular direction and the tensile force P_1 are equalized by the differential pressure Δp in the tube inflation zone:

$$\frac{P_1}{R_1} + \frac{P_3}{R_3} = \Delta p. \tag{12}$$

Equation (12) represents the solution of the equilibrium equation for a shell element (Laplace equation) [1] and determines the conditions for the existence of a free interface. The principal radii of curvature R_1 and R_2 of the film shell are defined as

$$R_1 = -\frac{[1 + (r_z')^2]^{3/2}}{r_z''} = -\frac{\sec^3\varphi}{r_z''}, \tag{13}$$

$$R_3 = r\sqrt{1 + (r_z')^2} = \frac{r}{\cos\varphi}. \tag{14}$$

It is obvious from geometrical considerations that $r_z' = \tan\phi$. Accordingly

$$R_1 = -\frac{\sec\varphi}{\varphi_z'}. \tag{15}$$

The value of the velocity v_1 can be expressed in terms of the volumetric flow rate Q and the parameters r and h:

$$v_1 = \frac{Q}{2\pi rh}. \tag{16}$$

The forces P_1 and P_3 can be expressed in terms of the stresses: $P_1 = h\sigma_{11}$, $P_3 = h\sigma_{33}$. However, before we make use of Eqs. (5) and (6), we lower the order of their derivatives by introducing the auxiliary function $y = h_t'$ and making the transformations

$$\frac{dr}{dt} = \frac{dr}{dz}\frac{dz}{dt} = v_z \operatorname{tg}\varphi = v_1 \sin\varphi, \tag{17}$$

$$\frac{d^2r}{dt^2} = -v_1\sin\varphi\left(\frac{v_1\sin\varphi}{r} + \frac{1}{h}\frac{dh}{dt}\right) + v_1^2\cos^2\varphi\,\frac{d\varphi}{dz}. \tag{18}$$

Equation (18) has been derived with allowance for Eqs. (16) and (17) together with the relation

$$\varphi_t' = \varphi_z' v_1 \cos\varphi.$$

We thus arrive at the system of four first-order differential equations

$$2\pi r h\sigma_{11}\cos\varphi = F - \pi\Delta p\,(R^2 - r^2) - 2\pi\rho g \int_z^{z_R} rh\sec\varphi\, dz, \tag{19}$$

$$-h\sigma_{11}\cos\varphi\cdot\varphi_z' + \frac{h\sigma_{33}}{r}\cos\varphi = \Delta p, \tag{20}$$

$$h_z' = \frac{y}{v_1 \cos\varphi}, \tag{21}$$

$$r_z' = \operatorname{tg}\varphi, \tag{22}$$

where

$$\frac{1}{2}\sigma_{11} = \eta_1\left(\Gamma + \frac{2y}{h}\right) + \eta_2\Gamma\left(\Gamma - \frac{y}{h} - \Gamma\varphi_z' r \operatorname{ctg}^2\varphi\right) -$$
$$- 2\eta_2 v_1 \cos\varphi \frac{y_z'}{h} + (2\eta_3 - \eta_2)\left(\Gamma + \frac{y}{h}\right)^2 - (2\eta_3 - 3\eta_2)\frac{y^2}{h^2},$$
$$\frac{1}{2}\sigma_{33} = \eta_1\left(\Gamma - \frac{y}{h}\right) - \eta_2\Gamma\left(\Gamma + \frac{y}{h} - \Gamma\varphi_z' r \operatorname{ctg}^2\varphi\right) -$$
$$- \eta_2 v_1 \cos\varphi \frac{y_z'}{h} + (2\eta_3 - 3\eta_2)\left(\Gamma^2 - \frac{y^2}{h^2}\right), \quad \Gamma = \frac{v_1 \sin\varphi}{r}.$$

To calculate η_1, η_2, and η_3 according to Eqs. (7)-(9), it is required to know the variation of the film temperature. The system (19)-(22) must be augmented with the heat-balance equation

$$\rho c v_1 h \cos\varphi T_z' = \alpha(T_c - T) + \lambda\varepsilon(T_c^4 - T^4) + \rho\xi\frac{d\Theta}{dt}, \tag{23}$$

where the dependence of the density and specific heat of the polymer on the temperature and degree of crystallinity can be regarded as linear:

$$\rho(T, \Theta) = [\rho_0 + \gamma_1(T - T_0)](1 - \Theta) + [\rho_c + \gamma_2(T - T_c)]\Theta, \tag{24}$$

$$c(T, \Theta) = [c_0 + \beta_1(T - T_0)](1 - \Theta) + [c_c + \beta_2(T - T_c)]\Theta. \tag{25}$$

The degree of crystallinity Θ with any temperature-time history of the state T(t) is calculated on the assumption of "isokinetic conditions," where the time dependences of the growth rate of the crystal formations and the rate of formation of active centers are similar functions [3]:

$$\ln(1 - \Theta) = -\left(\int_{t^*}^{t} K^{\frac{1}{m}}[T(t)]\,dt\right)^m. \tag{26}$$

A similar relation has been derived previously [8], but from different basic principles. Equation (26) is one of the simplest equations for nonisothermal crystallization, yet it is the most practical relation, since the literature is devoid of exhaustive experimental material requiring other more complex models [9]. The function K(T), which represents the reciprocal half-period of crystallization and characterizes the rate of crystallization, is determined by means of the equation for the nucleation rate in combination with the William–Lendel–Ferry equation for the activation energy [3]:

$$K(T) = K_0 \exp\left[-\frac{AT}{T - T_g + B} - \frac{CT_m}{T(T_m - T)}\right]. \tag{27}$$

We have thus obtained the system (19)-(23) of five first-order differential equations in the five unknown functions h(z), r(z), ϕ(z), y(z), and T(z), which can be solved numerically with allowance for Eqs. (7)-(9) and (24)-(27) to determine the fundamental parameters of the blown-film extrusion process. In the present study we use the Merson modification of the Runge–Kutta method. The shape of the tube (bubble) is shown in Fig. 2, along with the distributions of the film thickness and temperature along the height of the tube during the formation of an "unoriented" film from the melt. The model medium has the constants E = 60 kJ/mole, C_0 = 2500 J/kg K, T_m = 165°C, c_c = 1800 J/kg K, $\lambda = 5.67 \cdot 10^{-8}$ W/m²·K⁴, ρ_0 = 1104 kg/m³, ρ_c = 1210 kg/m³, η_{01} = 2700 Pa·sec², $b_2 = b_3 = 0$, n = 1, T_g = 20°C.

The parameters of the process are Δp = 1.2 Pa, T_c = 20°C, T_0 = 230°C, r_0 = 0.25 m, Q = $5.0 \cdot 10^{-5}$ m³/sec, $h_0 = 1.2 \cdot 10^{-3}$ m, α = 40 W/m²·K, F = 2.2 N.

Figure 3 shows the shape of the tube and the height distributions of the film thickness and temperature for the formation of an oriented film, where the tube is inflated during heating, but below the melting point of the polymer. The calculations are carried out on the assumption that the degree of crystallinity is constant and equal to the maximum attainable value. The physical properties of the polymer are the same as in Fig. 1, and the process parameters are $T_C = 125°C$, $T_0 = 90°C$, $\Delta p = 3000$ Pa, $E = 110$ kJ/mole, $r_0 = 0.3$ m, $Q = 5.2 \cdot 10^{-5}$ m^3/sec, $h_0 = 2.4 \cdot 10^{-4}$ m, $\alpha = 20$ W/m$^2 \cdot$K, $F = 11{,}000$ N.

We note in conclusion that the equations for the stresses σ_{11} and σ_{33} (5) and (6) are simplified considerably in the important practical case of the formation of isotropic films in the uniform biaxial stretching regime, when

$$\frac{1}{2h}\frac{dh}{dt} = -\frac{1}{r}\frac{dr}{dt},$$

viz.:

$$\frac{1}{3}\sigma_{11} = \frac{\eta_1}{h}\frac{dh}{dt} - \frac{\eta_2}{h}\frac{d^2h}{dt^2} + (2\eta_2 - \eta_3)\frac{1}{h^2}\left(\frac{dh}{dt}\right)^2,$$

$$\frac{1}{3}\sigma_{33} = -\frac{\eta_1}{h}\frac{dh}{dt} - \frac{\eta_2}{h}\frac{d^2h}{dt^2} + (2\eta_2 - \eta_3\lambda\frac{1}{h^2}\left(\frac{dh}{dt}\right)^2.$$

NOTATION

r, r_0, variable and initial radii of tubular bubble; h, h_0, variable and initial film thicknesses; T, T_0, instantaneous film temperature and film temperature at exit from extruder; F, axial force on drawing rollers; Δp, excess pressure inside tube; η_{01}, longitudinal viscosity at temperature T_0; η_{02}, η_{03}, elastic constants at temperature T_0; J_2, second invariant of strain-rate tensor; E, activation energy of longitudinal flow; R^*, gas constant; Θ, degree of crystallinity; ρ_0, c_0, density and specific heat of melt at temperature T_0; ρ_C, c_C, density and specific heat of polymer at ambient temperature T_C; α, heat-transfer coefficient; λ, Stefan–Boltzmann constant; ε, emissivity; Q, volumetric flow rate of polymer from die; v_1, velocity in x_1 direction; σ_{11}, σ_{22}, σ_{33}, normal stresses in x_1, x_2, x_3 directions, respectively; ξ, latent heat of crystallization.

LITERATURE CITED

1. C. D. Han, Rheology in Polymer Processing [Russian translation], Moscow (1979).
2. C. D. Han and J. Y. Park, J. Appl. Polym. Sci., 19, 3277-3283 (1975).
3. A. Zyabitskii, Theoretical Principles of Fiber Formation [in Russian], Moscow (1979).
4. C. J. S. Petrie, AIChE J., 21, 275 (1975).
5. M. L. Fridman, Technology of the Processing of Crystal Polyolefins [in Russian], Moscow (1977).
6. J. L. White, J. Appl. Polym. Sci., 8, 1129-1146 (1964).
7. V. G. Litvinov, Mekh. Polim., No. 9, 35-37 (1975).
8. V. A. Borokhovskii et al., Vysokomol. Soedin. B, 17, No. 1, 35-37 (1975).
9. D. M. Rein et al., Dokl. Akad. Nauk SSSR, 255, No. 6, 1422-1426 (1980).