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INFLUENCE OF THERMAL EFFECTS ON HYDRODYNAMIC STABILITY OF A POLYMERIZATION FRONT

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Two features are usually distinguished in investigating the stability of fronts of chemical reactions: diffusive-thermal and hydrodynamic stability [1]. The diffusive-thermal stability is analyzed under the assumption that perturbations of the front shape are not accompanied by perturbations of hydrodynamic fields in its vicinity [2], while the hydrodynamic stability is analyzed under the assumption that, on the contrary, no perturbations of the concentration and thermal fields are generated [3]. The last assumption is justified if the front width δ is negligibly small in comparison with the perturbation wavelength λ (the zeroth approximation in the small parameter $\varepsilon = \delta/\lambda$). For short-wave perturbations ($\delta \sim \lambda$) it is necessary to take into account the simultaneous occurrence of diffusive-thermal and hydrodynamic processes. This account was carried out most consistently in [4], as applied to a planar front in the gas phase. The analysis is restricted to the first approximation in ε, using an asymptotic method of solving singular perturbation problems with a surface discontinuity, as developed in [5].

The problem of creating a continuous technological process of obtaining polymer materials on the basis of the polymerization frontal effect [6] leads to the statement of stability problems of cylindrical and spherical polymerization fronts in radial flows. An important polymerization effect is a strong increase in medium viscosity; therefore, the hydrodynamic stability analyzed in [7, 8] is of basic interest in the given case. The thermal stability investigated in [9] for a cylindrical front is unrelated to any new physical effects relative to [2].

In the present study we consider the stability of a stationary cylindrical front in a radial flow with account of the mutual effect of thermal and hydrodynamic effects. The analysis is carried out within the first approximation in ε by the method of matched asymptotic expansions, in which case, unlike [4], one uses the convenient method of transition to a moving curvilinear natural coordinate system attached to the front [10]. First order corrections in ε to the solution obtained in [7] are found, and their effect on front stability is analyzed.

1. We restrict ourselves to the case of angular perturbations of a cylindrical front, in which case the problem can be considered as planar in the cross section normal to the front axis. Let the closed front F be located between two penetrable coaxial cylindrical surfaces S_- and S_+, and let it propagate in the direction of the normal **n** toward the flow (the internal supply of the medium), with the regions Ω_- and Ω_+ filled by the original mixture and by the final product (see Fig. 1). Following [2, 4], the problem is solved in the zeroth approximation in the small Frank-Kamenetskii parameter $\beta = \bar{R}T_r/E$ ($\bar{R}$ is the universal

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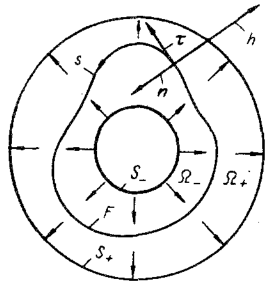

Fig. 1

gas constant, T_r is the temperature in the reaction zone, and E is the effective activation energy of the reaction). In this case the reaction zone is treated as a discontinuity surface of conversion and, consequently, viscosity. In the following it is identified with the front. The temperature dependence of the viscosity is not taken into account, while the other thermophysical parameters of the medium are assumed constant.

The flow in the regions Ω_- and Ω_+ is described by the Stokes and continuity equations [7, 8]:

$$\nabla p_{\pm} = \mu_{\pm}\nabla^2\mathbf{V}_{\pm},\ \nabla\cdot\mathbf{V}_{\pm} = 0 \tag{1.1}$$

(p is the pressure, **V** is the velocity vector, μ is the dynamic viscosity coefficient, and ∇ is the vector differential operator). We note that the Reynolds number $Re_{\pm} = R_0U/\nu_{\pm}$, constructed from the radius of the unperturbed front R_0, its normal propagation velocity with respect to the medium U, and the kinematic viscosity coefficient of the medium ν, is small only in the region Ω_+, filled by the highly viscous product, while in the region Ω_- it can be comparable to unity. As shown in [7, 8], however, in the latter case the motion of the perturbed front becomes dependent on the low-viscosity original mixture, and the results obtained by using Eq. (1.1) remain valid.

The continuity of the velocity and stress vectors is satisfied at the front F due to the incompressibility of the medium and the condition of flow "adhesion"

$$\mathbf{V}_- = \mathbf{V}_+,\ \mathbf{P}_{n-} = \mathbf{P}_{n+}. \tag{1.2}$$

The hydraulic resistances of the boundary surfaces S_- and S_+ are assumed linear, so that the following conditions are satisfied on them

$$p_{\pm} - \bar{p}_{\pm} = \pm\sigma_{\pm}V_{n\pm},\ \mathbf{V}_{\tau\pm} = 0, \tag{1.3}$$

where $\bar{p}_{\pm}$ is the pressure outside the reaction volume (near the boundaries $S_{\pm}$, respectively), and $\sigma_{\pm}$ are coefficients, which must be called local hydraulic resistances of the surface boundaries.

Equations (1.1) are also supplemented by the initial front shape, but the statement of the problem remains nonclosed: to close it, it is necessary to assign the condition of front motion with respect to the medium. With this purpose we turn to analyzing the internal diffusive-thermal structure of the front.

As is well known, in the gas phase all transport coefficients coincide in order of magnitude (D ~ κ ~ ν, where D is the diffusion coefficient, and κ is the temperature conductivity), in which case the transport processes are concentrated and interact in the preheating zone ahead of a front of thickness of order δ ~ κ/U.

In the condensed phase, as a rule, D ≪ κ ≪ ν and the thickness of the influence zone of diffusion effects $\delta_D \sim D/U$ is substantially smaller, while the thickness of the influence zone of viscous effects $\delta_{\nu\pm} \sim \nu_{\pm}/U$ is substantially larger than the thickness of the preheating zone δ. In the case of polymerization δ_D is not only much smaller than δ, but also smaller than the thickness of the reaction zone $\delta_r \sim \beta\delta$, in which case diffusion can be neglected [11]. Due to the sharp increase in medium viscosity at the front the δ_ν values are different for the regions Ω_- and Ω_+. In particular, $\delta_{\nu+}$ substantially exceeds not only δ, but also the radius of the unperturbed front R_0, playing the role of characteristic geometric scale of the problem ($Re_+ = R_0/\delta_{\nu+} = R_0U/\nu_+ \ll 1$). The quantity $\delta_{\nu-}$ is larger than or comparable to R_0 ($Re_- = R_0/\delta_{\nu-} = R_0U/\nu_- \lesssim 1$), and in both cases viscous effects are not concentrated in a narrow region, while they propagate over the whole flow region. If the perturbation wavelength is of scale R_0, then $\delta_{\nu-}$ is also the inhomogeneity scale of the velocity field.

The inhomogeneity in the temperature field T, generated at the boundary S_- and on the front F, drifts downward along the flow; the spatial scale of this inhomogeneity in all directions is R_0 (the external scale). At the same time the inhomogeneity, generated at the front F and the boundary S_+, is transported by the thermal conductivity upward along the flow and is "pressed" by the flow to the corresponding surfaces, forming in their vicinity temperature boundary layers with spatial scales R_0 in the tangential direction and δ in the normal direction (the internal scale). These two spatial inhomogeneity scales of the temperature field generate two time scales: the fast $[t]_1 = \delta/U \sim \kappa/U^2$, determining the front propagation time at a distance of the order of its thickness, and the slow $[t]_2 = R_0/U$, related to changes in the front shape.

Since the temperature distribution in the internal scale appears attached to the front, for its analysis it is convenient to use the moving curvilinear system of natural coordinates (h, s) attached to the front (see Fig. 1). In this system the nonstationarity is obviously related to a change in the front shape; therefore, the single time scale $[t]_2 = R_0/U$ is retained.

The scales of the remaining variables are selected according to the structure of the temperature field: $[h] = \delta$, $[R] = R_0$, $[V_h] = [V_s] = U$, $[T] = Q/c$ (R is the local radius of curvature of the front F, Q is the thermal effect of the reaction, and c is the heat capacity of the medium).

Not dwelling on the quite unwieldy technique of transition to the coordinates (h, s), we provide the equation of thermal conductivity in these coordinates in the dimensionless variables corresponding to the scales introduced (with the same notations as their dimensional analogs):

$$\varepsilon\frac{\partial T_{i\pm}}{\partial t} + (W+V_h)\frac{\partial T_{i\pm}}{\partial h} + \varepsilon\left[V_s - \frac{\varepsilon}{1+\varepsilon h/R}\frac{\partial}{\partial s}\left(\mathbf{g}\cdot\frac{\partial\mathbf{f}}{\partial t}\right)\right]\frac{\partial T_{i\pm}}{\partial s} = \frac{1}{1+\varepsilon h/R}\frac{\partial}{\partial h}\left[(1+\varepsilon h/R)\frac{\partial T_{i\pm}}{\partial h}\right] + \frac{\varepsilon^2}{(1+\varepsilon h/R)^2}\frac{\partial^2 T_{i\pm}}{\partial s^2}. \quad (1.4)$$

Here $T_{i\pm} = T_{i\pm}(h, s, t)$ are the temperature distributions in the internal scale, $\mathbf{f} = \mathbf{f}(s, t)$ is a vector representing the "natural" front shape, $\mathbf{g} = \mathbf{f} - \mathbf{r}$ is the vector joining the arbitrary point given by the radius-vector $\mathbf{r}$ and the "current" point of the front given by the vector $\mathbf{f}$, $W = (\partial\mathbf{f}/\partial t)\cdot\mathbf{n}$ is the normal velocity of the front relative to the resting coordinate system, and $\varepsilon = \delta/R_0 \sim \kappa/UR_0 = 1/Pe$ is a small parameter, having the meaning of the reciprocal Peclet number.

The temperature and total energy flux are continuous in the internal scale at the front F, while the conductive flux undergoes a jump, whose magnitude is found from the temperature distribution in the reaction zone in the zeroth approximation in β [2, 4]. In the case of the simplest gross kinetic process with a zeroth-order Arrhenius reaction we have

$$T_{i-} = T_{i+} = T_r \ (h = 0); \quad (1.5)$$

$$\partial T_{i-}/\partial h - \partial T_{i+}/\partial h = W + V_h \ (h = 0); \quad (1.6)$$

$$\partial T_{i-}/\partial h - \partial T_{i+}/\partial h = K\exp(-A/2T_r) \ (h = 0) \quad (1.7)$$

($K = (k\kappa)^{1/2}/U$, $A = Ec/\bar{R}Q$, and k is a preexponent). At large distances from the temperature front there is a smooth departure by a constant value

$$\partial T_{i\pm}/\partial h = 0 \ (h = \pm\infty). \quad (1.8)$$

In the external scale the front is considered as a surface with a temperature jump with the nonstationarity, as usual, related only to a change in the front shape and conservation of the single time scale $[t]_2 = R_0/U$. Besides, there is no scale difference here between the stresses normal and tangential to the front, so that there is a single spatial scale $[\mathbf{r}] = R_0$. The scales of the remaining quantities are not changed, in which case the heat conduction equation acquires the following form in the corresponding dimensionless variables

$$\partial T_{e\pm}/\partial t + \mathbf{V}_\pm \cdot \nabla T_{e\pm} = \varepsilon \nabla^2 T_{e\pm} \tag{1.9}$$

($T_{e\pm} = T_{e\pm}(\mathbf{r}, t)$ is the temperature distribution in the external scale).

At the boundary S_- we impose the following isothermal input in the reaction volume

$$T_{e-}|_{S_-} = T_0, \tag{1.10}$$

while a condition at the boundary S_+ is not specified.*

The temperatures in the internal and external scales coincide at the front F:

$$T_{e\pm}|_F = T_{i\pm}|_{h=\pm\infty}. \tag{1.11}$$

Equations (1.4), (1.9) with boundary conditions (1.5)-(1.8), (1.10), (1.11) make it possible to find the temperature distributions in the internal and external scales and the front velocity. This is done within the zeroth and first approximations in ε.

2. We expand the dependent variables in series in ε:

$$T_{i\pm} = T^0_{i\pm} + \varepsilon T^1_{i\pm} + \ldots,\ T_{e\pm} = T^0_{e\pm} + \varepsilon T^1_{e\pm} + \ldots,\ W = W^0 + \varepsilon W^1 + \ldots \tag{2.1}$$

Besides, in the external scale we expand the velocity vector in a Taylor series in the coordinate h in the vicinity of the front. Since the spatial inhomogeneity scale of the velocity field in all directions is R_0, in dimensionless variables we have

$$\mathbf{V}_\pm = \mathbf{V}_\pm\Big|_{h=0} + \varepsilon \frac{\partial \mathbf{V}_\pm}{\partial h}\Big|_{h=0} h + \ldots \tag{2.2}$$

Substituting these expansions into (1.1)-(1.11) and retaining first order terms in ε, we obtain a statement of the problem within the corresponding approximations. Within the zeroth approximation

$$\partial^2 T^0_{i\pm}/\partial h^2 - U^0 \partial T^0_{i\pm}/\partial h = 0; \tag{2.3}$$

$$\partial T^0_{i\pm}/\partial h = 0 \ (h = \pm\infty); \tag{2.4}$$

$$T^0_{i-} = T^0_{i+} = T^0_r,\ \partial T^0_{i-}/\partial h - \partial T^0_{i+}/\partial h = U^0, \tag{2.5}$$

$$\partial T^0_{i-}/\partial h - \partial T^0_{i+}/\partial h = K \exp\left(-A/2T^0_r\right)\ (h = 0);$$

$$\partial T^0_{e\pm}/\partial t - \mathbf{V}_\pm \cdot \nabla T^0_{e\pm} = 0; \tag{2.6}$$

$$T^0_{e-}|_{S_-} = T_0; \tag{2.7}$$

$$T^0_{e\pm}|_F = T^0_{i\pm}|_{h=\pm\infty} \tag{2.8}$$

($U^0 = W^0 + V_h|_{h=0}$ is the normal front velocity with respect to the medium within the zeroth approximation).

From (2.6), (2.7) we find $T_{e-}{}^0 = T_0$, i.e., the flow is isothermal in the region Ω_- in the external scale within the zeroth approximation. From (2.8) we have $T_{i-}{}^0|_{h=-\infty} = T_0$; integrating (2.3) with account of the last equality, as well as conditions (2.4) and the first two of conditions (2.5), the temperature distribution is obtained in the internal scale within the zeroth approximation:

$$T^0_{i-} = T_0 + \left(T^0_r - T_0\right) e^{U^0 h},\ T^0_{i+} = T^0_r = \mathrm{const}\,(h). \tag{2.9}$$

*We note that assigning on the boundary S_- conditions of the second or third kind assumes that this surface is at the same time a device implementing reaction supply to the medium and heat transfer in the direction normal to it. Such a combination of technological functions is practically quite complicated; therefore, the likelihood of controlling the front stability by means of heat exchange at the outlet in the reaction volume is considered to be problematic. This group of problems is not considered in the present study.

Substituting the last equality into the remainder of conditions (2.5), we obtain

$$T_r^0 = T_0 + 1 = T_m,\ U^0 = K\exp(-A/2T_m) = 1, \tag{2.10}$$

where T_m is the temperature of adiabatic reaction transmission (the last equality in (2.10) follows from the expression for the velocity U of a planar front in a resting medium with the use of simple kinetics [1]). Relations (2.9), (2.10) show that within the zeroth approximation the "internal" thermal front structure is not distorted with respect to a planar front in a resting medium.

From (2.6), (2.8) we find $T_{e+}{}^0 = T_0 + 1 = T_m$, i.e., the flow in the region Ω_+ is also isothermal in the external scale within the zeroth approximation.

The condition at the boundary S_+ determines the formation of a temperature boundary layer near this surface. Outside this layer the temperature field is determined by convective heat transport from regions located above the flow, therefore no condition was required on S_+.

Taking into account the solution found within the zeroth approximation, within the first approximation the statement of the problem looks as follows:

$$\frac{\partial^2 T_{i\pm}^1}{\partial h^2} - U^0 \frac{\partial T_{i\pm}^1}{\partial h} = \left(h \frac{\partial V_h}{\partial h}\bigg|_{h=0} + U^1 - \frac{1}{R}\right) e^h; \tag{2.11}$$

$$\partial T_{i\pm}^1/\partial h = 0 \quad (h = \pm\infty); \tag{2.12}$$

$$T_{i-}^1 = T_{i+}^1 = T_r^1,\ \partial T_{i-}^1/\partial h - \partial T_{i+}^1/\partial h = U^1, \tag{2.13}$$

$$\partial T_{i-}^1/\partial h - \partial T_{i+}^1/\partial h = ZT_r^1 \quad (h = 0);$$

$$\partial T_{e\pm}^1/\partial t + \mathbf{V}_\pm \cdot \nabla T_{e\pm}^1 = 0; \tag{2.14}$$

$$T_{e-}^1|_{S_-} = 0; \tag{2.15}$$

$$T_{e\pm}^1|_F = T_{i\pm}^1|_{h=\pm\infty}. \tag{2.16}$$

Here $U^1 = W^1$ is the first order correction to the normal front velocity, and $Z = A/2T_m^2$ is the Zel'dovich number (in dimensional variables $Z = E(T_m - T_0)2\bar{R}T_m^2$).

Following some transformations we obtain the problem solution within the first approximation

$$T_{i-}^1 = \left[\frac{1}{2}\frac{\partial V_h}{\partial h}\bigg|_{h=0} h^2 + \left(U^1 - \frac{1}{R} - \frac{\partial V_h}{\partial h}\bigg|_{h=0}\right) h + T_r^1\right] e^h, \tag{2.17}$$

$$T_{i+}^1 = T_r^1 = \mathrm{const}\,(h);$$

$$T_r^1 = 1/R + (\partial V_h/\partial h)_{h=0},\ U^1 = ZT_r^1. \tag{2.18}$$

Besides, $T_{e-}{}^1 = 0$, while the field $T_{e+}{}^1$ is found from Eq. (2.14) with the boundary condition $T_{e+}{}^1|_F = T_r{}^1$. Thus, in the external scale the flow remains isothermal in the region Ω_- within the first approximation, while corrections for the temperature distributions appear in the region Ω_+. Similar corrections are generated for the temperature distribution in the internal scale and for the front velocity.

Using the last of equalities (2.10), (2.18), we find the normal front velocity relative to a resting coordinate system. Turning to dimensional variables, we obtain, respectively, within the zeroth and first approximations

$$W = V_n|_F + U; \tag{2.19}$$

$$W = V_n|_F + U + \varkappa Z\left(\frac{1}{R} + \frac{\partial V_n}{\partial n}\bigg|_F \frac{1}{U}\right) \tag{2.20}$$

(V_n is the projection of the local flow velocity on the normal to the front, and $\partial/\partial n$ is the derivative in this direction).

Relation (2.20) shows that the first order corrections for the normal front velocity are related to its curvature and to the inhomogeneity in the velocity field, and they coincide with the well-known Markstein corrections and with the "stretch-effect" correction [1, 13]. Due to the quasistationarity of the Stokes and continuity equations (1.1) these corrections are completely determined by the front shape at a given moment of time (if inertial terms had been included in the equations of motion of the medium, the "stretch-effect" correction, containing the derivative $\partial V_n/\partial n$, would have also depended on the previous history of the front motion).

Conditions (2.19) or (2.20) close the statement of the problem of front stability (1.1)-(1.3) within the zeroth and first approximations, respectively. The zeroth approximation was investigated in [7], while here we investigate the effect of first-order corrections.

3. We consider initially stationary front states and their stability to perturbations of its radius, not accompanied by perturbations of its cylindrical shape (the zeroth mode). Taking into account that $W = -dR/dt$, $V_n|_F = -V_r(R)$, $(\partial V_n/\partial n)_F = (\partial V_r/\partial r)_{r=R} = -V_r(R)/R$, we rewrite (2.19):

$$dR/dt = [V_r(R) - U](1 + \varkappa Z/UR). \tag{3.1}$$

From the last relation we obtain that in the stationary state the front radius is determined by the equation

$$V_r(R_0) = U \tag{3.2}$$

and thus does not change with respect to the zeroth approximation. This is related to the fact that in the given case the effects related to front distortion and to velocity field inhomogeneity compensate each other.

Putting $R = R_0 + R'$ (R' is a small perturbation) and linearizing (3.1) in R', we find $dR'/dt = -\omega R'$, $R' \sim \exp(-\omega t)$, where

$$\omega = \omega^0 + \varepsilon\omega^1 + \ldots, \quad \omega^0 = [dV_r(R)/dR]_{R=R_0}, \quad \omega^1 = Z\omega^0 \tag{3.3}$$

(ω is the perturbation increment). From the last relation we see that the first-order correction increases the absolute value of the increment and does not change its sign; therefore, all conclusions concerning the front stability obtained within the zeroth approximation in ε [7] are only enhanced within the first approximation. In particular, in feeding the medium with a constant flow rate q ($V_r(R) = q/2\pi R$, $\omega^0 = 2\pi U^2/q$) for a single stable stationary state we have $0 < \omega^0 < \omega$, so that within the first approximation in ε the perturbations decay more quickly than in the zeroth approximation.

In the case of external supply to the medium (in Fig. 1 the front F propagates in a direction opposite to the normal $\mathbf{n}$, but, as previously, toward the flow, original mixture, and final product filling the regions Ω_+ and Ω_-, respectively) similar calculations lead to a first-order correction of opposite sign to the increment $\omega^1 = -Z\omega^0$. In this case the absolute value of the increment decreases, and there is a tendency to changing the conclusion concerning front stability. In particular, when the medium is supplied with constant flux ($V_r(R) = -q/2\pi R$, $\omega^0 = -2\pi U^2/q$) the single unstable stationary state has a tendency to stabilization ($\omega^0 < \omega < 0$). In reality, however, the change in sign of ω can occur only for small radii of curvature of the front, comparable with its thickness ($R_0 \sim \delta$). For these R_0 values the frontal regime of reaction transmission is practically degenerate, and the first order approximation in ε becomes operative. Nevertheless, extrapolating the results obtained to the small R_0 region, we find that in the case of external supply to the medium with constant flux the stationary state of the front becomes stable for

$$R_0 = R_0^* = Z\delta. \tag{3.4}$$

The last relation must be considered as orientational, and its accuracy is enhanced for strongly activated reactions, when $Z \gg 1$ and $R_0 \gg \delta$.

4. Consider perturbations of arbitrary shape

$$\mathbf{V}_\pm = \mathbf{V}_{0\pm} + \mathbf{V}'_\pm, \; p = p_{0\pm} + p'_\pm, \; f = f_0 + f', \; \mathbf{n} = \mathbf{n}_0 + \mathbf{n}', \; W = W_0 + W', \tag{4.1}$$

where the subscript 0 denotes the stationary value of quantities, the prime denotes perturbations, and f is a function giving the front shape in a cylindrical coordinate system: $r = f(\varphi, t)$ ($f_0 = R_0 = \text{const}\ (\varphi, t)$). Substituting (4.1) into (1.1)-(1.3) and (2.19) or (2.20), we linearize the nonlinear boundary conditions at the front in small perturbations. In this case relation (2.19), corresponding to the zeroth approximation in ε, acquires the form [7]

$$\partial f'/\partial t + (U/R_0) f' - V_r' = 0. \tag{4.2}$$

We dwell on the linearization of first order corrections to the front velocity. In a polar coordinate system we have for the front radius

$$1/R = [f^2 + 2(\partial f/\partial\varphi)^2 - f(\partial^2 f/\partial\varphi^2)]/[f^2 + (\partial f/\partial\varphi)^2]^{3/2}.$$

Substituting expansion (4.1) for f into this expression, following linearization we find

$$1/R = 1/R_0 - f'/R_0^2 - (\partial^2 f'/\partial\varphi^2)/R_0^2 + \ldots \tag{4.3}$$

To linearize the second correction it is convenient to use the following representation. From the continuity equations in the natural coordinates (h, s) we obtain at the front

$$\partial V_{h-}/\partial h + V_{h-}/R + \partial V_{s-}/\partial s = \partial V_{h+}/\partial h + V_{h+}/R + \partial V_{s+}/\partial s.$$

On the other hand, from the continuity condition of the velocity vector at the front it follows that $V_{h-} = V_{h+}$, $V_{s-} = V_{s+}$, implying $\partial V_{s-}/\partial s = \partial V_{s+}/\partial s$, and hence $\partial V_{h-}/\partial h = \partial V_{h+}/\partial h = \partial V_n/\partial n$. Substituting the last equality into the relations $-p_- + 2\mu_-(\partial V_{h-}/\partial h) = -p_+ + 2\mu_+(\partial V_{h+}/\partial h)$, which follows from the continuity conditions of the stress vector at the front, we find $(\partial V_n/\partial n)_F = (p_+ - p_-)/2\Delta\mu$ ($\Delta\mu = \mu_+ - \mu_-$). Following the substitution of expansion (4.1) for the pressure with subsequent linearization, the latter equality finally acquires the form

$$(\partial V_n/\partial n)_F = [(p_{0+} - p_{0-}) + (p_+' - p_-')]/2\Delta\mu + \ldots \tag{4.4}$$

Taking in account (4.2)-(4.4), the condition of front motion (2.20) corresponding to the first approximation in ε, is written in the form

$$\frac{\partial f'}{\partial t} + \frac{U}{R_0}(1 + \varepsilon Z) f' + \frac{U}{R_0}\varepsilon Z \frac{\partial^2 f'}{\partial\varphi^2} - V_r' - \frac{1}{2}\frac{R_0}{\Delta\mu}\varepsilon Z (p_+' - p_-') = 0. \tag{4.5}$$

Within the first approximation in ε the remaining linearized boundary conditions and the equations are not changed in comparison with the zeroth approximation. Not dwelling on the technique of finding the perturbation increment which is discussed in detail in [7], we provide the calculation results. We restrict ourselves to the case of a small radius of internal boundary surface S_- and large radius of the external S_+ ($R_-/R_0 \to 0$, $R_+/R_0 \to \infty$ is the front in the field of an axially symmetric hydrodynamic source). We then have for the increment

$$\omega = \omega^0 + \varepsilon\omega^1 + \ldots,\ \omega^0 = U/R_0, \qquad \omega^1 = Z[k^2 - 2k(1 - \alpha)/(1 + \alpha) + 1]U/R_0, \tag{4.6}$$

where $\alpha = \mu_-/\mu_+$; and k is the perturbation mode number ($f' \sim \exp(-\omega t + ik\varphi)$, and i is pure imaginary unity). The quantity in the square bracket in (4.6) is positive. Therefore, as is the case for the zeroth mode, the relation $0 < \omega^0 < \omega$ is satisfied for high modes, so that within the first approximation in ε the perturbations decay more quickly than in the zeroth approximation and the stability of high modes is enhanced. The ω^1 value increases with k, i.e., the higher mode perturbations decay more quickly. Thus, dispersion of the perturbation which has been absent in the zeroth approximation [7] appears within the first approximation.

Similarly to the case of external supply to the medium we obtain that in the second equality for ω^0 in (4.6) the sign changes to the opposite one and, thus, $\omega^0 < \omega < 0$. In this case, within the first approximation in ε the perturbations evolve more slowly than in the zeroth approximation, and there is a tendency to stabilization of the unstable higher modes. However, as is also the case for the zeroth mode, this stabilization can really occur only for small front radii R_0, comparable with its thickness δ, when the frontal regime of reaction transmission degenerates, and the first order approximation in ε becomes operative. Formally the value of the front radius, for which stabilization of the fixed mode takes place, is found by extrapolating the solution (4.6) to the region of small R_0:

$$R_0 = R_0^* = Z[k^2 - 2k(1 - \alpha)/(1 + \alpha) + 1]\,\delta. \tag{4.7}$$

The accuracy of the last relation is enhanced for the higher modes, when $k \gg 1$ and $R_0 \gg \delta$.

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