

THE APPEARANCE OF THERMOCAPILLARY CONVECTION IN  
THE NONMOVING LAYER OF A LIQUID OF VARIABLE VISCOSITY

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The Marangoni instability in the nonmoving layer of a liquid exhibiting constant viscosity was studied in [1-3]. In such chemical engineering processes as, for example, in the nonisothermal chemisorption of gases by liquid films, we frequently make use of viscous liquids exhibiting high Prandtl numbers [4]. The dynamic viscosity of such liquids diminishes markedly as temperature rises, and this effect, apparently, must significantly affect the critical Marangoni numbers which determine the conditions for the appearance of thermocapillary convection. Taking the variability in the physical properties of the liquid into consideration is an urgent problem also in the investigation of the processes in the hydro-mechanics and heat and mass exchange that occurs in the growth of crystals [5-8].

Below we present a solution for the problem of the stability of a nonmoving layer of a viscous liquid in contact with a gas, with provision made for the relationship between surface tension and the coefficient of dynamic viscosity to temperature. The neutral stability curve has been constructed analytically to link the critical Marangoni number, the viscosity gradient through the thickness of the layer, and the wave number of three-dimensional perturbation.

Let us examine the nonmoving layer of a liquid at whose free surface heat exchange takes place with the ambient medium in accordance with Newton's law. The Navier-Stokes equation and the equation for convective heat exchange are taken in the form

$$\rho \frac{\partial v_i}{\partial t} + \rho v_i \frac{\partial v_j}{\partial x_i} = - \frac{\partial p}{\partial x_j} + \frac{\partial}{\partial x_i} \left( \mu \frac{\partial v_j}{\partial x_i} \right) + \frac{\partial \mu}{\partial x_i} \frac{\partial v_i}{\partial x_j}, \quad (1)$$

$$\partial T / \partial t + (\mathbf{v} \nabla T) = a \Delta T, \quad \text{div } \mathbf{v} = 0,$$

where  $\mathbf{v} = \{v_x, v_y, v_z\}$  is the velocity of the liquid;  $p$ , pressure;  $T$ , temperature;  $\rho$ ,  $\mu$ , and  $a$ , density, the dynamic coefficient of viscosity, and the coefficient of thermal diffusivity;  $a = \lambda / \rho c_p$ ;  $\lambda$ , coefficient of thermal conductivity;  $c_p$ , specific heat capacity.

Let us assume that the viscosity of the liquid depends exponentially on the temperature, while the coefficient of surface tension is linearly dependent on temperature:

$$\mu = \mu_w e^{-\beta(T-T_w)}, \quad \sigma = \sigma_w - \varepsilon(T - T_w) \quad (2)$$

( $\beta$ ,  $\varepsilon$ , approximation parameters;  $T_w$ , wall temperature). Functions (2) are valid for a broad class of incompressible liquids [9-11].

Let the state of equilibrium be described by the steady-state solution of (1), corresponding to the heat-conduction regime:

$$\mathbf{v} = 0, \quad \frac{d^2 \widehat{T}}{dy^2} = 0, \quad \widehat{T}(0) = T_w, \quad \lambda \frac{d\widehat{T}}{dy}(h) = \alpha [T(h) - T_0]. \quad (3)$$

Here  $h$  is the thickness of the layer;  $\alpha$  is the coefficient of heat exchange;  $T_0$  is the temperature of the gas.

The solution of the heat-conduction equation with boundary conditions (3) has the form

$$\widehat{T} - T_w = - \frac{\text{Bi}(T_w - T_0)}{1 + \text{Bi}} \frac{y}{h}, \quad \frac{d\widehat{T}}{dy} = - \frac{\text{Bi}(T_w - T_0)}{(1 + \text{Bi})h} \quad (4)$$

( $\text{Bi} = \alpha h / \lambda$  is the Biot number). In the following we will assume that the temperature of the wall is greater than that of the gas, and that the temperature gradient is negative

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( $dT/dy < 0$ ). When the flow of heat at the wall is established, the first of the boundary conditions in (3) assumes the form  $\lambda(dT/dy)(0) = -q_w$  ( $q_w > 0$ ).

We will use the method of small perturbations to examine the steady state (4) for stability. Let  $\mathbf{v} = \mathbf{w}$ ,  $T = \hat{T}(y) + T'(x, y, z, \tau)$  ( $\mathbf{w}$  and  $T'$ , perturbations in velocity and temperature). Let us introduce the dimensionless quantities

$$y = y/h, x = x/h, z = z/h, \tau = t\mu_w/(\rho h^2), \text{Pr}_w = \gamma_w/a, \gamma_w = \mu_w/\rho, \\ \Theta = \frac{T - T_0}{T_w - T_0} \left( \frac{(T - T_0)\lambda}{q_w h} \right), \text{Ma} = \frac{\varepsilon(T_w - T_0)h}{\mu_n a} \frac{\text{Bi}}{1 + \text{Bi}} \left( \frac{\varepsilon q_w h^2}{\lambda \mu_n a} \right), \\ N = -\beta \frac{d\hat{T}}{dy} h = \frac{\beta(T_w - T_0)\text{Bi}}{1 + \text{Bi}}, \quad \omega = \frac{\mathbf{w}}{\gamma_w/h^2}$$

( $\mu_n = \mu_w e^{-N}$  represents the viscosity of the liquid at the surface temperature).

By applying the curl operation twice to the Navier-Stokes equation we will eliminate pressure and obtain a scalar equation for the normal velocity component  $\omega_y = \omega$ . This equation and the equation of convective heat exchange is linearized in the vicinity of the steady-state solution of (3), (4). As a result we have

$$\frac{\partial}{\partial \tau} \Delta \omega = f \Delta \Delta \omega + f'' \Delta \omega + 2f' \frac{\partial}{\partial y} \Delta \omega + 2f'' \frac{\partial^2 \omega}{\partial y^2}, \quad \text{Pr}_w \frac{\partial \Theta}{\partial \tau} - \omega = \Delta \Theta, \quad (5)$$

where  $f = e^{Ny}$ ;  $\Delta$  is the Laplace operator; the prime designates the derivative with respect to  $y$ .

Further, we will assume that the free surface of the liquid is undeformable (the distortion parameter is small), and the effect of the capillary forces reduces to the appearance of surface-tension gradients, which are offset by the tangential stresses. For temperature perturbations we will assume ordinary boundary-value conditions which take into consideration the exchange of heat at the free surface and at the solid wall:

$$\omega(0) = 0, \quad \frac{\partial \omega}{\partial y}(0) = 0, \quad \omega(1) = 0, \quad \frac{\partial^2 \omega}{\partial y^2}(1) = \text{Ma} \Delta_1 \Theta|_{y=1}, \\ \Theta(0) = 0 \text{ (or } \Theta'(0) = 0), \quad \frac{\partial \Theta}{\partial y}(1) = -\text{Bi} \Theta(1) \left( \Delta_1 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right). \quad (6)$$

The first and second of the boundary conditions in (6) follow from the hypothesis of adhesion and from the equation of continuity.

Let us note that with a constant viscosity  $N = 0$ ,  $f = 1$  we obtain the familiar Pearson problem [1] which deals with the appearance of thermocapillary convection in the nonmoving layer of the liquid.

Following the widely accepted Pelieu and Southwell hypothesis [11], we will look for the solution of (5) with boundary conditions (6) in the form

$$\omega(x, y, z, \tau) = -F(x, z)\omega(y)e^{\sigma\tau}, \quad \Theta(x, y, z, \tau) = F(x, z)\Theta(y)e^{\sigma\tau} \quad (7)$$

[ $F(x, z)$  is the function of the horizontal structure, satisfying the equation  $\Delta_1 F = -\alpha^2 F$ ]. Using (5)-(7), we obtain the spectral problem

$$\sigma(\omega'' - \alpha^2 \omega) = f(y)[\omega^{IV} - 2\alpha^2 \omega'' + \alpha^4 \omega - N^2(\omega'' - \alpha^2 \omega) + \\ + 2N(\omega''' - \alpha^2 \omega') + 2N^2 \omega''], \\ \text{Pr}_w \sigma + \omega = \Theta'' - \alpha^2 \Theta, \\ \omega(0) = 0, \omega'(0) = 0, \omega(1) = 0, \omega''(1) = \text{Ma} \alpha^2 \Theta(1), \\ \Theta(0) = 0 \text{ (or } \Theta'(0) = 0), \Theta'(1) = -\text{Bi} \Theta(1). \quad (8)$$

Let us examine the neutral perturbation  $\sigma = 0$ . In this case we obtain an equation with constant coefficients

$$\omega^{IV} - 2\alpha^2 \omega'' + \alpha^4 \omega - N^2(\omega'' - \alpha^2 \omega) + 2N(\omega''' - \alpha^2 \omega') + \\ + 2N^2 \omega'' = 0, \quad \Theta'' - \alpha^2 \Theta = 0 \quad (9)$$

with boundary conditions (8).

Let us construct the Green's function of the operator  $\theta'' - \alpha^2\theta = 0$  with the boundary conditions  $\theta(0) = 0$ ,  $\theta'(1) = -Bi\theta(1)$  and let us calculate its value at the interphase boundary ( $y = 1$ ):

$$G(\xi, \alpha, Bi) = -\frac{(e^{\alpha\xi} - e^{-\alpha\xi})}{e^{-\alpha}(\alpha - Bi) + e^{\alpha}(\alpha + Bi)}.$$

The temperature perturbation of the free surface in this case

$$\Theta(1) = \frac{Bi}{1+Bi} \int_0^1 G(\xi, \alpha, Bi) \omega(\xi) d\xi.$$

The neutral stability curve assumes the form

$$Ma = \frac{\omega''(1, \alpha, N)}{\alpha^2 \int_0^1 G(\xi, \alpha, Bi) \omega(\xi) d\xi} \quad (10)$$

[ $\omega$  is the solution of the hydrodynamic equation (9)]. From relationship (10) it follows that  $Ma \rightarrow \infty$  as  $\alpha \rightarrow 0$  and  $\alpha \rightarrow \infty$ . It is comparatively simple to construct the asymptote of (10). As  $\alpha \rightarrow \infty$  the influence of the parameter  $N$  is insignificant and Eq. (10) has the same asymptotes as the neutral stability curve in the Pearson problem [1].

As  $\alpha \rightarrow 0$  the hydrodynamic equation (9) is simplified:

$$\omega^{IV} + 2N\omega^{III} + N^2\omega'' = 0. \quad (11)$$

Equation (11) has a fundamental system of solutions  $\omega = \{1, \xi, \xi e^{-N\xi}, e^{-N\xi}\}$ . Using this system, we obtain

$$Ma_{\alpha \rightarrow 0} = \frac{1+Bi}{\alpha^2} \varphi_1(N) \quad \text{wall temperature given,}$$

$$Ma_{\alpha \rightarrow 0} = \frac{Bi}{\alpha^2} \varphi_2(N) \quad \text{heat flow given.}$$

The functions  $\varphi_1(N)$  and  $\varphi_2(N)$  are found as a result of solving the hydrodynamic equations

$$\varphi_1(N) = \frac{6N^4 e^{-N}(N^2 - 2N + 2 - 2e^{-N})}{(N^3 - 48N + 12) + (2N^4 + 5N^3 + 12N^2 + 12N - 24)e^{-N} + (6N^2 + 12)e^{-2N}},$$

$$\varphi_2(N) = \frac{2N^3 e^{-N}(N^2 - 2N + 2 - 2e^{-N})}{N^2 - 4N + 2 + (N^3 + N^2 + 4N - 4)e^{-N} + 2e^{-2N}}.$$

We can verify that as  $N \rightarrow 0$ ,  $\varphi_1 \rightarrow 80$ ,  $\varphi_2 \rightarrow 48$ , i.e., we will obtain the neutral Pearson curve for a liquid layer of constant viscosity.

Let us introduce the functions  $\chi_1 = \varphi_1/80$ ,  $\chi_2 = \varphi_2/48$ , so that

$$Ma/Ma(0) = \chi_1(N), \quad Ma/Ma(0) = \chi_2(N), \quad (12)$$

where  $Ma(0)$  represents the critical Marangoni numbers in the case of constant viscosity, these numbers having been calculated for the viscosity of the liquid at the film surface.

Relationships (12) reflect the influence exerted by the relationship of the viscosity to the temperature on the critical conditions for the appearance of thermocapillary convection in the nonmoving liquid layer. We present the numerical expressions of the functions  $\chi_1$ ,  $\chi_2$ :

$$N = 0; \quad 0.5; \quad 1; \quad 2; \quad 3;$$

$$\chi_1 = 1; \quad 0.86; \quad 0.65; \quad 0.41; \quad 0.26;$$

$$\chi_2 = 1; \quad 0.83; \quad 0.62; \quad 0.38; \quad 0.23.$$

Our attention is called to the strong reduction in the critical Marangoni numbers due to the reduction in the viscosity of the liquid. For example, in the case of glycerine, with

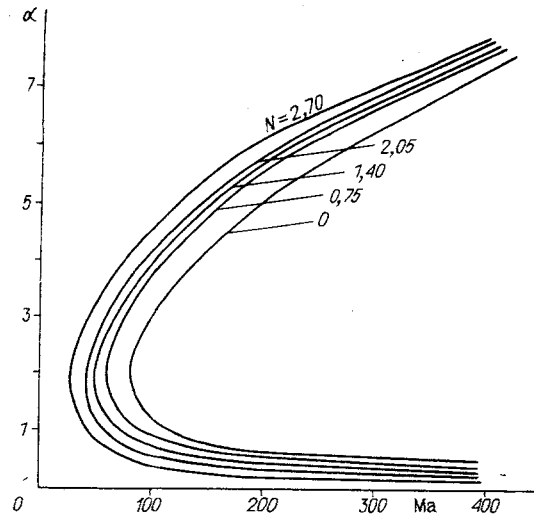


Fig. 1

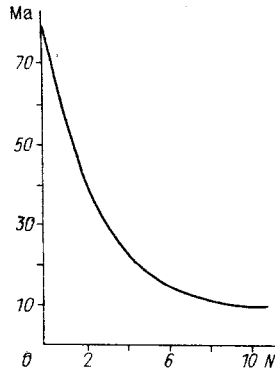


Fig. 2

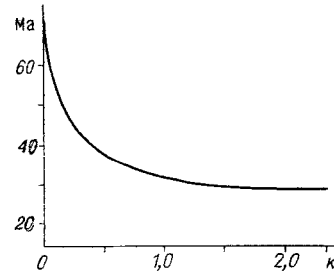


Fig. 3

the temperature drop of  $15^{\circ}\text{C}$ ,  $\beta = 0.07 \text{ deg}^{-1}$ ,  $N = 1$  the Marangoni numbers are reduced by a factor of approximately 1.5. Let us construct the neutral-stability curve over the entire range of wave numbers  $0 < \alpha < \infty$ . For this it is necessary to solve the hydrodynamic equation (9). We will seek the solution of (9) in the form of  $\omega(y) = e^{\beta_n y}$ ; substituting this expression into (9) gives us the following algebraic equation for the determination of  $\beta_n$ :

$$\beta_n^{\text{IV}} - 2\alpha^2\beta_n^2 + \alpha^4 - N^2[\beta_n^2 - \alpha^2] + 2N(\beta_n^3 - \alpha^2\beta_n) + 2N^2\beta_n^2 = 0, \quad n = 1, 2, 3, 4. \quad (13)$$

It is not difficult to find the roots of (13):

$$\beta_n = z_n - N/2, \quad z_n^2 = \alpha^2 + N^2/4 \pm N\alpha i.$$

Here  $z$  is determined by the root of the quadratic equation;  $i$  is imaginary unity. Let  $\tan \varphi_0 = N\alpha/(N^2/4 + \alpha^2)$ ,  $k = \sqrt[4]{(N^2/4 + \alpha^2) + N^2\alpha^2}$ . Then the roots of (13) will be the following:

$$\begin{aligned} \beta_1 &= k \left[ \cos \frac{\varphi_0}{2} + i \sin \frac{\varphi_0}{2} \right] - \frac{N}{2}, & \beta_2 &= k \left[ \cos \frac{\varphi_0}{2} - i \sin \frac{\varphi_0}{2} \right] - \frac{N}{2}, \\ \beta_3 &= -k \left[ \cos \frac{\varphi_0}{2} + i \sin \frac{\varphi_0}{2} \right] - \frac{N}{2}, & \beta_4 &= -k \left[ \cos \frac{\varphi_0}{2} - i \sin \frac{\varphi_0}{2} \right] - \frac{N}{2}. \end{aligned}$$

Thus the fundamental system for the solution of (9) has the form

$$\omega(\xi) = \{ e^{k_1 \xi} \cos k_2 \xi, e^{k_1 \xi} \sin k_2 \xi, e^{k_3 \xi} \cos k_2 \xi, e^{k_3 \xi} \sin k_2 \xi \},$$

where  $k_1 = k \cos \frac{\varphi_0}{2} - \frac{N}{2}$ ;  $k_2 = k \sin \frac{\varphi_0}{2}$ ;  $k_3 = -k \cos \frac{\varphi_0}{2} - \frac{N}{2}$ . Using this system and calculating the accompanying integrals, we can calculate the neutral-stability curve, the minimum critical Marangoni numbers, and the critical wave numbers (Figs. 1 and 2,  $Bi = 0.01$ ).

Since the derived solution makes no provision for the relationship between density and temperature, it is valid for small Rayleigh numbers, i.e., for rather thin layers of the liquid. In this connection let us make the following comment. As is well known [6, 11], there exists a critical film thickness  $h_*$  such that when  $h \gg h_*$  natural convection predominates, while when  $h \ll h_*$  the thermocapillary mechanism of lost instability is significant. In the intermediate region both mechanisms of stability loss for the liquid layer are active, in the case in which the liquid layer is heated from below. It would be interesting to evaluate the critical values of the film thickness  $h_{**}$  at which the influence of the relationship between viscosity and temperature is significant.

Let us introduce the dimensionless parameter  $K = Ne^N/Ma = \mu_w \beta a / \epsilon h$ , characterizing the relationship of the two destabilizing effects: the extent to which viscosity and surface tension are dependent on temperature. Using the calculations for the critical Marangoni numbers, we will construct the function  $Ma^*(K)$  (Fig. 3), analysis of which demonstrates that even when  $K = 0.05$  there is a significant drop (of up to 30%) in the critical Marangoni numbers. The film thicknesses on the order of  $h_{**} = \mu_w \beta a / \epsilon K$  correspond to this region of change in  $K$ , from which it follows that with an increase in the viscosity of the liquid and in the parameter  $\beta$ , proportional to the activation energy of the viscous flow, the critical film thicknesses increase. Therefore, the significant reduction in the minimum Marangoni numbers due to reduction in viscosity with a rise in temperature is characteristic of liquids with large Prandtl numbers. For example, in the case of glycerine  $T = 20^\circ\text{C}$ ,  $Pr = 12,490$ ,  $h_{**} = 4.8$  mm, for MS-20 oil  $T = 30^\circ\text{C}$ ,  $Pr = 7300$ ,  $h_{**} = 1.7$  mm, for methanol  $T = 25^\circ\text{C}$ ,  $Pr = 6$ ,  $h_{**} = 0.28 \cdot 10^{-3}$  mm, for toluene  $T = 25^\circ\text{C}$ ,  $Pr = 9$ ,  $h_{**} = 0.16 \cdot 10^{-3}$  mm, while for water  $T = 20^\circ\text{C}$ ,  $Pr = 6$ ,  $h_{**} = 0.3 \cdot 10^{-3}$  mm. The quantity  $h_{**}$  is on the order of 1-5 mm and for conditions at the surface the mechanism for reducing stability as a consequence of a reduction in viscosity therefore always acts on liquids with larger Prandtl numbers ( $Pr > 100$ ), while for liquids with moderate Prandtl numbers (organic fluids, water) the destabilizing effect of variability in viscosity is insignificant.

A related problem dealing with the effect on Marangoni convection as a consequence of a change in viscosity, dependent on temperature, was covered in [12].

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## STABILITY IN THE SHEAR LAYER OF A COMPRESSIBLE GAS

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In the mixing of two parallel streams of a viscous gas, moving at different velocities, near the boundary of separation a flow is formed that is referred to as an ordinary free shear layer. Such flows in actual practice are encountered in the boundary layer of a jet discharging into a submerged space, in the wake trailing a nonsymmetrically streamlined body, etc. The free shear flows are extremely unstable to small perturbations, i.e., the shear layer of an incompressible gas, for example, is unstable for all Reynolds numbers  $Re$  [1]. The stability of the compressible shear layer in the case of finite  $Re$  has, apparently, not been studied earlier. Without provision for viscosity, this problem is solved in [2-4], with a number of additional simplifications having been introduced: the temperature throughout the entire flow was assumed to be constant and the dynamic profile was given by the function  $U(y) = \tanh y$ .

In the present study in investigating the stability of the compressible shear layer we assume the gas to be viscous and capable of conducting heat, with the velocity and temperature profiles calculated from corresponding boundary-layer equations [5]. Approximations of an incompressible or inviscid gas are thereby attained in the form of limit cases in which the Mach number  $M \rightarrow 0$  or  $Re \rightarrow \infty$ . The calculations were performed numerically by the orthogonalization method [6]. It is demonstrated that when  $M \leq 1$  the stability of the flow is determined by wave perturbations exhibiting a phase velocity  $c_r = 0$  and a zero critical Reynolds number  $Re_{*}$ . With an increase in  $M$  the region of unstable wave numbers narrows. When  $M \geq 1$ , as in the case of the inviscid problem [3], stability is determined by traveling waves with  $c_r \neq 0$  (the second perturbation mode). It has been observed that for the second mode  $Re_{*}$  is different from zero and diminishes as  $M$  increases. We have constructed the neutral stability curves, the eigenfunctions, and we have studied the relationship between the characteristics of stability and  $M$  for the case in which  $0 \leq M \leq 2$ .

1. Let us examine the plane flow in the shear layer of a compressible viscous heat-conducting gas. We will assume the gas to be ideal, with constant heat capacities  $c_V$  and  $c_P = \gamma c_V$ , viscosity  $\mu$ , and thermal conductivity  $k$  directly proportional to temperature, so that the Prandtl number  $Pr = \mu c_P / k$  is constant and that the second viscosity is equal to zero. The Navier-Stokes equations, written in dimensionless form, in this case have the form

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \frac{\partial(\rho u_i)}{\partial x_i} &= 0, \quad \rho \left( \frac{\partial u_i}{\partial t} + u_j \frac{\partial u_i}{\partial x_j} \right) + \frac{\partial p}{\partial x_i} = \frac{1}{Re} \frac{\partial \sigma_{ij}}{\partial x_j}, \\ \rho \left( \frac{\partial \theta}{\partial t} + u_j \frac{\partial \theta}{\partial x_j} \right) + (\gamma - 1) \rho \theta \frac{\partial u_j}{\partial x_j} &= \frac{\gamma}{Re Pr} \frac{\partial}{\partial x_j} \left( \mu \frac{\partial \theta}{\partial x_j} \right) + \frac{\gamma(\gamma - 1) M^2}{Re} \sigma_{ij} e_{ij}, \\ \mu(\theta) &= \theta, \quad p = \rho \theta / \gamma M^2, \\ \sigma_{ij} &= 2\mu e_{ij} - \frac{2}{3} \mu e_{kk} \delta_{ij}, \quad e_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right), \quad i, j, k = 1, 2, \\ M &= U_* / \sqrt{\gamma R T_*}, \quad Re = \rho_* U_* \delta / \mu_*, \quad \delta = (\pi \mu X / \rho_* U_*)^{1/2}. \end{aligned} \quad (1.1)$$

Here  $u_1 \equiv u$  and  $u_2 \equiv v$  are the longitudinal and transverse components of velocity in the direction of the  $x_1 \equiv x$  and  $x_2 \equiv y$  axes, respectively;  $\rho$ ,  $p$ , and  $\theta$  are the density, pressure, and temperature of the gas;  $R$  is the gas constant. The region in which the independent variables  $x$  and  $y$  change represents the entire plane  $-\infty < x < \infty$ ,  $-\infty < y < \infty$ . We have

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