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NONISOTHERMAL FLOW OF CHEMICALLY REACTING MEDIA WITH VISCOSITY DEPENDING ON TEMPERATURE AND PRESSURE

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The article investigates heat exchange and resistance when non-Newtonian, chemically reacting liquids, whose viscosity depends on the flow rate, the temperature, the pressure, and the degree of conversion, flow through flat pipes.

Flow of highly viscous materials (e.g., molten thermosets or rubber mixtures) occurs at high temperatures and pressures, and it is accompanied by considerable dissipative heat liberation, and also by chemical reactions (e.g., the reactions of curing or vulcanization). As a result of these reactions, the viscosity of the materials increases, and this imposes certain limits on the duration of their state of viscous flow. This is of great technological importance because it makes it possible to establish how long the material can be permitted to remain in the working components of the processing equipment.

The authors of [1] and [2] examined questions of nonisothermal flow of chemically reactive media; their solutions are correct only for the flow of Newtonian liquids, and they do not take the effect of the pressure on the physical properties into account.

Below we examine nonisothermal flow of non-Newtonian liquids in a flat pipe on whose outer surface heat exchange with the environment proceeds according to Newton's law (the heat transfer coefficient and the ambient temperature are known). In consequence of the high viscosity of the liquid and its low thermal diffusivity (which is a characteristic feature of many converted polymer materials), the generalized Reynolds criterion does not exceed 10^{-2} whereas the generalized Prandtl number attains 10^5. Therefore, the hydrodynamic initial section is practically lacking, and the speed profile at the pipe inlet may be taken to be fully developed, with no slip on the wall. The temperature distribution at the inlet is taken as uniform over the entire cross section of the channel. As the physical model of heat exchange of the liquid with the inner pipe walls we adopted the model of the thermal boundary layer [3, 4] which assumes that with high Graetz numbers, heat exchange proceeds only in the region near the wall, and that this region increases with increasing distance from the pipe inlet, until the liquid is heated completely over the entire cross section of the channel. It is also assumed that in the material flowing through the pipe, there occurs a chemical reaction of first order. The flow pattern of the liuqid in accordance with the adopted assumptions is shown in Fig. 1.

Assuming that between the stress extra tensor and the strain rate tensor there exists a correlation in the form of Ostwald de Vila's exponential equation, we represent the system of differential equations describing the process of nonisothermal flow of a rheologically complex liquid in the Cartesian system of coordinates in the following manner:

$$\rho\left(v_x\frac{\partial v_x}{\partial x}+v_y\frac{\partial v_x}{\partial y}\right)=-\frac{dP}{dx}+2\frac{\partial}{\partial x}\left[K\left|\frac{\partial v_x}{\partial y}\right|^{n-1}\frac{\partial v_x}{\partial x}\right]+$$
$$+\frac{\partial}{\partial y}\left[K\left|\frac{\partial v_x}{\partial y}\right|^{n}\operatorname{sign}\left(\frac{\partial v_x}{\partial y}\right)+K\left|\frac{\partial v_x}{\partial y}\right|^{n-1}\frac{\partial v_y}{\partial x}\right], \tag{1}$$

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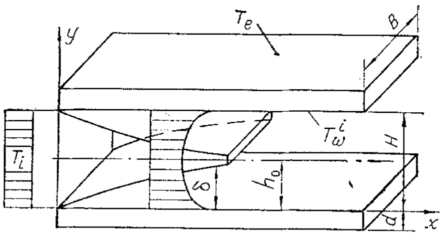

Fig. 1. Flow pattern of the liquid in the pipe.

$$\frac{\partial v_x}{\partial x} + \frac{\partial v_y}{\partial y} = 0, \tag{2}$$

$$v_x \frac{\partial T}{\partial x} + v_y \frac{\partial T}{\partial y} = a_f \frac{\partial^2 T}{\partial y^2} + \frac{K}{\rho c_p} \left| \frac{\partial v_x}{\partial y} \right|^{n+1}, \tag{3}$$

$$\int_0^\delta v_x dy + \int_\delta^{h_0} v_x dy = \bar{v} h_0. \tag{4}$$

The consistency constant in Eqs. (1) and (3) depends on the temperature T, the hydrostatic pressure p, and the degree to which the reaction C proceeds, in accordance with the equation

$$K = K_0 F(C) \exp(-b|T - T_0|) \exp(s_1 p). \tag{5}$$

The expression for the consistency constant in the form of relation (5) considerably complicates the mathematical model of the process of flow of a reacting material, therefore in practice various criteria are widely used that make it possible to calculate the time of the induction period of the reaction in the course of which the material is in the state of viscous flow. The following expression [5] may serve as one of these criteria:

$$J = \int_0^{t_i} \exp\left[-\frac{U}{RT(t)}\right] dt \cdot \tag{6}$$

To determine the length of the induction period of the reaction with the aid of Eq. (6) it suffices to know the values of U and J* which are determined from two experiments in isothermal regime at the temperatures T_1 and T_2 by the following formulas:

$$U = [R \ln(t_{i1}/t_{i2})] \Big/ \left[\frac{1}{T_1} - \frac{1}{T_2}\right], \tag{7}$$

$$J^* = t_{i1} \exp\left(-\frac{U}{RT_1}\right). \tag{8}$$

The inequality $J \leqslant J^*$ corresponds to the condition of maintaining the state of viscous flow of the material.

The use of criterion (6) makes it possible to simplify expression (5) because during the induction period of the reaction it may be adopted that F(C) = 1.

In accordance with the adopted assumptions the boundary conditions have the form

$$x \geqslant 0,\ y = 0,\ v_x = v_y = 0,\ \lambda_f \left(\frac{\partial T}{\partial y}\right) = K^* (T_W^i - T_e), \tag{9}$$

$$x \geqslant 0,\ h_0 \geqslant y \geqslant \delta,\ T = T_i,\ \frac{\partial T}{\partial y} = 0, \tag{10}$$

$$x \geqslant 0,\ y = h_0,\ \frac{\partial v_x}{\partial y} = 0, \tag{11}$$

where $K^* = \lambda_s \Big/ \left(d + \frac{\lambda_s}{\alpha_e}\right)$.

We introduce the following dimensionless variables:

$$X = x/h_0,\ Y = y/h_0,\ \Delta = \delta/h_0,\ Z = a_f x/\bar{v} H^2,\ m = 1/n,\ V_X = v_x/\bar{v},$$

$$\theta = (T - T_e)/(T_i - T_e),\ \varphi = b(T_i - T_e),\ \psi = m\varphi,\ \beta = 2/\mathrm{Bi},\ \mathrm{Bi} = K^* h_0/\lambda_s,$$

$$\mathrm{Br} = [K_e \bar{v}^{n+1} / \lambda_f (T_i - T_e)\, h_0^{n-1}, \quad \mathrm{Pr}^* = \frac{K_e}{12\rho a_f}\left(\frac{4n+2}{n}\right)^n \left(\frac{\bar{v}}{H}\right)^{n-1}.$$

In solving the stated problem, we have to replace the exponential curve describing the dependence K = K(T) in expression (5) by an approximating polynomial of fourth degree, using Chebyshev's orthogonal polynomials

$$K = K_e \left[1 + \sum_{i=1}^{4} a_i (\theta\psi)^i\right] \exp(s_1 p). \tag{12}$$

If we solve the system of equations (1)-(3), (12) with a view to the boundary conditions (9)-(11) by the approximate analytical method based on averaging the inertial and convective terms of the equations over the height of the channel and the thickness of the thermal boundary layer, respectively, we can obtain the expressions for the profile of the speed V_X and of the temperature θ in any cross section of the channel and for its hydraulic resistance ΔP which have the following form [6]:

$$V_X = N \sum_{i=1}^{9} \frac{M_i}{m+i}\left[(1-Y)^{m+i} - 1\right] \quad (\Delta \geqslant Y \geqslant 0), \tag{13}$$

$$V_X = \frac{N\chi_0^m}{m+1}\left[(1-\Delta)^{m+1} - (1-Y)^{m+1}\right] + \\ + N \sum_{i=1}^{9} \frac{M_i}{m+i}\left[(1-\Delta)^{m+1} - 1\right] \quad (1 \geqslant Y \geqslant \Delta), \tag{14}$$

$$\theta = -\frac{Y^2}{\Delta(\beta+\Delta)} + \frac{2Y}{\beta+\Delta} + \frac{\beta}{\beta+\Delta} = A_1 Y^2 + A_2 Y + A_3, \tag{15}$$

$$\Delta P = 6\rho\bar{v}^2 \left(\frac{n}{2n+1}\right) \mathrm{Pr}^* \int_0^\Delta \frac{\exp(-A_3\varphi)\left(-N\sum_{i=1}^{9} M_i\right)^n (F_1+F_2)}{\mathrm{Br}\, F_3 \exp(s_1 p) - 2/(\Delta+\beta)}\, d\Delta, \tag{16}$$

$$F_1 = N \sum_{i=1}^{9} \frac{M_i}{m+i}\left[\frac{(B_1+B_2+B_3)(1-\varepsilon^{s+i})}{s+i} - \frac{(2B_1+B_2)(1-\varepsilon^{r+i})}{r+i} - \right. \\ \left. - \frac{B_1(1-\varepsilon^{q+i})}{q+i}\right] - \left(\frac{B_1\Delta^3}{3} + \frac{B_2\Delta^2}{2} + B_3\Delta\right),$$

$$F_3 = N \sum_{i=1}^{9} \frac{M_i}{m+i}\left(\varepsilon^{m+i} - 1\right),$$

$$F_2 = N \sum_{i=1}^{9} \frac{NGM_i + D_i}{m+i}\left[\frac{\Delta^2(4A_1\Delta + 3A_3)}{6} + \frac{(2A_1+A_2)(1-\varepsilon^{r+i})}{(s+i)(r+i)} - \frac{2A_1(1-\varepsilon^{q+i})}{(s+i)(q+i)} - \frac{\Delta(A_1\Delta + A_2)}{s+i}\right],$$

$$N = \left[\frac{\chi_0 \varepsilon^r}{m+2} - \sum_{i=1}^{9} \frac{M_i}{s+i}\left(1 - \varepsilon^{s+i}\right)\right]^{-1},$$

$$G = \chi_0^m \varepsilon^s + \sum_{i=1}^{9} M_i \varepsilon^{m+i} + \sum_{i=1}^{9} \frac{D_i}{s+i}\left(1 - \varepsilon^{s+i}\right),$$

$$B_1 = \frac{\beta + 2\Delta}{\Delta^2(\beta+2\Delta)^2}, \quad B_2 = -\frac{2}{(\beta+\Delta)^2}, \quad B_3 = -\frac{\beta}{(\beta+\Delta)^2};$$

$$M_i = M_i(\psi, \Delta); \; D_i = \frac{dM_i}{d\Delta}, \; \varepsilon = 1 - \Delta, \; s = m+1, \; r = m+2, \; q = m+3.$$

The dependence of the reduced length on the thickness of the thermal boundary layer is found from the equation

$$Z = 0.25 \int_0^\Delta \frac{F_1 + F_2}{\mathrm{Br}\, F_3 \exp(s_1 p) - 2/(\beta+\Delta)}\, d\Delta.$$

The obtained expressions make it possible to describe the development of the speed profile as well as of the temperature profile along the pipe, and consequently, to determine

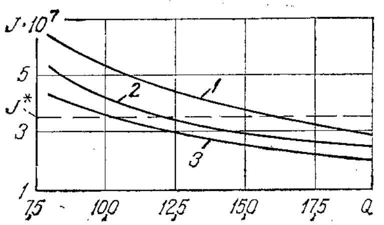

Fig. 2

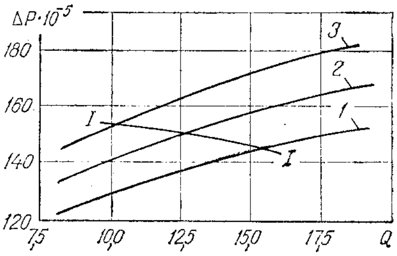

Fig. 3

Fig. 2. Dependence of the criterion J* on the flow rate Q with different initial temperatures of the liquid (Bi = 17): 1) T_i = 80°C; 2) 100; 3) 120; J*, sec; Q, cm^3/sec.

Fig. 3. Determination of the range of permissible values of the thermal and hydrodynamic flow parameters of a chemically reactive liquid: 1) T_i = 80°C; 2) 100; 3) 120; ΔP, N/m^2.

the temperature–time regime of affecting an arbitrary microvolume of material flowing in the pipe. If we know the dependence $T = T(t)$ for an arbitrary elementary volume of the medium, we can then calculate the value of the integral (6) for it. By comparing the obtained value of the criterion with its permissible limit value calculated by expression (7), we determine the possibility of the given microvolume of the medium remaining in the state of viscous flow.

As a practical application of the obtained results we will examine the following problem. A chemically reactive material, whose consistency constant depends on temperature and pressure in accordance with Eq. (5), is fed into a cavity with volume V through a flat pipe whose length is L = 150 mm and height H = 3 mm. The flowing material has the following rheological and thermophysical characteristics:

$$K_e\big|_{T=373\,^\circ\mathrm{K}} = 55\cdot 10^3\,\frac{\mathrm{N\cdot sec^n}}{\mathrm{m^2}},\ n = 0.26,\ b = 0.0076\ \mathrm{deg}^{-1},$$

$$\rho = 1150\,\mathrm{kg/m^3},\ \lambda_f = 0.26\,\mathrm{W/m\cdot deg},\quad U = 60\ \mathrm{kJ/mole},$$

$$J^* = 3.5\cdot 10^{-7}\,\mathrm{sec},\ s_1 = 0.6\cdot 10^{-8}\ \mathrm{Pa}^{-1}.$$

It is indispensable to establish the region of values of flow rate Q, initial temperature T_i, and pressure gradient ΔP along the channel for which it may be asserted that no particles of the medium converted to the solid state as a result of a chemical reaction had reached the cavity. Here we determine the time of filling the cavity $t_{fil} = V/Q$, then the time of dwelling of elementary volumes of the medium, situated at different distances from the inner pipe wall, in the pipe $t_{Lp} = \sum_{j=1}^{k} \frac{\Delta l_j}{v_{avj}}$, where k is the number of sections of the channel within which the temperature and speed of an elementary volume of the medium are considered constant and are calculated by the formulas

$$\Delta l_j = x_{j+1} - x_j,\ v_{avj} = \frac{v_{j+1} + v_j}{2},\quad T_{av} = \frac{T_{j+1} + T_j}{2},$$

v_{j+1}, T_{j+1}, v_j, T_j are the magnitudes of the longitudinal component of speed and temperature at the end x_{j+1} and at the beginning x_j of each section, calculated by the above formulas.

Then for an elementary volume of material, whose dwelling time in the pipe is equal to the time of filling the cavity, we determine by a numerical method the magnitude of the integral (5) which is compared with its limit value J*. If J does not exceed J*, the cavity is filled with material that is exclusively in the state of viscous flow.

Figure 2 shows the results of numerical calculations by the presented method. Above the dashed line, which corresponds to the limit value of the criterion J*, there is the region of impermissible values of volume velocity of flow of the medium and of its initial temperature at the inlet to the channel because there it is possible that cured particles of material reach the cavity.

Using the data of Fig. 2, and also taking Eq. (16) into account, we can plot the volume–flow rate characteristic of the pipe–cavity system taking into account the rheokinetics of flow of the material as well as the fact that it is not isothermal. An example of plotting such a characteristic is presented in Fig. 3. To the right of the line I–I there is the range of permissible values of flow rate and pressure gradient along the pipe whose joint realization ensures that only material in the state of viscous flow reaches the cavity.

The obtained results may find application in engineering calculations of the optimum operating parameters of equipment converting chemically reactive materials.

NOTATION

K_0, K_e, values of the consistency constant at atmospheric pressure and at the temperatures T_0 and T_e, respectively; n, flow index; b, s_1, temperature and baric coefficients of viscosity, respectively; λ_f, a_f, ρ, $\bar{v}$, thermal conductivity, thermal diffusivity, density, and mean flow rate of the liquid, respectively; λ_s, thermal conductivity of the material of the pipe; K^*, heat-transfer coefficient from the inner wall surface of the pipe to the environment; a_i, coefficients of the Fourier expansion of the exponential curve into a series with respect to the orthogonal Chebyshev polynomials; α_e, heat-transfer coefficients from the outer pipe surface to the environment with temperature T_e; U, activation energy of the chemical reaction; F(C), function taking into account the effect of the degree of the course of the reaction on the viscosity of the liquid; t_i, t_{i1}, t_{i2}, time of the induction period of the reaction; R, universal gas constant.

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