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MATHEMATICAL MODEL OF STRAIN OF DROPLETS OF A DISPERSED-PHASE POLYMER IN FLOW OF MOLTEN POLYMER BLENDS

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A mathematical model of strain of a dispersed-phase-polymer droplet in flow of a molten polymer blend from a wide reservoir to a narrow one has been created with the use of the structural-continuum approach. The system of differential equations obtained has numerically been solved by the Runge–Kutta method. The model satisfactorily describes the actual processes of flow of molten polymer blends in the entrance zone of a molding orifice: the values of the droplet strains are a function of the relation of the viscosities of the starting components and their absolute values, the volume concentration of the dispersed phase, the interphase tension, and the elasticity of the droplet. The adequacy of the model created has been confirmed by comparison of the droplet strains calculated using the equations obtained and the theoretical conclusions and experimental results.

The use of polymer blends is one efficient method of creating materials with a prescribed set of properties. Polymer blends may be considered as specific colloidal systems whose properties are determined by the type of structure formation and the surface phenomena at the phase boundary [1]. Processing of molten blends offers a new method of molding of ultrafine synthetic fibers (microfibers) of diameter from several fractions to tenths of a fraction of a micrometer. This phenomenon has been called specific fiberization [2]. The empirical approach is dominant in investigating the processes of formation of a structure in polymer dispersions. Romankevich et al. have made an attempt to describe the process of formation of microfibers in the matrix of the other polymer in the case of flow of a molten binary mixture; the emphasis was on the strain of a polymer droplet in the channel of the molding orifice and after the exit from it or in subsequent thermoorientational stretching [3, 4]. The mechanism of the phenomenon of specific fiberization, whose essence is that the fiberization of one polymer in the mass of the other (matrix) polymer is carried out in the field of tensile forces arising in transition from a wide reservoir to a narrow one, has been formulated and experimentally confirmed in [2].

The present work seeks to create a mathematical model of strain of a droplet of the dispersed-phase component in flow of a molten polymer blend in the entrance zone of a molding orifice.

To describe the processes occurring in dispersion flow one must know of numerous characteristics of the dispersion: the viscoelastic properties of the components, the volume concentration of the dispersed phase, the shape, size, and interaction of particles, etc. It is possible to allow for these indices in detail within the framework of the structural (microscopic) approach which has widely been used in investigating comparatively simple media, such as diluted suspensions with simply shaped particles. The possibility of using the structural method in the rheology of dispersions is limited by their diversity and complex morphology. The structural-continuum method proposed in [5, 6] combines the phenomenological (macroscopic) and structural (microscopic) approaches and enables one to allow for all the fundamental propositions of the continuum mechanics (continuity of the medium and discontinuity of functions characterizing its motion and state) and the distinctive features of the behavior of the dispersed phase. Thus, in the structural-continuum model, each point of the dispersion is characterized by the density, velocity, and pressure (just as in the classical mechanics) and by the so-called internal parameters, in addition. The latter can be scalars, vectors, and tensors; their number and form are determined by the nature of the microstructure of the dispersion under study. In

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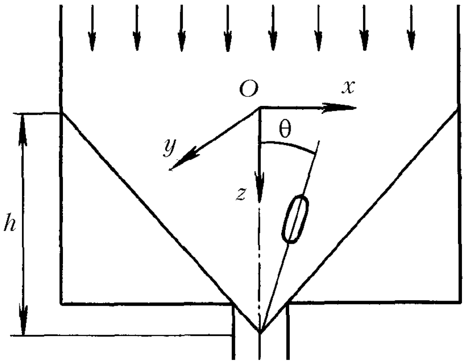

Fig. 1. Diagrammatic representation of the zone of entry into the molding orifice.

the case of diluted suspensions of deformable particles one introduces one internal parameter, i.e., the vector allowing for the influence of the orientation, hydrodynamic interaction, and strain of suspended particles on the behavior of the medium, into the rheological equation. The coordinates of a particle in the space described by the vector determine the dependence of the rheological properties of the suspension on the particles' orientation (anisotropy), whereas the modulus of the vector determines the value of strain in the case of flow [7].

Objects and Methods of Investigation. To create the model of strain of droplets of the dispersed-phase polymer in the other polymer we took the droplet to be an ellipsoid that changes its dimensions in the process of interaction with the dispersion medium but in so doing retains its volume. The dispersion medium was modeled by a Newtonian fluid with the aim of simplifying mathematical calculations and solving the equations obtained. The zone of entry into the narrow reservoir from the wide one is shown in Fig. 1.

The system of equations describing the isothermal motion of a continuous medium consists of the equation of uniaxial-tension flow, the flow-continuity equation, and the rheological equation of state of the fluid. The velocity field resulting from the superposition of the longitudinal strain on the shear flow can be described as follows:

$$V_x = -u/2x\,, \quad V_y = -u/2y\,, \quad V_z = u\,, \quad u = V/h\,. \tag{1}$$

The rate tensor of uniaxial tension strain (d_{ij}) is determined as [8]

$$d_{ij} = \begin{pmatrix} u & 0 & 0 \\ 0 & -u/2 & 0 \\ 0 & 0 & -u/2 \end{pmatrix}; \tag{2}$$

it is allowed that circulatory flow is absent.

The flow-continuity equation in the case of a constant density has the form

$$u_{i,i} = 0\,. \tag{3}$$

The rheological state of the dispersion under study has been described by the equation obtained from the viewpoint of the structural-continuum approach for diluted suspensions with ellipsoidal deformable particles [5]. This equation contains one internal parameter — the vector $\mathbf{n}_i$ related to the microstructural element — of a suspended deformable particle, depends on the character of flow of the medium, and may change in space and with time. Its direction coincides with the vector of the axis of symmetry of an ellipsoid, and its modulus coincides with the length of the semiaxis of rotation a, i.e., $|\mathbf{n}_i| = a$. The rheological equation describing the anisotropic fluid has the form [5]

$$\dot{n}_i = \lambda_1 n_i + \lambda_2 d_{km} n_k n_m n_i + \lambda_3 d_{ij} n_j\,, \tag{4}$$

where the value of $\dot{n}_i$ is taken at a corresponding point and at a corresponding instant of time.

In the equations used, "·" above the symbol denotes the total derivative with respect to time; the double subscripts denote summation from 1 to 3 over the given subscript.

The quantities λ_1, λ_2, and λ_3 characterize the rheological properties of the system. Equations for their determination have been obtained in [6]:

$$\lambda_1 = \frac{-2ab^2\beta_0'' G \dfrac{a}{a_0}\left(1 - \dfrac{q}{q_0}\right)}{\mu\left(2 + 3ab^2\beta_0''\dfrac{\eta}{\mu}\right)}(1 - M\Phi)\,,$$

$$\lambda_2 = \frac{2 + (N - 2M)\Phi}{a^2\left(2 + 3ab^2\beta_0''\dfrac{\eta}{\mu}\right)} - \frac{a^2 - b^2 + \dfrac{15(a^2 - b^2)\,(\alpha_0 + \beta_0) + 4\,(a^2 + b^2)\,(\beta_0 - \alpha_0)}{6ab^2\beta_0' B}\Phi}{a^2 + b^2 + \dfrac{15\,(a^2 - b^2) + 4\,(a^2 + b^2)^2}{6ab^2 B}\Phi}\,, \qquad (5)$$

$$\lambda_3 = \frac{a^2 - b^2 + \dfrac{15(a^2 - b^2)\,(\alpha_0 + \beta_0) + 4\,(a^2 + b^2)\,(\beta_0 - \alpha_0)}{6ab^2\beta_0' B}\Phi}{a^2 + b^2 + \dfrac{15\,(a^2 - b^2)^2 + 4\,(a^2 + b^2)^2}{6a^2 bB}\Phi}\,,$$

where

$$M = \frac{4}{ab^2\left(2 + 3ab^2\dfrac{\eta}{\mu}\beta_0''\right)}\left\{\frac{5}{6\,(\alpha_0 + 2\beta_0 - 2\beta_0'\,(a^2 + b^2))} - \right.$$

$$\left. - \frac{100\beta_0' a^2\,(2\beta_0' a^2 - \alpha_0 - 2\beta_0)}{(\alpha_0 + 2\beta_0)\,(\alpha_0 + 2\beta_0 - 2\beta_0'\,(a^2 + b^2))}\left[\frac{1}{24a\beta_0} - \frac{1}{2\beta_0'^2 a^2 - (\alpha_0 + 2\beta_0)}\right]\right\};$$

$$N = \frac{4}{ab^2}\left[\frac{10}{18\beta_0''} + \frac{5\beta_0''}{8b^4\alpha_0'^2} - \frac{10\,(\beta_0'' - \alpha_0'')\,(2b^2\alpha_0' + 3\beta_0'')}{72b^4\alpha_0'^2}\right];$$

$$B = a^2\alpha_0 + b^2\beta_0\,.$$

The quantities a_0, β_0, a_0', β_0', a_0'', and β_0'' have been determined in [9]. To solve Eq. (4) for the strain and orientation of an ellipsoid (polymer droplet) in the flow we expand Eq. (4) in the coordinate axes x, y, and z:

$$\dot{n}_x = \lambda_1 n_x + \lambda_2\frac{u}{2}(-n_x^2 - n_y^2 + 2n_z^2)\,n_x + \lambda_3\left(-\frac{u}{2}\right)n_x\,,$$

$$\dot{n}_y = \lambda_1 n_y + \lambda_2\frac{u}{2}(-n_x^2 - n_y^2 + 2n_z^2)\,n_y + \lambda_3\left(-\frac{u}{2}\right)n_y\,, \qquad (6)$$

$$\dot{n}_z = \lambda_1 n_z + \lambda_2\frac{u}{2}(-n_x^2 - n_y^2 + 2n_z^2)\,n_z + \lambda_3 u n_z\,.$$

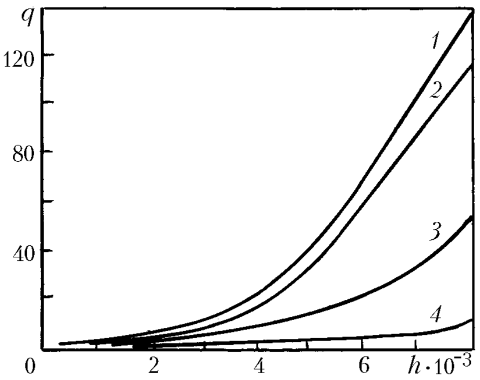

Fig. 2. Strain of a dispersed-phase droplet vs. depth of descent and angle of orientation θ (rad) for $\eta/\mu = 1$: 0 (1), 0.2 (2); 0.4 (3); 0.7 (4).

In the spherical coordinate system, the location of the vector $\mathbf{n}_i$ may be determined as follows:

$$n_x = a \cos \varphi \sin \theta , \quad n_y = a \sin \varphi \sin \theta , \quad n_z = a \cos \theta . \tag{7}$$

Taking the derivatives of the left-hand and right-hand sides of Eqs. (7), equating the right-hand sides of the corresponding equations (6) and (7), and carrying out the required transformations, we obtain the system of differential equations

$$\dot{\varphi} = 0 ; \quad \dot{\theta} = -\frac{3}{4} u \lambda_3 \sin (2\theta) ; \quad \frac{\dot{a}}{a} = \lambda_1 + \frac{u}{2} (\lambda_2 a^2 + \lambda_3) (2 - 3 \sin^2 \theta) . \tag{8}$$

It represents a mathematical model (created for the first time) of strain of droplets of the dispersed-phase components in flow of a molten polymer blend in the entrance zone of the molding orifice. The model enables us to determine the strain and the orientation of droplets in the flow as functions of the rheological properties of the components of the blend and the volume concentration of the dispersed phase.

Results and Their Discussion. For convenience of solution of the system of equations (8) we have passed to dimensionless variables in the equation for determination of the strain rate from the formula $q = a/b$. The system of equations (8) may be written in new variables as

$$\dot{\varphi} = 0 ; \quad \dot{\theta} = -\frac{3}{4} u\lambda_3 \sin (2\theta) ; \quad \frac{\dot{q}}{q} = \frac{3}{2} \left(\lambda_1 + \frac{u}{2} \left(\lambda_2 r_0^2 q^{4/3} + \lambda_3 \right) \left(2 - 3 \sin^2 \theta \right) \right) . \tag{9}$$

This system of differential equations was solved numerically by the Runge–Kutta method using a program specially written in the Object Pascal language in the Delphi environment.

The adequacy of the model created was checked by comparison of the droplet strains calculated using the equations obtained to experimental results. For this purpose we used the data of [10], where the influence of the composition of the blend and the relation of the viscosities of the components on the phenomenon of specific fiberization was investigated with the example of polyoxymethylene/ethylene-vinyl acetate copolymer (POM/EVAC) blends. Using the model, we calculated the strain and orientation of a POM droplet in the flow as it approached the entrance to the molding orifice. The depth of descent of the droplet over the height of the entrance zone was determined from the equation

$$h (t) = h_0 (\exp (ut) - 1) . \tag{10}$$

The results obtained demonstrate that the strains q substantially depend on the orientation of the droplet in the flow (Fig. 2). It is seen that polymer droplets are strained in the direction of flow when tensile stresses begin to be generated in the reservoir. The value of q grows as the droplet approaches the entrance to the molding orifice. The strain is maximum on the flow axis ($\theta = 0$) and decreases with distance from the axis. The q values calculated using

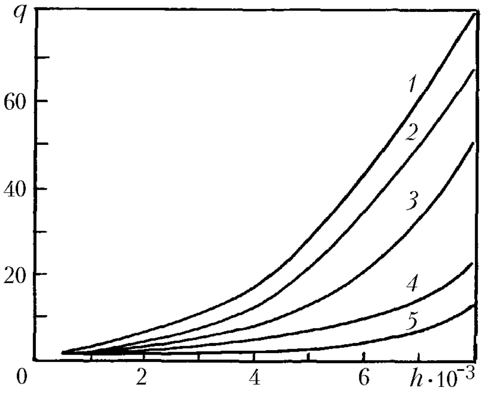

Fig. 3. Strain of a dispersed-phase droplet vs. depth of descent and viscosity relation η/μ for $\theta = 0.4$ rad: 0.65 (1); 0.85 (2); 1.05 (3); 4.14 (4); 10.77 (5).

the model are in good agreement with the conclusions of [8], according to which the melt moves with acceleration in transition from a wide reservoir to a narrow one: the velocity grows as the entrance is approached and has its maximum values on the axial line. Different degrees of strain of POM droplets in the flow, in addition to aggregation, are one reason for the spread in the diameters of POM microfibers, which has been established experimentally [2, 10].

It is well known that the microstructure of extrudates of polymer blends is formed in flow and is determined by such microrheological processes as strain, disintegration, coalescence, and migration of droplets of the dispersed-phase polymer. The degree of manifestation of one enumerated process or another largely depends on the relation of the viscosities of the dispersed-phase polymer and the matrix (η/μ) [2, 10]. The calculations (carried out using the model) of the strain of a POM droplet as a function of the relation of the POM and EVAC viscosities have shown that, for $\eta_{\mathrm{pom}}/\mu_{\mathrm{evac}} < 1$, the quantity q grows as compared to the strain for $\eta_{\mathrm{pom}}/\mu_{\mathrm{evac}} \geq 1$ (Fig. 3). The latter is natural and has been confirmed experimentally. It has been established in [11] that when the viscosities of the melts of the dispersed phase and the medium are equal, the deformable component experiences the same strain as the continuous phase, but if η of the dispersed phase is lower than the viscosity of the medium it is strained better. An increase in η/μ is accompanied by a poorer strain of the dispersed-phase component. The data on the strain calculated using the model agree with these conclusions (Fig. 3). A comparison of experimental results that reflect the character of structure-formation processes in extrudates of the POM/EVAC blends with change in the viscosity relations [10] and the strain q of a POM droplet, calculated from the model, demonstrates that the mathematical model developed describes the actual process of strain of a droplet of the dispersed-phase polymer in the entrance zone (Tables 1 and 2). The increase (established experimentally) in the average diameter of POM microfibers and the decrease in q for $\eta_{\mathrm{pom}}/\mu_{\mathrm{evac}} = 0.65$ disagree with the calculated data and are inconsistent with theoretical conclusions [11]. This is attributable to the migration of droplets of a low-viscosity dispersed phase to the capillary walls, because of which the center of the flow is depleted and most of the dispersed phase is strained at the periphery, where the q values are much lower than those on the flow axis.

The elasticity of a molten droplet of the dispersed-phase polymer in the model developed was allowed for in terms of the elastic modulus G. The q values calculated from the model show that the theory proposed correctly describes the influence of the elasticity on the degree of strain of the dispersed phase: droplets with a higher elasticity are strained to a lesser extent, i.e., they are more tension- and shear-resistant.

In addition to the relation of the viscosities of the blend components, their absolute values also make the determining contribution to microrheological processes in flow of molten polymer blends. The latter is confirmed by the results of calculations, using the model, of the strains of dispersed-phase droplets for the cases where $\eta/\mu = 1$ (Table 3). The theory developed demonstrates that the lower the absolute values of the component viscosities, the larger the strains of the components, e.g., for $0.5\eta/0.5\mu$ and $h = 8$ the strain is 102.9, whereas for $2\eta/2\mu$ it is only 9.4. However, the minimum values of the viscosities are bounded by the capacity of polymers for fiberization and by the enhancement of coalescence processes. Evidently, there are optimum values of the viscosities of the initial parameters for which the strain will be maximum for $\eta/\mu = 1$. Thus, selection of the absolute values of the viscosities of polymers

TABLE 1. Experimental Diameters of Fibers

η_{pom}/η_{evac}	d_{av}	d_{min}	d_{max}
0.65	6.9	2.6	31.2
0.85	4.9	1.7	20.8
1.05	5.5	2.1	26.0
4.14	7.3	2.6	30.8
8.64	8.5	5.2	31.2
10.77	10.2	5.2	31.2

TABLE 2. Strains q of a Droplet of the Dispersed-Phase Polymer

η_{pom}/η_{evac}	d_{av}	d_{min}	d_{max}	θ, rad		
				0	0.4	0.7
0.65	22.5	2.3	97.3	249.1	76.9	11.2
0.85	37.6	4.3	184.0	207.8	64.9	10.1
1.05	31.6	3.1	134.0	138.7	48.1	7.0
4.14	20.7	2.4	96.3	112.5	19.7	3.8
8.64	16.5	2.3	97.3	103.4	14.3	2.0
10.77	12.5	2.3	34.4	57.9	9.5	1.3

TABLE 3. Influence of the Absolute Values of the Viscosities of the Starting Components on the Strain q (θ = 0.4 rad and η/μ = 1)

$h \cdot 10^{-3}$	$2\eta/2\mu$	$1.5\eta/1.5\mu$	η/μ	$0.75\eta/0.75\mu$	$0.5\eta/0.5\mu$
2	1.2	1.5	1.8	4.9	13.2
4	3.1	4.4	7.1	31.2	61.1
8	9.4	24.5	48.1	79.2	102.9

TABLE 4. Strain of a Dispersed-Phase Droplet q as a Function of the Interphase Tension (θ = 0.4 rad and η/μ = 1)

$h \cdot 10^{-3}$	$\gamma_{\alpha\beta}$				
	0.1	0.5	1.0	2.0	3.0
2	2.4	2.1	2.0	1.9	1.8
4	70.8	44.8	32.0	21.1	11.3
8	228.4	67.5	40.4	29.0	17.7

in the blend may be one method of control of structure formation with the aim of producing microfibers with diameters smaller than tenths of a fraction of a micrometer.

It is well known that there is a competitive influence of the rheological properties of the polymers blended and the composition of the blend on the microstructure of extrudates [12]. Calculations of the strains using the model have shown a growth in q as the concentration of the dispersed-phase polymer increases from 4 to 16 vol. %. The latter is in agreement with the Starita theory [13], according to which the degree of strain is in direct proportion to the volume concentration of the dispersed phase. However, in practice, this conclusion is not necessarily confirmed for polymer blends. In the case of realization of specific fiberization, an increase in the content of the fiberizing component is accompanied by the growth in the average microfiber diameter [2, 10].

Recently, it has been shown theoretically and experimentally that one can control the type of structure in multicomponent polymer compositions, changing the interphase tension ($\gamma_{\alpha\beta}$) [14]. A decrease in $\gamma_{\alpha\beta}$ leads to an improvement of the strain of the dispersed-phase component. On the other hand, the capacity of a droplet for straining is largely determined by its elasticity. On condition that the forces of internal elasticity and surface tension are equal, the rheological constant λ_1 (5) in the model created will have the form

$$\lambda_1 = \frac{-2ab^2\beta_0'' \frac{\gamma_{\alpha\beta}}{R_0} \frac{a}{a_0} q^{2/3}}{\mu\left(2 + 3ab^2\beta_0'' \frac{\eta}{\mu}\right)} (1 - M\Phi) .$$

The droplet strains calculated using the model as functions of the degree of interphase tension are given in Table 4. The results demonstrate that the model created adequately describes the process of strain of a droplet of a dispersed-phase polymer: a decrease in $\gamma_{\alpha\beta}$ is accompanied by the growth in the strain. The calculated results are in good agreement with experimental data on investigation of structure-formation processes in compatibilized polymer blends. A decrease in the interphase tension in polypropylene/copolyamide (PP/CPA) blends due to the introduction of compatibilizers contributes to the growth in the degree of dispersity and to the improvement of the fiberization of PP in the CPA matrix [15].

Thus, we have created a mathematical model of strain of a droplet of a dispersed-phase polymer in the case of flow of a molten polymer blend in the entrance zone of the molding orifice. The model enables us to calculate the strains of the droplet in the flow and allows for the influence of the volume concentration and elasticity of the dispersed-phase polymer, the relation of the viscosities of the components, and the interphase tension. The system of differential equations obtained adequately describes the actual processes of flow of molten polymer blends in transition from a wide reservoir to a narrow one.

NOTATION

$\dot{a}/a$, rate of strain of an ellipsoid; a, b, a_0, and b_0, semiaxes of an ellipsoid in strained and unstrained states; d_{av}, d_{min}, and d_{max}, microfiber diameter (average, minimum, and maximum ones), μm; d_{ij}, components of the rate tensor of uniaxial tension strain; G, elastic modulus of the dispersed phase; h, height of the molten-blend layer, m; h_0, initial height of the molten blend, m; $\mathbf{n}_i$, internal parameter characterizing the dispersion microstructure; $\dot{n}_i$, time derivative of the orientation vector $\mathbf{n}_i$; n_k, n_m, n_i, vector coordinates; $q = a/b$ and $q_0 = a_0/b_0$, strains; R_0, initial radius of a droplet, μm; t, time, sec; u, intensity of uniaxial-tension flow; V, velocity of descent of the upper layer of the molten blend, m/sec; V_z, V_x, and V_y, components of the velocity V in the direction of flow and in perpendicular directions respectively, m/sec; $\gamma_{\alpha\beta}$, interphase tension at the boundary of the phases α and β, mN/m; η, viscosity of the dispersed phase, Pa·sec; θ, angle between the direction of flow and the ellipsoid's axis of rotation, rad; λ_1, λ_2, and λ_3, rheological parameters of the system; μ, viscosity of the dispersion medium, Pa·sec; φ, angle between the Ox axis and the projection of the axis of rotation of an ellipsoidal particle onto the area Oxy, rad; Φ, volume concentration of the dispersed phase, vol. %. Subscripts: av, average; max, maximum; min, minimum; 0, initial (starting); α and β, polymers of the dispersed phase and the dispersion medium; x, y, and z, coordinate axes; pom, polyoxymethylene; evac, ethylene-vinyl acetate copolymer.

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