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THERMOCAPILLARY MOTION IN A GAS—LIQUID MIXTURE

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1. Equation of Motion. Let a viscous incompressible liquid with gas bubbles be found in the region of space Ω. The number of bubbles is sufficiently large that a number $a \ll d$ can be found, where d is the diameter of Ω, such that any sphere of radius a belonging to Ω contains a number of bubbles $N \gg 1$. The bubbles are assumed to be spheres of identical radius R. If the characteristic distance between bubble centers l is sufficiently small in comparison with the characteristic distance L over which the mean mixture parameters vary, the concepts of mechanics of heterogeneous media (see, e.g., [1]) are valid.

If the external mass forces are small and the acceleration of the liquid is also small, the main source of motion is the inhomogeneity of the temperature field in the liquid and the thermocapillary effect induced by it [2].

We denote by c the bulk concentration of the gas, by u and v the average velocities of the gas and liquid phases, respectively, and by T the temperature. The exact continuity equation (within the scope of fluid mechanics) for the liquid phase is

$$\partial(1-c)/\partial t + \operatorname{div}[(1-c)\mathbf{v}] = 0. \qquad (1.1)$$

Allowing the gas density to satisfy $\rho_g = \text{const}$, the continuity equation for the gas phase is similar to (1.1):

$$\partial c/\partial t + \operatorname{div}(c\mathbf{u}) = 0. \qquad (1.2)$$

The possible gas-exchange process between bubbles and the liquid due to diffusion processes is not taken into account. For simplicity, we do not take into account either the more important process of bubble coagulation, which up to a certain extent is justified in the case of a dilute system.

Taking into account that the shear viscosity of a suspension of gas bubbles equals $(1 + c)\nu$, where ν is the viscosity of the liquid, and neglecting quadratic terms of order c^2 in the viscous stresses, one can write the momentum equation of the liquid in the form

$$(1-c)d\mathbf{v}/dt = -\rho^{-1}\nabla p + (1-c)\mathbf{g} + 2\operatorname{div}[(1+c)\nu S], \qquad (1.3)$$

where S is the velocity deformation tensor; p, pressure; and $\mathbf{g}$, acceleration of the external mass forces. Equation (1.3) is valid for small Reynolds numbers of bubble flow and for sufficiently large characteristic times of motion, when effects of associated bubble masses can be neglected.

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Equation (1.3) is at the same time a momentum equation of the mixture, since the gas density ρ_g is low in comparison with the liquid density ρ. It is important to note that the thermocapillary effect does not give a direct contribution to (1.3), since in the quasi-stationary approximation the force acting on the gas bubbles vanishes [2].

Turning to the heat-transfer equation, it is natural to assume that the heat conductivity of the gas $\varkappa_g$ is small in comparison with that of the liquid $\varkappa$. For the case $\varkappa_g \ll \varkappa$ the heat conductivity of the mixture, determined by Maxwell's equation [3, 4], equals

$$\varkappa' = \varkappa(1 - 3c/2). \tag{1.4}$$

Generally speaking, convective transport during fluid flow of separate bubbles can affect heat transport. The inhomogeneity of the velocity field of the fluid, generating bubble motion, has little effect on heat transport if the Peclet number calculated by the characteristic bubble velocity relative to the liquid and by the mean distance l between bubble centers is sufficiently small:

$$\mathrm{Pe}_l = wl/\chi \ll 1 \tag{1.5}$$

(χ is the temperature conductivity coefficient of the liquid). Under condition (1.5) the heat conductivity of the gas—liquid mixture is determined by Eq. (1.4), valid in the absence of relative phase motion.

It is well known [5, 6] that the temperature dependence of the density, heat conductivity, and heat capacity of the liquid is weaker by two orders of magnitude than the viscosity, and significantly weaker than the surface tension. Therefore, in the heat-transfer equation it is not necessary to take into account the variation in heat capacity and heat conductivity, and as a result one obtains the following equation for the temperature:

$$(1-c)\left(\frac{\partial T}{\partial t} + \mathbf{v}\cdot\nabla T\right) = \chi\,\mathrm{div}\left[\left(1 - \frac{3}{2}c\right)\nabla T\right]. \tag{1.6}$$

The thermocapillary effect has the meaning of considering small Reynolds and Peclet numbers

$$\mathrm{Re} = wR/\nu \ll 1,\quad \mathrm{Pe} = wR/\chi \ll 1.$$

The latter condition is satisfied due to (1.5). The condition of small Reynolds number is usually satisfied for small Marangoni numbers

$$M = \frac{R^2}{\rho\nu^2}\left|\frac{d\sigma}{dT}\right||\nabla T|.$$

Since the bubble size is small in comparison with the flow scale ($R \ll L$), a change in σ along its surface is relatively small and is confined to a situation in which the surface tension σ varies linearly with increasing temperature (usually decreasing).

For the given liquid—gas pair, the condition $M < 1$ can be attained by choosing a sufficiently small $R^2|\nabla T|$. For example, for pure water near T = 20°C, the Marangoni number is $M < 1$ if $R^2|\nabla T| < 10^{-3}$ cm•deg. For liquid copper near the melting temperature $M < 1$ for $R^2|\nabla T| < 1.3\cdot 10^{-4}$ cm•deg. For small M the values of w and ∇T are related by a linear dependence, having the following shape according to similarity and dimensionality theories [7]:

$$\mathbf{w} = K\nabla T,\quad \mathbf{w} = \mathbf{u} - \mathbf{v}, \tag{1.7}$$

where $K = \varphi(c)|d\sigma/dT|R/2\mu$ accurately up to terms of order Re, Pe, and the ratios of dynamic viscosities and heat conductivities in the gas and in the liquid μ_g/μ and $\varkappa_g/\varkappa$. For small bulk concentrations of the gas ($c \ll 1$) the equation must transform to the equation for the velocity of thermocapillary motion relative to the bubble [2]. Therefore, $\varphi = 1$ for $c = 0$.

We stress that the coefficient K in (1.7) depends strongly on temperature. This occurs mostly due to the temperature dependence of the viscosity ν.

System (1.1)-(1.3), (1.6), (1.7) contains nominally nine scalar equations for the nine unknown quantities: c, the three components of **u** and **v**, p, and T. In practice, equality (1.7) is an ultimate relation between **u** and **v**, and makes it possible to reduce the number of unknown functions to six.

We refine (1.7), related to taking into account the effect of a variance of velocities of separate bubbles.

2. Account of the Effect of Chaotic Motion. The velocities of motion of even nearest bubbles in the gas—liquid mixture differ strongly, generally speaking, among thenselves and are not equal to the mean bubble velocity. The reason for fluctuations is the impossibility of stable stationary states. It can be shown that a stationary state of thermocapillary motion of an ordered bubble system is exponentially unstable even for small Reynolds numbers. This is already clear from stability considerations of thermocapillary motion of an unconfined bubble chain moving perpendicularly to the chain axis. It is therefore natural to assume that in such a system chaotic bubble motion is established due to hydrodynamic forces. It is well known that one reason for stochastization of a dynamic system is instability of its stationary states.

Stochastization of motion due to hydrodynamic forces is, obviously, a general property of two-phase media in the presence of relative phase motion. It occurs for arbitrary Reynolds numbers and various forces generating relative motion. Exponential instability of motion of an ordered particle system in an ideal liquid was proved in [8]. Vibrational motion of bubbles floating in a heavy liquid at finite Reynolds numbers was repeatedly observed in experiments [9].

The chaotic bubble motion in the nonuniform problem must be given by a diffusion type correction to Eq. (1.7):

$$w_i = K\nabla_i T - c^{-1}D_{ij}\nabla_j c, \quad i, j = 1,2,3. \tag{2.1}$$

The shape of the tensor $D_{ij} = D_{ij}(c, w)$ is uniquely determined by dimensionality considerations and space isotropy

$$D_{ij} = R|\mathbf{w}|[f_\perp(c)\delta_{ij} + (f_\parallel(c) - f_\perp(c))w_i w_j/|\mathbf{w}|^2].$$

It is important to note that the second term in (2.1) is of small order $R/L \ll 1$, where L is the scale of variation of mean quantities. Generally speaking, besides ∇c ∇w can provide a contribution to (2.1). It is sensible, however, to restrict ourselves to the physically obvious dependence of (2.1).

Until recently, there were no closed expressions of type (2.1) in the theory of two-phase media, with the coefficients D_{ij} depending only on c and the relative phase velocities (or on ∇T to the same accuracy). The diffusion term in the expression for the relative motion velocity or in the phase interaction force is of significant value in problem of medium stability.

The order of the coefficients D_{ij} in (2.1) is determined by hydrodynamic interactions in the system. It is obvious, therefore, that $D_{ij} \to 0$ for $c \to 0$, when the bubble system is rarefied.

We estimate approximately the order of the bubble diffusion coefficient in a rarefied system, when the bubble radius R is much smaller than the mean distance between their centers. Obviously, $D \sim w_* s$, where w_* is the mean-square velocity fluctuation of the bubble, and s a characteristic distance at which the characteristic velocity fluctuation w_* occurs. The perturbation ∇T at the given point, occurring due to the presence of bubbles at distance r, is of order r^{-3}. The velocity perturbation due to these bubbles (the "Stokelet" term, decreasing as r^{-1}, is absent) is of the same order r^{-3}. Using a characteristic value $r \sim l$, we find $w_* \sim |\mathbf{w}|R^3/l^3$. Using the quantity $s \sim l$, we finally obtain the estimate

$$D \sim w_* s \sim |\mathbf{w}| R c^{2/3}. \tag{2.2}$$

3. One-Dimensional Motion. Most simple classes of solutions of system (1.1)-(1.3), (1.6), and (1.7) describe one-dimensional motion with planar, cylindrical, and spherical waves. For one-dimensional motion the system admits further reduction of the order. Firstly, Eqs. (1.1), (1.2), (1.6), and (1.7) form in this case a closed subsystem, after whose solution the pressure is found from (1.3) by quadrature. Secondly, combining (1.1) and (1.2) and integrating over the spatial coordinate, we obtain a linear relation between the phase velocities. Using this relation and equality (2.1) to express the velocities mentioned in terms of c and ∇T, ∇c, and substituting the results into (1.1), (1.6), we obtain two coupled equations for concentration and temperature, both being of second order if $D \neq 0$. We provide this system for motion with planar waves (the notation is obvious):

$$c_t + [(KcT_x - Dc_x - f)(1 - c)]_x = 0; \tag{3.1}$$

$$(1 - c)[T_t - (KcT_x - Dc_x - f)\,T_x] = (\chi' T_x)_x. \tag{3.2}$$

Here $f = f(t)$ is an arbitrary function of time which must be determined together with the solution from further conditions, and χ' is the heat-conductivity coefficient of the mixture, generally dependent on c.

For the system of quasilinear equations (3.1), (3.2) one can formulate various initial- or boundary-value problems. We assume for simplicity that $D = 0$, and that K and χ' are positive constants. It is required to find functions $c(x, t)$, $T(x, t)$ in the rectangle $0 < x < L$, $0 < t < t_0$ and a function $f(t)$ on the interval $0 < t < t_0$, such as to satisfy Eqs. (3.1), (3.2), the initial conditions

$$c(x, 0) = c_0(x),\ T(x, 0) = T_0(x); \tag{3.3}$$

the boundary condition at $x = 0$

$$T_x(0, t) = \psi_0(t),\ f(t) = Kc(0, t)\psi_0(t); \tag{3.4}$$

and boundary conditions at $x = L$

$$T_x(L, t) = \psi_L(t),\ f(t) = Kc(L, t)\psi_L(t). \tag{3.5}$$

We assume that the following conditions are satisfied: $c_0 \in C^{1+\alpha}[0, L]$ $(0 < \alpha < 1)$, $0 \leqslant c_0 \leqslant 1$ for $0 \leqslant x \leqslant L$; $T_0 \in C^{3+\alpha}[0, L]$, $dT_0/dx > 0$ for $0 \leqslant x \leqslant L$; $\psi_0 \in C^{(3+\alpha)/2}[0, t_0]$, $\psi_L \in C^{(3+\alpha)/2}[0, t_0]$, and $c_0(0)\psi_0(0) = c_0(L)\psi_L(0)$. There exists then a $t_0 > 0$, so that problem (3.1)-(3.5) has a unique solution, with $0 \leqslant c \leqslant 1$ and $T_x > 0$ for all $x \in [0, L]$, $t\ [0, t_0]$.

The physical interpretation of the problem is the following. The gas—liquid mixture occupies the space between the parallel solid impenetrable planes $x = 0$ and $x = L$. Initially, the concentration and temperature distributions are given, depending on x. The thermal flux is given at the boundaries $x = 0$ and $x = L$ (the first two conditions (3.4), (3.5)), as well as the nonflow conditions for the liquid phase $v = 0$ (the second two conditions). If the thermal fluxes at the boundaries are independent of coordinates along the planes, the generated thermocapillary motion is one-dimensional.

Problem (3.1)-(3.5) belong to a class of the so-called "inverse problems," where along with solution of a system of differential equations one also finds the unknown coefficients of this system (in the present case function f(t)). The function can sometimes be found a priori. This situation arises, e.g., in the problem of spherically symmetric motion inside a sphere, on whose boundary is given a heat-exchange condition with the surrounding gas. Let now u and v denote the radial velocity components of the gas and liquid phases and let $r = |x|$. From the conditions $u = v = 0$ at $r = 0$ we find $f = 0$, leading to the expressions $u = (1 - c)KT_r$, $v = -cKT_r$. For $D = 0$ the problem reduces to searching the function s(t), determining the free boundary, and the functions $c(r, t)$, $T(r, t)$ from the relations $c_t + r^{-2}[Kr^2 \cdot c(1 - c)T_r]_r = 0$ at $0 < r < s(t)$, $(1 - c)\ (T_t - KcT_r^2 = r^{-2}(\chi' r^2 T_r)_r$; $s(0) = s_0 > 0$, $c(r, 0) = c_0(r)$, $T(r, 0) = T_0(r)$; c and T are bounded, c_r, $T_r \to 0$, and $r \to 0$; $T_r = -q[T - T_s(t)]$ for $r = s(t)$, $ds/dt = -KcT_r$ at $r = s(t)$. In the penultimate conditions $q = q(T)$ is the coefficient of interphase heat exchange, and $T_s(t)$ is the given temperature outside the medium. The last condition implies that the free boundary $r = s(t)$ confines the liquid volume. It can be expected that under conditions $T_0(r) \geqslant 0$, $T'_s(t) \geqslant 0$ and appropriate smoothness conditions and matching in input data, the problem stated is correctly formulated, though we have not yet succeeded in proving that. Monotonic increase of the functions T_0 and T_s can be guaranteed in this case by positive u, and ultimately guarantees purification of the liquid medium from bubbles.

If $f\sqrt{t} = \text{const}$ and $D = 0$, system (3.1), (3.2) admits the self-similar solutions

$$c = c(x/\sqrt{t}),\ T = T(x/\sqrt{t}).$$

For $f = 0$ the solution in this case consists in the following: the gas—liquid mixture fills the half space $x > 0$, and for $t = 0$ has constant parameters $c = c_0$, $u = v = 0$, $T = T_0$. Initially, the temperature at the plane $x = 0$ undergoes a jump discontinuity to the value T_1, which is then maintained constant. The bubble motion generated at the edge of the plane $x = 0$ is determined by a system of ordinary differential equations which is not given here.

The simplest nontrivial solution of system (3.1), (3.2) with $D \neq 0$ is admitted in the case of a coefficient K independent of T. It corresponds to $f = KGc_0$, and has the form

$$c = c_0,\ T = Gx, \tag{3.6}$$

where c_0 and G are constant, and $0 < c_0 < 1$. It corresponds to motion with constant pressure, gas phase velocity KG, and vanishing velocity of the liquid phase. Below solution

(3.6) is investigated for stability with respect to one-dimensional perturbations.

4. Stability of Thermocapillary Motion. Consider stability of stationary motion with plane waves to short-wave perturbations, when the wavelength of the perturbation is significantly smaller than the size of the flow region L. For this we linearize the equation for the perturbation following from (3.1), (3.2). The expression for f is not perturbed, $\delta f = 0$, which is directly related to the unperturbed boundary conditions. For short wavelengths one may neglect the dependence of coefficients on coordinates in the equations of perturbed motion, as well as the effect of boundaries, and seek small perturbations of the concentration δc and the temperature δT in the approximate form

$$\delta c = A \exp(\alpha t + ikx), \ \delta T = B \exp(\alpha t + ikx),$$

where A and B are constants; α, a complex frequency; k, wave number; and $kL \gg 1$. The dispersion equation is

$$\begin{gathered}\alpha^2 + \alpha\left[\chi\left(1 - \frac{c}{2}\right)k^2 + (1-c)\,ik\left(cK_c' + K\right)T_x + (1-c)\,Dk^2\right.\\ \left.- ickKT_x - cK_T'T_x^2\right] + \chi k^2\left[\frac{3}{2}cT_x^2K_T' + \left(1 - \frac{3}{2}c\right)Dk^2 + ik\left(cK_c' + K\right)T_x\right] = 0.\end{gathered} \tag{4.1}$$

At low concentrations $c \ll 1$ the roots of Eq. (4.1) can be written in the form

$$\alpha_1 = -(1-c)\left[ik\left(K + cK_c'\right)T_x + Dk^2\right] + c\left(1 - \frac{5}{2}\frac{\chi k}{\chi k - Dk - iKT_x}\right)\left(ikKT_x + T_x^2K_T'\right) + O(c^2),$$

$$\alpha_2 = -\chi k^2 + O(c).$$

Within the applicability of the model discussed, we always have $\operatorname{Re}\alpha_2 < 0$. The stability of the medium is determined by the expression

$$\operatorname{Re}\alpha_1 = -Dk^2(1-c) + cT_x^2K_T' + \frac{5}{2}cT_x^2\chi k^2\frac{K^2 - (\chi - D)K_T'}{(\chi - D)^2k^2 + T_x^2K^2} + O(c^2) \quad \text{for} \quad c \to 0. \tag{4.2}$$

If the coefficient of thermocapillary drift is independent of temperature ($K_T' = 0$) and diffusion is absent ($D = 0$), Eq. (4.2) describes an instability. This instability has essentially the nature of thermal instability (it vanishes for $\chi \to \infty$), and has no analog in mechanics of two-phase media.

Taking into account diffusion, the instability is retained in the region of sufficiently long waves. Due to the approximate estimate (2.2)

$$D \sim c\chi \mathrm{Pe}_l \ll c\chi. \tag{4.3}$$

It is seen from Eq. (4.2) that accurately to small orders of c, the quantity $\operatorname{Re}\alpha_1 > 0$ is in the region

$$|k| < k_* = \frac{|w|}{\chi}\sqrt{\frac{5c\chi}{2D} - 1} \sim l^{-1}\sqrt{\mathrm{Pe}_l}.$$

It is here taken into account that $KT_x \approx w$. In a significant region of wave numbers, the growth increment of the perturbation is nearly constant:

$$\operatorname{Re}\alpha \approx 5cw^2/2\chi, \quad |w|/\chi \ll |k| \ll k_*.$$

In the small wave number region, when $k\chi/|w| \to 0$, we have $\operatorname{Re}\alpha_1 \to 0$. Taking into account the temperature dependence of the thermocapillary drift coefficient ($K_T' \neq 0$) changes substantially the asymptotic nature of $\operatorname{Re}\alpha_1$ for $k \to 0$.

It is convenient to introduce the dimensionless notation

$$\Pi = \frac{K_T'}{K^2}\chi, \quad Q = \frac{w^2}{\chi^2k^2}, \quad Q_* = \frac{2D}{5c\chi} \ll 1.$$

Taking into account (4.3), Eq. (4.2) can be reduced to simple form, neglecting the small c^2 and D/χ:

$$\operatorname{Re}\alpha_1 \approx \frac{5cw^2}{2\lambda}\left(-\frac{Q_*}{Q} + \frac{2\Pi}{5} + \frac{1-\Pi}{1+Q}\right).$$

The effect of the temperature dependence of the thermocapillary drift coefficient ($\Pi \neq 0$) is determined by the sign of K'_T, and is differently manifested in various wave number regions.

For $\Pi < 0$ the critical Q value decreases with respect to Q_*, the instability advances toward the region of shorter waves, and the system becomes at the same time stable in the region $Q \gg 1$ (long waves).

For $\Pi > 0$ the boundary of the region of short-wave stability can be extended: from $Q \approx Q_* \ll 1$ at $\Pi = 0$ to $Q \sim 1$ at $\Pi > 2/3$. In this region of long waves, however (i.e., for $Q \to \infty$), we have $\operatorname{Re} \alpha_1 \to \text{const} > 0$.

The regions of large and small Q have a clear physical meaning. For $Q > 1$ the bubbles move relatively faster than the thermal wave front (by a distance on the order of the perturbation wavelength), and the situation is the opposite for $Q < 1$. Thus, the increase of the thermocapillary drift coefficient with temperature leads to instability enhancement if the bubbles move faster than the thermal wave ($|w| > \chi k\sqrt{3/2}$), and significantly enhances the stability if the bubbles move relatively slowly. When $K'_T < 0$ the situation is the opposite.

Usually $K'_T > 0$ for bubbles in the liquid. The quantity Π is appreciable for sufficiently small bubbles, as well as for low-viscosity liquids. Thus, for water at 20°C we have $\Pi \sim 1$ for bubbles of radius $R \sim 10^{-5}$ cm.

The instability is significant only for sufficient flow extension L if the Peclet number satisfies $|w|L/\chi \gg 1$ (for moderate values $|\Pi| \lesssim 1$). In other words, as can be shown from Eq. (4.2), the perturbations are removed together with the bubbles from the region under consideration until there is a considerable increase in amplitude.

If $K'_T = 0$, the solution (3.6) is the exact solution of system (3.1), (3.2) with $f = \text{const}$. In this case the preceding analysis makes it possible to draw conclusions concerning the stability of the uniform bubble distribution in space in the presence of a constant temperature gradient. For $D = 0$ this distribution is absolutely unstable. Account of bubble diffusion ($D \neq 0$) stabilizes short-wave perturbations with $|k| < k_*$, but outside this region of wave numbers the instability is retained.

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