

CONVECTIVE TRANSFER PROCESSES IN CHEMICALLY STABLE
AND REACTING LIQUIDS WITH AN INTERNAL STRUCTURE

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Dynamic processes are studied in media with softly and hardly internal destroyable configurations. A method is examined for the measurement of properties of media with spatial separation of the chemical transformation stages and thermal hydrodynamic measurements.

The specifics of the mechanical and thermal behavior of rheologically complex liquids is determined macroscopically by the developed internal configuration. There is a fluctuating molecular network enclosing the whole liquid volume [1] in solutions and melts of polymers. The network node exists just a certain time and decays because of thermal motion. As the stress grows the rate of node formation and their number reduce while the decay tempo grows. In many colloidal and disperse systems the filler particles form a branched skeleton with elastic properties by mutual interaction [2]. In contrast to the macromolecular network, the skeleton is destroyed only under sufficiently high stresses. Phenomenologically, two limiting kinds of internal configuration destruction can be extracted depending on the stress-strain state of the liquid: "soft," starting with arbitrarily small stresses, and "hard," being developed only under stresses greater than a certain critical value. In the first case the relaxation properties of the configuration are important and in the second the magnitude of the critical stress also. The listed mechanisms for the change in internal configuration of a liquid in time are reversible. For a wide circle of flowing systems the internal configuration varies irreversibly in a real-time scale because of chemical transformations. Among such chemically unstable liquids are polymerizing systems, reactive oligomers, many disperse and colloidal media, etc. Studied most is the physical chemistry of reactive systems during polymer synthesis and transformation [3].

CHEMICALLY STABLE MEDIUM

Required for the mathematical simulation of flow and heat transfer problems of liquids with time-varying internal configuration is a rheological equation of state (RES) connecting the stress being developed in the liquid with the prehistory of the velocity or deformation field. Most studied are the RES of polymer solutions and melts. Around 30 kinds of RES of media are collected in [4] for which all parameters have been measured for at least one liquid. Parameters of the internal configuration are not contained explicitly in the majority of RES. This is related to the large multivariety of structural transformation mechanisms in different media and the significant difficulties in measuring their characteristics. Such an approach is completely acceptable for chemically stable materials. Within its framework the appearance of properties of liquids with soft and hard destruction of the internal configuration in nonstationary channel flows is investigated in [5-10]. They illustrate the specific features of nonstationary transport processes in liquids with internal configuration.

Most important for practice are the flow development and cessation modes as well as the oscillating mode. The strong dependence of the rheological properties of a liquid on the temperature specifies the influence of the thermal mode on the nature of the flow. The velocity profile in a hydraulically and thermally long coaxial cylindrical channel (the length considerably exceeds the size of the input hydrodynamic and thermal sections) is determined from the equations

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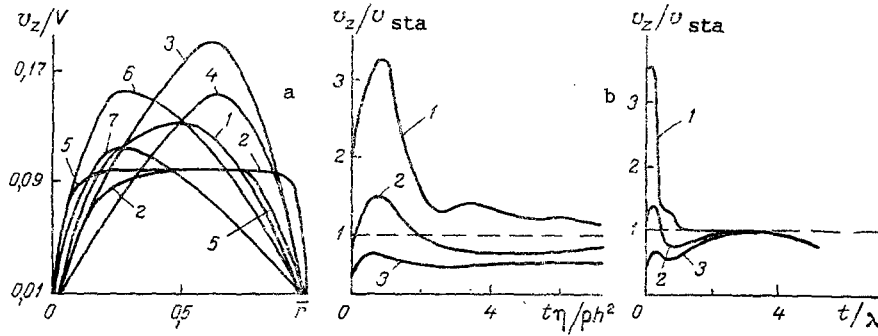


Fig. 1. Flow development for $El = 10$, $\alpha = 2$: a) velocity profiles for $R_2 = 2R_1$; $We = 1$; $b = 0.1$; $\tau\eta/\rho h^2 = 0.1$ (2, 5); 4.5 (3, 6); 30 (1, 4, 7); $\nu = 0$ (1); 0.25 (5, 6, 7); -0.25 (2, 3, 4); b) velocity change for $R_2 = 1.1R_1$; $\nu = 0$; $We = 1$ (1), 10 (2), 20 (3). The wave stage is displayed in an enlarged scale $\tau\eta/\rho h^2 = El t/\lambda$.

$$\rho \frac{\partial v_z}{\partial t} = -\frac{\partial p}{\partial z}(t) + \frac{1}{r} \frac{\partial}{\partial r}(r\tau),$$

$$v_z|_{t=0} = \tau|_{t=0} = 0, \quad v_z|_{r=R_1} = v_z|_{r=R_2} = 0.$$

(1)

SOFT DESTRUCTION OF THE CONFIGURATION

On the basis of results of numerical computations performed in [5-8], we analyze the principal features of nonstationary flows of liquids with soft destruction of the configuration. An RES with a dependence of the memory function on the strain rate tensor and temperature invariants that allow a temperature-time superposition is used in computations for the excess stress tensor. For the shear flow under consideration the tangential stress is related to the prehistory of the shear velocity by the integral relation

$$\tau(r, t) = \int_{-\infty}^t m[t, t', s(r, t'), \Theta(r, t')] \int_{t'}^t \frac{\partial v_z}{\partial r}(r, t'') dt'' dt',$$

$$m = \sum_{k=1}^{\infty} \frac{\eta_k}{\lambda_k^2} f_k[s(r, t')] \exp\left\{-\int_{t'}^t \frac{g_k[s(r, t'')]}{\lambda_k} dt''\right\}, \quad s = \left| \frac{\partial v_z(r, t)}{\partial r} \right|,$$

$$\lambda_k = \lambda_s a(\Theta)/k^\alpha, \quad \eta_k = \eta_0 a(\Theta)/\zeta(\alpha) k^\alpha, \quad a(\Theta) = \exp\left[\frac{E}{R_T} \left(\frac{1}{\Theta} - \frac{1}{\Theta_s}\right)\right],$$

which includes a large quantity of RES relative to slightly viscous polymer liquids [4]. The difference is in the selection of the functions $f_k(s)$ and $g_k(s)$ that characterize the influence of the strain rate on the probability of formation and decay of the fluctuation network nodes. Numerical computations were executed for liquids with constant properties $f_k = g_k = 1$ and the rheological models of Meisster (M), BByrd-Carreaux (BC) and Macdonald-Byrd-Carreaux (MBC). The M and BC models correspond to extreme cases of RES physical nonlinearity, the MBC to the intermediate situation. Taken into account in M is the reduction of the relaxation time as the strain rate grows $f_k = 1$, $g_k = 1 + c\lambda_k s$, $c \sim 1$, in BC the reduction of the elasticity moduli $f_k = 1/(1 + \lambda_k^2 s^2)$, $g_k = 1$, and both characteristics in the MBC

$$f_k = \frac{1 + \lambda_k' s}{1 + \lambda_k s}, \quad g_k = \frac{(1 + \lambda_k' s)^{3/2}}{(1 + \lambda_k s)^{1/2}}, \quad \frac{\lambda_k'}{\lambda_k} = \text{const} < 1.$$

The fundamental criteria of convective and thermal nonstationarity, the physical nonlinearity of the RES, and the thermal factor are: the elasticity number $El = \lambda\eta/\rho h^2$, the correlative relaxation time (λ) and the viscous liquid flow development in the gap ($\rho h^2/\eta$); the Weissenberg number $We = \lambda V/h = \lambda h(\partial p/\partial z)_0/\eta_0$ constructed according to the velocity scale $V = h^2(\partial p/\partial z)_0/\eta_0$ and governing the influence of the strain rate on the rheological properties; the characteristic of the linear relaxation time spectrum α ; the relative temperature

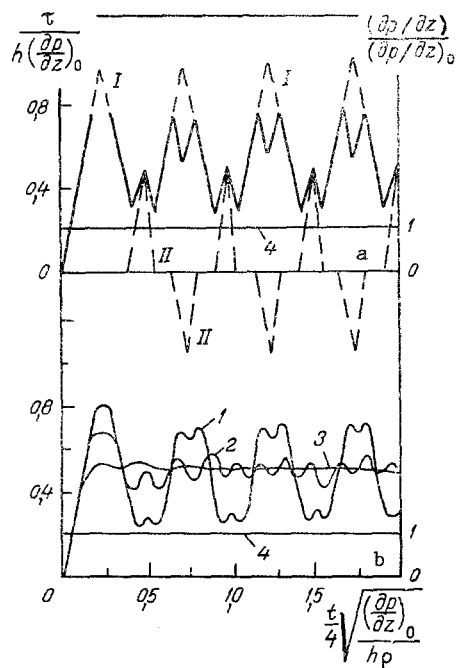


Fig. 2

Fig. 2. Change in stress: a) analytic solution for $Y/h(\partial p/\partial z)_0 = 0.75$; b) numerical computation $Y/h(\partial p/\partial z)_0 = 0.75$ (1); 0.6 (2); 0.45 (3); $E = 50$, $Y/G = 1$; 4) $(\partial p/\partial z)/(\partial p/\partial z)_0$.

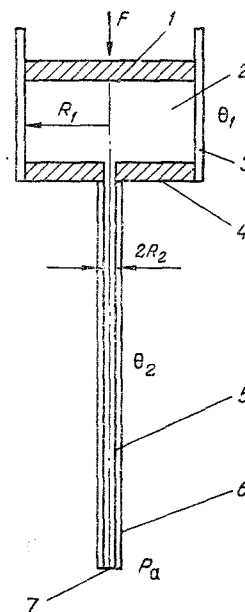


Fig. 3

Fig. 3. Diagram of the experimental apparatus: 1) piston; 2) tank; 3) thermostatted tank skin; 4) heat insulated cover; 5) capillary; 6) thermostatted capillary skin; 7) thermocouple.

drop across the gap $v = (\theta_1 - \theta_2)/\theta_S$; the temperature sensitivity of the viscosity $b = R_T\theta_S/E$; the Prandtl number Pr displaying the relationship of the times of thermal and shear perturbation passage through the gap. The ratio between the relaxation and propagation times is determined by the complex $\sim El/Pr$. For polymer and disperse systems $Pr \gg 1$, for relatively low-viscosity mixtures $El/Pr \sim 1$. Consequently, the change in thermal conditions with time can be neglected in unsteady hydrodynamic modes while hydrodynamics can be considered quasistationary in unsteady thermal modes.

Elastic properties do not appear in transition hydrodynamic modes with $El \ll 1$. The liquid behaves as a Newtonian fluid for $We \ll 1$ and nonlinearly viscous for $We \gg 1$. Propagation of shear perturbations is of diffusion nature for $El \ll 1$. Consequently, the velocity and tangential stress grow monotonically for an increase, and diminish for a decrease in the pressure drop. The capability of the liquid to accumulate reversible strains appears for $El \sim 1$. In this case, it reacts elastically to a change in the hydrodynamic characteristics. For $We \ll 1$ the liquid behaves as linearly viscoelastic. The dependence of the properties on the stress-strain state is felt for $We \gtrsim 1$. For $El \gtrsim 1$ propagation of the perturbations is of wave nature. Shear waves reflected from the channel walls pass repeatedly through the gap causing damped vibrations of the velocity and tangential stress at each point. Figure 1 illustrates the development of nonisothermal flows under the impulsive application of a pressure drop for a MBC RES. For $El \gtrsim 1$ two flow stages are clearly separated: wave (duration on the order of $\rho h^2/\eta$) and quasistationary (duration on the order of λ). The inertial forces are inessential in the second stage and a stationary stress distribution is established, the motion is determined by creep during flow development and by retardation during its cessation. After removal of the forces acting on the liquid, it moves in the reverse direction because of unloading of the reversible strains. For $El \gtrsim 1$ the elasticlike behavior of the liquid is magnified with the growth of α . The change in time of the velocity at the point of the maximum of the stationary profile is shown for the MBC RES in Fig. 1b for an impulsive application of a pressure drop. The maximum of the velocity that occurs in the flow wave stage gradually diminishes and vanishes as the pressure gradient grows. The behavior of the liquid becomes more viscouslike as the strain rate rises

(the number We). The magnitude of the elastic return after dumping of the pressure drop increases as the numbers El , α grow and the number We diminishes. The irreversible deformation of the liquid changes to the opposite as these parameters change. Numerical computations for the BC, M, MBC RES showed that the qualitative regularities of nonstationary liquid flows with these RES agree. Only the quantitative characteristics differ.

Experimental investigations of nonsteady flows of polymer liquids for the rise and fall of the pressure are numerous [11] and are mainly qualitative in nature. Results of computations of the change in the mass flow rate in time for the BC, M, MBC RES for We , $El \gtrsim 10$ are qualitatively in agreement with the dependences observed.

Many papers (see [11]) are devoted to a theoretical and experimental investigation of polymer liquid flows subjected to a fluctuating pressure drop

$$\frac{\partial p}{\partial z}(t) = \left(\frac{\partial p}{\partial z} \right)_0 (1 + \xi \sin \omega t), \quad \xi < 1,$$

For liquids with constant properties (Newtonian and linearly viscoelastic) the pressure fluctuations do not alter the mean mass flowrate: $\langle Q \rangle = Q_0$. The relative excess of the mass flow rate $I = (\langle Q \rangle - Q_0)/Q_0 \xi^2$ initially increases with the rise in the pressure gradient and then is reduced. The pressure fluctuations are effective only for low values of the vibrational Reynolds number $Re_\omega = \rho \omega h^2 / \eta_0$. Such a situation is realized in three limit cases [8]: 1) $El \ll 1$, $\omega \lambda \ll Re_\omega \ll 1$; 2) $El \gg 1$, $Re_\omega \ll \omega \lambda \ll 1$; 3) $El \gg 1$, $Re_\omega \ll 1 \ll \omega \lambda$. The elastic properties appear for the third mode. Numerical computations [6, 8] showed that in this case there are two different sections on the frequency dependence $I(\omega)$: the frequency diminishes with growth in the first ($\omega \lambda \lesssim 1$) and increases in the second ($\omega \lambda \gtrsim 1$).

A monotonic reduction in I with the rise in frequency is observed for nonlinear inelastic liquids in tests with fluctuating flows. Theoretical results also lead to the same deduction. Both a diminution and an increase in the relative excess of the flow rate with the rise in frequency [11] is detected in tests with nonlinear elastic liquids. The presence of two branches in the frequency dependence $I(\omega)$, established in [8], shows that such a discrepancy in the results can be related to different test regimes.

The strong dependence of the rheological properties of a liquid on the temperature governs the influence of the thermal mode on the flow. A change in the viscosity and relaxation time across the gap caused by inhomogeneity of the temperature field deforms the velocity and stress profiles, changes the time of flow development and the magnitude of the elastic return. As the parameter $|v|/b$ grows the role of the thermal action increases. The thermal factor appears stronger in the quasistationary stage than in the wave stage. In unsteady thermal and stationary hydrodynamic conditions the elasticity is felt weakly because of the high values of the Prandtl number of polymer liquids. The influence of elasticity is stronger for an impulsive temperature change than for an impulsive change in the thermal flux.

HARD DESTRUCTION OF THE CONFIGURATION

Numerical computations which were performed in [9, 10] permit analysis of the features of nonstationary flows of liquids with hard destruction of the configuration. The simplest RES that does not take account of relaxation processes in the skeleton was taken for the computations in [9, 10]. The structural skeleton is deformed elastically up to the critical stress. It is destroyed instantaneously at a high stress and restored instantaneously at a lower stress. The irreversible deformation for stresses above the critical value is accompanied by destruction of isolated elements of the skeleton. For a shear flow in the channel the tangential stress and the shear velocity are connected by the relationship

$$\frac{1}{\eta} \left[\tau - Y \operatorname{sign} \left(\frac{\partial v_z}{\partial r} \right) \right] 1(|\tau| - Y) + \frac{1}{G} \frac{\partial \tau}{\partial t} 1(Y - |\tau|) = \frac{\partial v_z}{\partial r}. \quad (3)$$

The relative strength of the skeleton is ordinarily $Y/G \sim 1$. For steady flows the RES (3) is equivalent to the Schwedow-Bingham model in a domain with undestroyed structure ($|\tau| < Y$) of the system (1), and (3) is reduced to a hyperbolic equation while it is parabolic in the domain with destroyed structure ($|\tau| > Y$). On the boundary between the zones the tangential stress equals the yield point and the condition of continuity of the velocity is satisfied. In the zone with undestroyed structure the shear wave moves with velocity $c = \sqrt{G/\rho}$, while the shear perturbation with destroyed structure is propagated by diffusion means with the diffusion coefficient η/ρ .

Upon a sudden application of a constant pressure gradient $(\partial p/\partial z)_0$ to the liquid, as long as $|\tau(r, t)| < Y$ the structure is not destroyed and elastic shear waves are propagated across the channel. The maximal value of the tangential stress in the elastic wave is $h(\partial p/\partial z)_0$ for a plane channel. For $h(\partial p/\partial z)_0 < Y$ the structural skeleton fluctuates elastically without being destroyed. The maximal value of the stress is reached at the wall in the stationary flow of a liquid. Stationary flow in a coaxial cylindrical channel is realized for $h\left(\frac{\partial p}{\partial z}\right)_0 > 2Y$. For pressure gradients in the band $Y < h\left(\frac{\partial p}{\partial z}\right)_0 < 2Y$ the structural skeleton in a plane channel is destroyed only near the wall upon the approach of a shear wave. After wave reflection from the wall the stress drops and the structure of the liquid in this region is restored. Doubling of the maximal stress in the wave in comparison with a stationary flow is due to the transition of the liquid kinetic energy into elastic energy. After several cycles of destruction and restoration of the structural skeleton near the wall the velocity field in the channel is rebuilt and a vibrational mode of reversible deformation of the medium is set up without destruction of the structural skeleton. This rebuilding is related to the generation of elastic waves of the boundaries separating the destroyed and undestroyed structure zones. An analytic solution of (1) and (3) is found for a plane channel under the condition $\varepsilon = 1 - Y/h(\partial p/\partial z)_0 \ll 1$, when the size of the destroyed structure zone is considerably less than the channel width. Shown in Fig. 2a is the change in the tangential stress at the wall in time. For $\varepsilon \ll 1$ destruction of the structure near the wall occurs only once. After restoration of the structure the elastic wave has two components: I is the result of a sudden application of a pressure gradient in an elastic medium, and II was formed at the times of the appearance and disappearance of the structure destruction zone. Afterwards the tangential stress at the wall does not exceed the yield point. Numerical computations [9, 10] showed that a qualitatively similar pattern of periodic vibration build-up of the medium without destruction of the structure is observed even for high values of ε , when the steady liquid flow is not realized (Fig. 2b). For $\varepsilon > 0.5$, after several passages of the shear wave across the gap causing stress and velocity fluctuations at each point, stationary velocity and stress profiles are established in the channel (Fig. 2b). The dynamics of the transient is determined by the parameter $E = \sqrt{\rho Gh}/\eta$ which characterizes the relationship between the times of viscous $\rho h^2/\eta$ and elastic h/c wave propagation. After the removal of the pressure drop the liquid moves in the reverse direction because of unloading of the elastic strains of the structural skeleton.

In a steady oscillatory flow subjected to a pressure gradient varying according to the harmonic law $\frac{\partial p}{\partial z} = \left(\frac{\partial p}{\partial z}\right)_0 \sin \omega t$, the influence of elastic properties of the liquid are most essential at moderate frequencies when the inertial forces are small and elastic wave propagation time in the undestroyed structure is of the same order as the period of vibration $Re_\omega = \rho \omega h^2/\eta \lesssim 1$, $St = \rho \omega^2 h^2/G \sim 1$. Reversible strain accumulation in the pressure gradient growth phase and their unloading in its diminution phase increase the amplitude of the fluctuations of the average mass flow rate of the liquid as the relative strength Y/G of the skeleton rises. For low-frequency vibrations ($St \ll 1$) the liquid flow is quasistationary, and its behavior is described completely by the inelastic Schwedov-Bingham model. For high-frequency oscillations ($St \gg 1$) the inertial forces play the main role and the rheological properties of the liquid appear weakly. The role of elasticity of the structure is felt most strongly at moderate frequencies in fluctuating flows, when the Stokes number computed according to the size of the undestroyed structure zone of the main flow h_c is small: $St_h = \rho \omega^2 h_c^2/G \lesssim 1$. This is manifest in the resonance effects of abrupt growth at definite frequencies of the amplitude of fluctuation of the mean velocity, the stresses on the wall, and the width of the undestroyed structure zone. The relative excess of the liquid flow rate in a fluctuating flow diminishes monotonically as the frequency grows.

RELAXATION-CAPABLE MEDIA

Chemically unstable running media can be separated into two kinds by the degree of change in the rheological properties during reaction. The first includes media whose rheological properties vary slightly during transformation and the other includes media with an abrupt change in the rheological properties during reaction. Among the first kind are low-molecular liquids that remain low-molecular even after completion of the chemical reactions.

Among the second are the majority of reaction systems during polymer synthesis and transformation. For liquids of the first kind chemical reactions do not exert direct influence on the nature of the flow. The thermal effects of the reactions and the convective heat transfer conditions are of fundamental value. A change in the viscosity of the liquid because of chemical processes, associated with a change in its temperature, indirectly itself influences the hydrodynamics. For liquids of the second kind chemical reactions decisively influence the motion. The RES of such media should explicitly contain the characteristics of the internal structure and the parameters governing the degree of chemical transformation in contrast to the RES of chemically stable liquids. It must be noted that a softly destroyable structure is formed initially in many liquids during chemical reactions, and then a hardly destroyable structure. Consequently, the characteristics of dynamic processes for chemically stable media with soft and hard destruction of the structure are "bench marks" for reaction-capable systems.

Existing methods of measuring the transformation kinetics of polymer liquids can be separated into three groups:

- 1) intrinsically kinetic, when directly chemical or indirectly physical (by optical, dielectric, etc.) properties can be used to determine the change in time of the content of different reactive substances;
- 2) thermal, when the change in time of the quantity of heat being liberated (DTA, DSK, etc.) is used to assess the behavior of the reactions that occur;
- 3) rheological, in which the effective degree of transformation is determined by the change in time of the mechanical characteristics: viscosity, elastic moduli, etc.

The first method, which is most direct and informative from the chemical aspect, is quite tedious and yields no representation of the macromolecular configuration of the material obtained which depends on the strain mode during the transformation. The second method, most widespread, integrally reflects the progress of a whole set of chemical reactions in the liquid. It also does not yield information about the changes in configuration of the medium. The third method permits an estimate of the rebuilding of the liquid configuration during the transformation. However, it does not characterize the chemical reactions since the change in configuration is not related directly to them. It is expedient to combine the thermal and rheological methods for the simultaneous measurement of the chemical transformation kinetics and the formation of the liquid inner configuration. These same results are required for mathematical modelling of the hydrodynamics and convective heat transfer of chemically unstable liquids. The topologies of the medium configuration being formed, and even the kinetics of the chemical reactions in a number of cases, are determined by the strain mode during the transformations [3, 11, 12]. Consequently, by giving the thermodynamic conditions of chemical reactions and estimating their kinetics by a thermal method, the rheological properties of the liquid must be measured at each stage of the transformation at different temperatures and for different strain conditions. The rheological properties (mainly the viscosity) are measured in existing methodologies only under given thermohydrodynamic transformation conditions. This results in the need to introduce different a priori assumptions relative to the dependence of the rheological properties on the degree of transformation, the strain rate, the temperature, etc., during modelling of chemical and structural transformations in a moving liquid (see [3] and the survey [13]).

Shown in Fig. 3 is the diagram of an experimental installation in which the chemical transformation and rheological properties measurement stages are spatially separated. This permits measurement of the dependence of the effective viscosity of the liquid on the temperature and shear velocity at different stages of the chemical transformation. The reactive fluid for which the heat liberation kinetics is measured first in a microcalorimeter in isothermal regimes at different temperatures is held in a cylindrical tank at a temperature θ_1 for a time t_1 . The corresponding degree of transformation is determined from the microcalorimetry data. The liquid is then squeezed rapidly at a constant load by a piston through a capillary with wall temperature θ_2 . The magnitude of the load, the size of the reservoir and capillary are selected such that 1) the hydraulic resistance of the capillary exceeds by many times the hydraulic resistance of the tank and the entrance to the capillary ($S_1 \gg S_2$); 2) the hydrodynamic entrance section is much less than the capillary length ($l_2 \gg 0.065 \text{ Re } d_2$); 3) the time of liquid efflux from the tank is considerably less than the characteristic time scale of the reaction as determined from tests in the microcalorimeter.

The first and second conditions permit computation of the dependence of the effective viscosity of the liquid on the shear velocity in the capillary by means of the steady velocity of the piston motion under different loads. The third condition eliminates the influence of the chemical transformation on the flow in the capillary. To find the temperature dependence of the viscosity of the liquid, the capillary length should exceed the dimension of the thermal entrance section ($l_2 \gg 0.055 Pe d_2$). Measurement of the dependence of the liquid temperature at the exit from the capillary on its length permits computation of the heat transfer coefficient at the initial section of the channel.

Separation of chemical transformation and thermohydrodynamic measurement stages permits a direct determination of the rheological properties, flow characteristics, and convective heat transfer of chemically unstable liquids at different reaction stages. The possibility of studying the influence of the temperature-time regime of a chemical reaction on the formation of the rheological, hydrodynamic, and heat transfer properties of a liquid is disclosed. The described diagram of the experiment is realized in an installation assembled in the rheophysics laboratory of the Institute of Heat and Mass Transfer of the Academy of Sciences of the Belorussian SSR. The solidification of epoxy resins is investigated thereon.

NOTATION

v_z , axial velocity; $\partial\rho/\partial z$, pressure gradient; τ , tangential stress; R_1, R_2 , channel inner and outer radii; m , memory function; $h = R_2 - R_1$, gap width; λ , greatest relaxation time in the spectrum; θ , temperature; E , activation energy; R_T , gas constant; $a(\theta)$, temperature coefficient of reduction; $\zeta(\alpha) = \sum_{h=1}^{\infty} k^{-\alpha}$, zeta function; α , a relaxation spectrum characteristic; the subscript s refers to the reduction temperature; Q , liquid mass flow rate; Y , yield point; G , elastic modulus of the undestroyed skeleton ($|\tau| < Y$); η , plastic viscosity after destruction of the skeleton ($|\tau| > Y$); $l(x)$, Heaviside unit function; c , elastic wave velocity; S_1, S_2 , cross-sectional areas of the tank and the capillary; l_2, d_2 , length and diameter of the capillary; Re , Reynolds number; Pe , Peclet number.

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