THERMOCAPILLARY MOTION IN AN EMULSION

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ABSTRACT

The phenomenological model for the motion of an emulsion or a gas-liquid mixture exposed to the thermocapillary forces and microaccelerations is formulated. The analytical and numerical investigation of one-dimensional flows for these media is fulfilled, the structure of discontinuous motion is studied. The stability conditions of a space-uniform state and of the interface between an emulsion and a pure liquid are obtained.

MAIN ASSUMPTIONS

Let us consider the motion of a liquid with inclusion of small drops of another liquid immiscible with the first one. It is supposed that the system is in local thermodynamic equilibrium. The relative phase motion is primarily due to the nonuniformity of the temperature field which produces the thermocapillary effect by virtue of the dependence of the coefficient of the surface tension σ on the temperature T. For simplicity, we suppose that this dependence is linear, $\sigma = \sigma_0 - \sigma_T(T - T_0)$ where σ_0, T_0 and σ_T are some positive constants. In addition to the thermocapillary forces, the system is exposed to the microgravity with acceleration $\vec{g}$.

Further, it is supposed that the drops or bubbles have the spherical form, that they are of equal radius R, and that the volume concentration c of the disperse phase is small. Moreover, we assume that the mean distance between the drops l satisfies the inequalities $R << l << diam\Omega$, where Ω is the volume containing the liquid. In this case, the concepts of mechanics of heterogeneous media are applicable to describe the motion of such a system. Besides, below the Peclet numbers for the liquid matrix and disperse phase are assumed small. This allows us to restrict ourselves to the medium model with the single temperature.

GOVERNING EQUATIONS.

The system of governing equations involves the following unknown functions: T, c, averaged velocity of drops or bubbles $\vec{u}$, and carrying phase $\vec{v}$, pressure of carrying phase p. Let us use the indices d and m to denote the parametres of the disperse phase and liquid matrix, correspondingly. The thermal conductivity of the mixture is determined from the Maxwell formula, whereas, the dynamic viscosity coefficient from the Taylor formula. The resulting system is as follows

$$c_t + div(c\vec{u}) = 0, \tag{1}$$

$$(1-c)_t + div[(1-c)\vec{v}] = 0, \tag{2}$$

$$\rho_d\, c\left(\vec{u}_t + \vec{u}\cdot\nabla\vec{u}\right) + \rho_m(1-c)\left(v_t + \vec{v}\cdot\nabla\vec{v}\right) =$$

$$= -\nabla p + div\left\{\mu_m\left[1 + c\frac{\mu_m + 5\mu_d/2}{\mu_m + \mu_d}\right]\left[\nabla\vec{v} + (\nabla\vec{v})^*\right]\right\} + \rho_d\, c\vec{g} + \rho_m(1-c)\vec{g}, \tag{3}$$

$$\rho_d \lambda_d c\left(T_t + \vec{u}\cdot\nabla T\right) + \rho_m\lambda_m(1-c)\left(T_t + \vec{v}\cdot\nabla T\right) = div\left\{k_m\left[1 - c\frac{3(k_m - k_d)}{2k_m + k_d}\right]\nabla T\right\}, \tag{4}$$

$$\vec{u}-\vec{v}=\frac{2R(\mu_m+\mu_d)}{3\mu_m(2\mu_m+3\mu_d)}\times\left[(\rho_d-\rho_m)R\vec{g}+\frac{3k_m\mu_m\sigma_T\nabla T}{(2k_m+k_d)(\mu_m+\mu_d)}\right], \tag{5}$$

where ρ, μ, λ, k stand for density, dynamic viscosity, heat capacity, and thermal conductivity of the corresponding phase. These coefficients are assumed constant.

System (1) - (5) was obtained in [1]. It should be noticed that this system does not contain any empiric parameter. Equations (1) and (2) are the exact forms (within the framework of the approach of heterogeneous mechanics) of the mass conservation law for the disperse and carrying phases. When deriving the momentum equation (3) and the energy equation (4) we neglected the terms of the second order with respect to c in viscous stresses, and the diffusion heat flow, correspondingly. Besides, we omitted the dissipation function in (4) taking into account the smallness of the arising velocities of the motion.

The completing equation of system (1) - (5) requires a special comment. This relation gives an approximate value of the relative phase velocity in the equation (5) we neglected the effect of added masses and Basset's hereditary force, which is inefficient for the flow with low Reynolds numbers calculated from the drop radius and for smoothly changing external conditions. The coefficients in the right-hand part of (5) correspond to the known dependences in the Hadamard-Rybczinski, [2,3], and Young-Goldstein-Block, [4], formulas. In this connection, it is worth mentioning work [5] where the problem of creeping motion of a small drop with the gravity acceleration collinear to the temperature gradient at infinity was solved explicitly. Also, this paper discussed limits of applicability of formula (5).

Let us consider now the motion of a gas-liquid mixture under the effect of thermocapillary forces and microaccelerations. In the case when the gas is the disperse phase we have $\rho_d << \rho_m$, $\mu_d << \mu_m$, $k_d << k_m$. The corresponding system of governing equations can be obtained from (1) - (5) by a simple limiting procedure. Historically, the model of a gas-liquid motion in weak force fields was derived before in work [6]. Its further analytical and numerical investigation was fulfilled in [7] where, in particular, self-similar solutions and travelling waves were studied.

STABILITY OF SPACE - UNIFORM STATE

The simplest solution of system (1)-(5) corresponds to a uniform relative phase motion with a constant concentration and the temperature distribution being linear function of Cartesian coordinate x in the absence of gravity:

$$c=c_0,\ T=Gx+Ht,\ p=const,$$
$$u=(1-c_0)LG,\ v=-c_0LG \tag{6}$$

where $c_0 \in (0, 1)$ and G are given constants,

$$H=c_0(1-c_0)LG^2(\rho_m\lambda_m-\rho_d\lambda_d)\left[\rho_d\lambda_d c_0+\rho_m\lambda_m(1-c_0)\right]^{-1}$$
$$L=2Rk_m\sigma_T(2\mu_m+3\mu_d)^{-1}(2k_m+k_d)^{-1}; \tag{7}$$

u and v designate projections of vectors $\vec{u}$ and $\vec{v}$ on the direction x.

The linear analysis of the stability of solution (6) with respect to the 3D disturbances proportional to $\exp(\alpha t+i\vec{\beta}\cdot\vec{x})$ where α is the complex frequency and $\vec{\beta}$ is the wave vector leads to a cubic equation for $\alpha(\vec{\beta})$. It turns that two of its roots, α_2 and α_3, always have the negative real part. As for the first one, the real part of α_1 has no definite sign. For the small concentration c_0 we have

$$\mathrm{Re}\,\alpha_1=\frac{c_0(LG)^2\chi_m|\vec{\beta}|^2}{(LG\beta)^2+\chi_m^2|\vec{\beta}|^4}\times\left(\frac{5k_m-2k_d}{2k_m+k_d}\beta^2-\frac{\rho_d\lambda_d}{\rho_m\lambda_m}|\vec{\beta}|^2\right)+O(c_0^2),$$

where $\chi_m=k_m/\rho_m\lambda_m$ is the thermodiffusion coefficient of the carrying phase, β is x-component of the vector $\vec{\beta}$ and L is defined by formula (7). Therefore, the stability conditions is

$$2\left(\frac{\rho_d \lambda_d}{\rho_m \lambda_m}+\frac{k_d}{k_m}\right)+\frac{\rho_d \lambda_d k_d}{\rho_m \lambda_m k_m} \geq 5, \tag{8}$$

moreover one-dimensional disturbances are the most dangerous ones.

It follows immediately from condition (8) that the uniform distribution of bubbles in the liquid $(\rho_d << \rho_m,\ k_d << k_m)$ under the constant gradient of temperature is unstable. The mentioned unstability is of the heat nature. It disappears when $\chi_m \to 0$ and $\chi_m \to \infty$ and has no analogy among the flows in the two-phase media.

On the other hand, given a liquid matrix, we can point out such properties of the disperse phase that inequality (8) will be fulfilled. An example of such a situation is delivered by the emulsion "aluminium-lead" with aluminium as the carrying phase. We also note that the emulsion having the same coefficients of the carrying and disperse phases is neutrally stable in the sence indicated above.

ONE - DIMENSIONAL MOTION

System (1)-(5) is very complicated due to its nonlinearity, high order and mixed type. However, in the case of the one-dimensional motion with plane waves it can be simplified radically. (In this case, we suppose that vectors $\vec{g}$ and ∇T are collinear and parallel to the axis x). In fact, the two first equations of (1)-(5) provide the integral

$$cu+(1-c)v = f(t) \tag{9}$$

where $f(t)$ is the mean volume velocity of the mixture. Now we can express u and v from (9) and the last equation of system (5) in terms of c, T and f. As a result, we obtain a system to determine T and c:

$$c_t + \left\{\left[c\,(Kg + L T_x) - f\right](1-c)\right\}_x = 0; \tag{10}$$

$$\left[\rho_d \lambda_d c + \rho_m \lambda_m (1-c)\right]\left(T_t + f T_x\right) + \left(\rho_d \lambda_d - \rho_m \lambda_m\right) c\,(1-c)(Kg + L T_x) = k_m\left[(1-Mc)\,T_x\right]_x \tag{11}$$

Here we introduce the following notations:

$$K = \frac{2R^2(\rho_d - \rho_m)(\mu_m + \mu_d)}{3\mu_m(2\mu_m + 3\mu_d)}, \qquad M = \frac{3(k_m - k_d)}{2k_m + k_d};$$

parameter L is defined by formula (7) and $g = |\vec{g}|$.

System (10), (11) should be completed with appropriate initial and boundary conditions. An additional boundary condition is necessary to determine the new sought function $f(t)$ (for example, $f = 0$ in the motion possessing a plane of symmetry). The typical problem of such kind was set up in [6]. The gas-liquid mixture is restricted by two parallel solid impermeable walls. The initial distributions of concentration and temperature are given. The heat flux at both walls is prescribed as a function of time. In addition, the impermeability condition for liquid phase is fulfilled at boundaries of the flow domain. The solvability of formulated problem is proved in [8].

DISCONTINUOUS SOLUTIONS

System (10), (11), as well as the original one, does not have a definite type. In some sence, it contains both hyperbolic and parabolic parts. This peculiarity leads to a special structure of discontinuities in its solutions. Namely, the concentration has a jump across the line of discontinuity $x = X(t)$, while the temperature and heat flux are continuous across this line. The jump of concentration gives rise to a jump of the phase velocities. Proceeding in the standard way and using the notation $D = X'(t)$ for the jump velocity, we obtain the conditions on the line of discontinuity:

$$[c]D = \left[\left\{c\,(Kg + L T_x) - f\right\}(1-c)\right],$$

$$[T] = 0,\ \left[(1-Mc)\,T_x\right] = 0. \tag{12}$$

Here the symbol $[r]$ denotes the difference of values of the function r in front and behind the jump.

One of the important cases of concentration discontinuity is a boundary that separates the emulsion and the pure liquid ($c = 0$). In this case $D = u^+$ where u^+ is a velocity of drops in the emulsion (after a jump). Symbol "+" denotes below the emulsion characteristics. The problem of the thermocapillary motion of a single drop near the moving boundary of the emulsion at weightlessness is considered on the base of relations (12) and the Young-Goldestein-Block formula. The difference of the velocities of the boundary u^+ and the drop u^- equals

$$u^+ - u^- = L(k_m - 4k_d)(2k_m + k_d)^{-1} c^+ T_x^+. \qquad (13)$$

If T increases in the direction from the boundary of emulsion into the pure liquid, then the drop is absorbed by the emulsion when $k_m > 4k_d$ and it runs away from the boundary when $k_m < 4k_d$. If the sign of T_x is the opposite, the effect also changes its sign. The loss or the absorption of a drop by the weighted layer is important for its stability. Either lower or upper boundary is unstable. Correspondingly, the suspensed layer will occur near a heated or a cooled wall of the vessel. The formula (13) shows where exactly.

Let us consider now the problem of the motion of a single drop relatively the boundary of a stationary weighted layer of drops. The equilibrium of this layer is possible if $KG = -L T_x^+$. The absence of coalescention of drops is important for the realization of such a layer. The conditions of "attraction" or "repulsion" of the drop from the layer differs in this case from that given above. Namely, if $k_m > k_d$, the single drops are absorbed by the lower boundary of the layer and run away from the upper one (the direction of the increasing of temperature is denoted as the upper one). If $k_m < k_d$, the single drops run away from the upper boundary and, on the contrary, are absorbed by the lower boundary. In the first case the layer is located at the bottom of the vessel, and in the second case it is positioned overhead.

EXAMPLE OF NUMERICAL SOLUTION

Nonlinear initial boundary value problem for system (10), (11) was solved numerically for different combinations of disperse and carrying phases and for different initial distributions of temperature and concentration. We present here the results of its solution in case when the carrying phase is a melted aluminium and the disperse one is a liquid lead. Initial distributions of temperature and concentration are shown on Figures 1 and 2, correspondingly; two other pairs of curves on these figures demonstrate profiles of temperature and concentration at moments $t = 90\,\text{sec}$ and $t = 180\,\text{sec}$. The boundary conditions are the following: $T = T_0 = 1000\ K$ if $x = 0$, $T_x = 0$ if $x = 10\ cm$; $c = 0.03$ if $x = 0$. The gravity is absent, the parameter $\sigma_T R$ entering into the definition (7) of the coefficient L is chosen as $0.001\ gram \cdot cm / \text{sec} \cdot K$.

CONCLUDING REMARKS

In the process of derivation of system (1)-(5), we supposed that all drops or bubbles are of the equal radius. This assumption allows us to minimize dispersion effects. The equations of motion of a polidisperse emulsion or gas-liquid mixture can be obtained using the procedure described above.

A specific trait of the space-uniform state (6) is the dependence of temperature distribution of time. Namely this peculiarity, together with the dependence of mixture heat conductivity coefficient of concentration, lead to the origin of a non-zero real part of the first root in the dispersion equation. Ignoring these factors, we obtain the neutral stability of the convective mode corresponding to the root α_1.

The conclusion concerning the instability of uniform distribution of bubbles in a gradient temperature field correlates with the results of numerical simulation of 1D thermocapillary motion in a gas-liquid mixture [7]. Here we mention one of them. Let as suppose that initial distribution of concentration is like to a smoothed shelf while initial distribution of temperature is close to a linear one, moreover, the derivatives of both functions are of the same sign. Nonlinear development of process brings to the steepening of the concentration profile and to the loss of its monotonicity: after passing the region of large gradients, the concentration peak appears and this peak is growing rapidly. At the same time, the temperature profile remains close to linear and deforms slightly in the zone where the concentration changes abruptly.

As for stability conditions of the interface "emulsion-pure liquid", they were obtained on the base of conservation laws on the discontinuity. In spite of that, an intrinsic mechanism of instability of two-phase flow was not taken into account. Therefore, the mentioned conditions of stability should be considered as the necessary ones.

In conclusion, we remark that system (1)-(5) can be used for the study of solidification process of an emulsion. To this end, we have to add to these equations appropriate Stefan-type conditions at the front of solidification as well as, in the simplest case, the heat equation in the solid phase. Generally speaking, the solidification temperature of disperse phase is not the same with one of liquid matrix. This circumstance leads to complication of the problem under consideration due to possible appearance of two fronts of solidification. On the other hand, the situation is feasible when drops are driven back from the front of solidification of the pure liquid. Besides the direct problem, the inverse one has an interest: to get the given (for example, constant) distribution of inclusions in the solid material by the control of the boundary regime of cooling.

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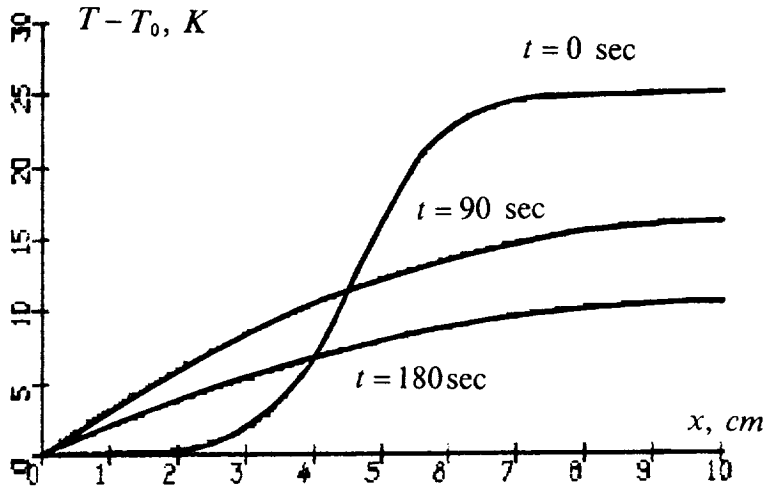

Fig.1

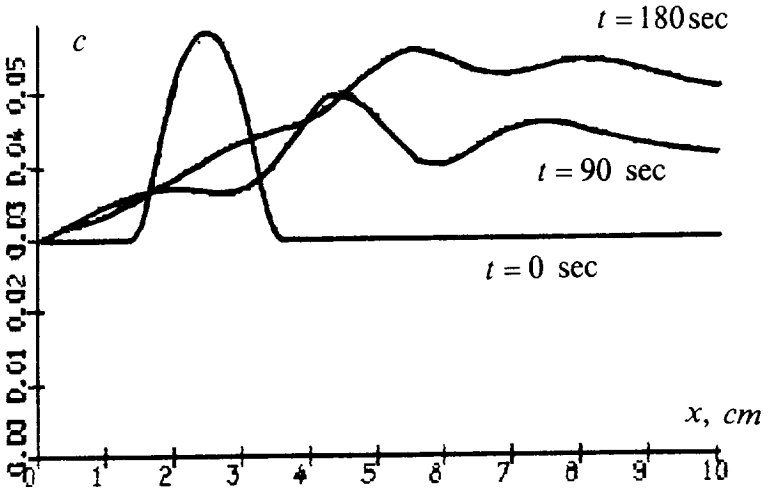

Fig.2

Nonlinear evolution of temperature (Fig. 1) and concentration (Fig.2) in the process of the motion of lead droplets ($\sigma_T R = 0.001\ gram \cdot cm / \sec \cdot K$) in a melted aluminium.