CONVECTIVE INSTABILITY IN A TWO-LAYER SYSTEM WITH AN INTERFACIAL REACTION

L. M. Rabinovich and I. A. Belikova

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Linear analysis of convective stability was carried out for a system with two liquid layers of finite thickness in the presence of the concentration—capillary effect and a second-order interfacial reaction. It was assumed that the surface tension at the liquid—liquid interface depends on the concentrations of the reaction components.

As a result of the Marangoni effect [1], the dependence of the surface tension at the boundary between two immiscible liquids on the concentration of a diffusing substance can lead to concentration—capillary convection [2], in the presence of the mass flux across the interface. Chemical reactions which take place in the two phases in contact affect the values of concentration gradients and corresponding mass fluxes. The interfacial reactions, on the other hand, change the mass balance and the effective value of the surface tension. This change in interfacial conditions can lead to the loss of hydrodynamic stability even in the absence of the mass flux across the interface [3].

Hydrodynamic stability of a reacting liquid—liquid system is of great practical interest in the problems of enhancement of mass transfer in liquid extraction in the presence of chemical reactions, and also in the liquid phase catalytic processes in heterogeneous media.

The effect of the first order volume chemical reactions on the onset of the Marangoni instability in a liquid—liquid system, with infinite depth of the two phases, was considered first in [4] within the framework of the Sternling—Scriven model [5]. It was shown in [4] that chemical reactions can lead to convective instability in a system which is stable in the absence of reactions, even for quite small values of the rate constant. The effect of interfacial reactions, at the boundary between two liquid media of infinite depth, on the onset of the Marangoni convection has been considered in [6], also in the Sternling—Scriven approximation. In addition, it was assumed in [6] that one of the two reagents is in great excess, i.e., the reaction is quasistationary. With these assumptions, it was shown how the rate constant of the reaction affects the growth rate of perturbations and the dispersion curves. The effect of a heterogeneous second order reaction on the concentration—capillary instability in a two-phase system with the spherical interface was investigated in [7]. The linear analysis of the Marangoni instability was used to determine the considerable effect of the reaction rate constant on stability limits of the system under consideration.

We consider the convective stability of a system consisting of two liquid layers of finite depth in the presence of an interfacial reaction of the second order, such that one of the reagents and the product affect the value of the surface tension. It was found that, in the presence of two surface—active components, the instability conditions are qualitatively different from those in a system with only one surface-active component [3]. In addition, it is shown that the nature of instability in a two-layer system with an interfacial reaction depends on the diffusion coefficients in the two phases in contact.

Consider a system of two horizontal layers with a nondeformable interface at $y = 0$, bounded by solid surfaces at $y = h_1$ and $y = -h_2$, and infinite in the x and z directions. The layers $0 < y < h_1$ and $-h_2 < y < 0$ are filled with immiscible liquids in which reagents are dissolved: the reagent A in the first liquid and the reagent B in the second. A second order reaction $A + B \rightarrow C$ takes place only at the interface. It is further assumed that this reaction is irreversible; that its rate is given by Ka_sb_s, where a_s and b_s are the concentrations of reagents at the interface and K is the rate constant; that the reagents react com-

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L. Ya. Karpov Physicochemical Research Institute, Moscow. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 58, No. 6, pp. 972-979, June, 1990. Original article submitted February 22, 1989.

pletely to form the product which diffuses into the first phase; and that the reagent B and the product C affect the interfacial tension. The Rayleigh–Taylor instability of the interfacial boundary in the external mass field is neglected.

Equations of motion of the viscous incompressible liquids in the first and second phases (j = 1, 2) can be written using the stream function ψ and vorticity ω:

$$\frac{\partial \omega_j}{\partial t}+\frac{\partial \psi_j}{\partial y}\frac{\partial \omega_j}{\partial x}-\frac{\partial \psi_j}{\partial x}\frac{\partial \omega_j}{\partial y}=\nu_j\Delta\omega_j,$$
$$\omega_j=\Delta\psi_j,\quad \Delta=\partial^2/\partial x^2+\partial^2/\partial y^2. \tag{1}$$

Concentrations of the components A and C in the first phase, and of B in the second phase, are determined by the convection–diffusion equations:

$$\frac{\partial c_n}{\partial t}+\frac{\partial \psi_j}{\partial y}\frac{\partial c_n}{\partial x}-\frac{\partial \psi_j}{\partial x}\frac{\partial c_n}{\partial y}=D_n\Delta c_n\quad (n=1,\ 2,\ 3), \tag{2}$$

where $c_1 = a$; $c_2 = b$; $c_3 = c$; $j = 1$ for $n = 1, 3$ and $j = 2$ for $n = 2$; and $D_1 = D_a$, $D_2 = D_b$, $D_3 = D_c$.

Solutions of Eqs. (1)-(2) must satisfy the following boundary conditions at the flat interface at $y = 0$ and solid surfaces at $y = h_1$ and $y = -h_2$:

$$y=0:\quad \frac{\partial \psi_j}{\partial x}=0,\quad \frac{\partial \psi_1}{\partial y}=\frac{\partial \psi_2}{\partial y}, \tag{3}$$

$$y=0:\quad \left(\frac{\partial^2}{\partial y^2}-\frac{\partial^2}{\partial x^2}\right)(\mu_2\psi_2-\mu_1\psi_1)=\frac{\partial \sigma}{\partial x}, \tag{4}$$

$$y=0:\quad \sigma=\sigma_0-f_b b-f_c c, \tag{5}$$

$$y=0:\quad D_a\frac{\partial a}{\partial y}=-D_b\frac{\partial b}{\partial y}=-D_c\frac{\partial c}{\partial y}=Kab, \tag{6}$$

$$y=h_1:\quad \frac{\partial \psi_1}{\partial x}=\frac{\partial \psi_1}{\partial y}=0,\quad a=a_0,\quad c=0, \tag{7}$$

$$y=-h_2:\quad \frac{\partial \psi_2}{\partial x}=\frac{\partial \psi_2}{\partial y}=0,\quad b=b_0. \tag{8}$$

Condition (4) expresses the continuity of tangential stresses at the interface, and condition (6) the mass conservation at the interface. The continuity of normal stresses at the interface is not considered since, for simplicity, σ is assumed to be sufficiently large so that the interface remains flat even in the perturbed state.

We introduce the following dimensionless variables and parameters:

$$x^*=\frac{x}{h_1},\quad y^*=\frac{y}{h_1},\quad t^*=\frac{D_a t}{h_1^2},\quad \psi_j^*=\frac{\psi_j}{D_a},$$
$$\omega_j^*=\frac{h_1^2\omega_j}{D_a};\quad \mu=\frac{\mu_1}{\mu_2},\quad c_n^*=\frac{c_n}{a_0},$$
$$h=\frac{h_2}{h_1},\quad d=\frac{D_c}{D_b},\quad d_1=\frac{D_b}{D_a},$$
$$\mathrm{M}_b=\frac{f_b b_0 h_2}{\mu_2 D_b},\quad \mathrm{M}_c=\frac{f_c a_0 h_1}{\mu_1 D_c},\quad \mathrm{T}=\frac{K a_0 h_1}{D_a}. \tag{9}$$

In the following the asterisk used to denote dimensionless variables will be omitted.

The system of equations (1)-(8) has, in general, two trivial stationary solutions:

$$\omega_j^{(s)} = \psi_j^{(s)} = 0, \quad a^{(s)} = l\,(y-1)+1,$$

$$b^{(s)} = -\frac{l}{d_1}(y+h)+\frac{b_0}{a_0}, \quad c^{(s)} = \frac{l}{dd_1}(1-y),$$

$$l = \frac{Rd_1}{2h} \pm \left[\left(\frac{Rd_1}{2h}\right)^2 - \frac{d_1 b_0}{ha_0}\right]^{1/2},$$

$$R = \frac{b_0}{a_0}+\frac{h}{d_1}+\frac{1}{\mathrm{T}}. \tag{10}$$

In practical calculations, the sign in the expression for ℓ is chosen so that concentrations at the interface are positive.

Consider the stability of the stationary solution (10) with respect to small perturbations of the stream function and concentrations, which are represented as the Fourier components with a dimensionless, real and positive wave number α and a dimensionless complex frequency β:

$$\psi_j = \Psi_j(y)\exp(i\alpha x+\beta t), \quad c_n - c_n^{(s)} = C_n(y)\exp(i\alpha x+\beta t),$$

where i is the imaginary unit.

Linearizing Eqs. (1)-(2) we obtain:

$$\Psi_j^{\mathrm{IV}} - (\alpha^2+\gamma_j^2)\Psi_j'' + \alpha^2\gamma_j^2\Psi_j = 0, \tag{11}$$

$$C_n'' - \lambda_n^2 C_n + r_n\Psi_m = 0, \tag{12}$$

$$\gamma_j^2 = \alpha^2 + D_b\nu_j^{-1}\beta, \ \lambda_n^2 = \alpha^2 + D_b D_n^{-1}\beta,$$

$$r_n = i\alpha D_b D_n^{-1}(\partial c_n^{(s)}/\partial y),$$

where $D_1 = D_a$; $D_2 = D_b$; $D_3 = D_c$; $c_1^{(s)} = a^{(s)}$; $c_2^{(s)} = b^{(s)}$; $c_3^{(s)} = c^{(s)}$; in Eq. (12) and below m = 1 for n = 1, 3 and m = 2 for n = 2.

The solutions of Eqs. (11) and (12) are of the form

$$\Psi_j = A_j\exp(-\gamma_j y) + B_j\exp(\gamma_j y) + E_j\exp(-\alpha y) + F_j\exp(\alpha y), \tag{13}$$

$$C_n = G_n\exp(\lambda_n y) + H_n\exp(-\lambda_n y) - r_n Q_n(y), \tag{14}$$

$$Q_n(y) = \frac{A_m\exp(-\gamma_m y) + B_m\exp(\gamma_m y)}{\gamma_m^2-\lambda_n^2} + \frac{E_m\exp(-\alpha y) + F_m\exp(\alpha y)}{\alpha^2-\lambda_n^2}.$$

Substituting Eqs. (13) and (14) into the dimensionless boundary conditions (3)-(8) we obtain a set of fourteen algebraic equations for the constants A_1, A_2, B_1, B_2, E_1, E_2, F_1, F_2, G_1, G_2, G_3, H_1, H_2, H_3. This set of equations has nonzero solutions only if the determinant vanishes. The characteristic equation of this set, which can be written as $\mathcal{F}(\alpha, \beta, M_b, M_c, T, h, d, \ldots) = 0$, determines the dispersion relations which relate the wave characteristics of the perturbation to all physicochemical parameters.

Consider neutral stability for perturbations with $\alpha \neq 0$, $\beta = 0$. In this case the solutions of Eqs. (11) and (12) are of the following form:

$$\Psi_j = A_j'\exp(\alpha y) + B_j' y\exp(\alpha y) + E_j'\exp(-\alpha y) + F_j' y\exp(-\alpha y), \tag{15}$$

$$C_n = G_n'\exp(\alpha y) + H_n'\exp(-\alpha y) - r_n Q_n'(y), \tag{16}$$

$$Q_n'(y) = [2A_m' y + (y^2 - y/\alpha)B_m'](\exp(\alpha y)/4\alpha) +$$

$$+ [(2y+1/\alpha)A_m' - (y^2 + y/\alpha + 1/2\alpha^2)F_m'](\exp(-\alpha y)/4\alpha).$$

Substituting (15) and (16) into the boundary conditions, and taking into account that from (3) we have $E_j' = -A_j'$, we obtain a set of twelve equations for the constants A_j', B_j',

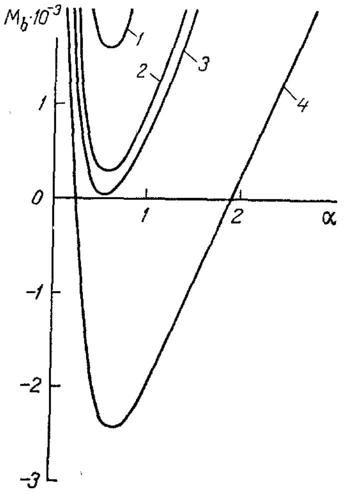

Fig. 1

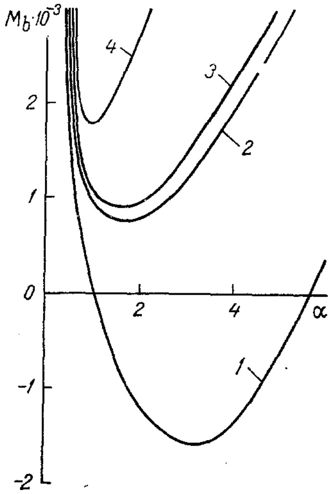

Fig. 2

Fig. 1. Neutral stability curves for $h = 1.5$, $a^* = 1.3$, $\mu = 0.5$, $d_1 = 5/6$, $M_c = 10^4$, $T = 1$: 1) $d = 0.8$; 2) 1; 3) 1.05; 4) 2.

Fig. 2. Neutral stability curves for $h = 1.5$, $a^* = 1.3$, $\mu = 1.8$, $d = 0.8$, $d_1 = 5/6$, $T = 0.8$: 1) $M_c = -10^3$; 2) 10; 3) 10^2; 4) 10^3.

F_j', G_n', H_n'. The characteristic equation is

$$\det\|a(p, q)\| = 0 \quad (p, q = 1, 2, \ldots, 12). \tag{17}$$

All the nonzero elements of the matrix $\|a(p, q)\|$ are given in the Appendix.

Dependence of the Marangoni number on the wave number of the perturbation, for different values of physical parameters, is found by numerical solutions of Eq. (17). These solutions also yield the neutral stability curves, which separate the regions of stability ($\beta < 0$) and instability ($\beta > 0$). Extrema of these curves correspond to the critical values of the Marangoni number, which determine the threshold of instability, and also to the wavelength of the perturbation which grows as the Marangoni number is changed. Such a perturbation determines to a large extent that dominant mode which is responsible for the dissipative structure formed as a result of the onset of convective instability and stabilization of perturbations.

The problem of determining the neutral stability curves is the problem of finding roots of a complex-valued function given as a determinant of the twelfth order. This was done numerically using the Mueller method (parabolic method) [8], while the value of the determinant was evaluated using the standard program. Calculations were carried out in double precision.

Neutral stability curves $M_b(\alpha)$ are shown in Figs. 1-4, for different values of parameters defined in Eq. (9). It follows from the analysis of these curves that the nature of stability of the system under consideration depends considerably on the relation between the diffusion coefficients of the surface-active components. If the diffusion coefficient of the reagent B is greater than that of the product C ($d < 1$), and if C lowers the interfacial tension ($M_c > 0$), then the system is always stable if, in addition, the reagent activity is positive, i.e., the reagent increases the interfacial tension ($f_b < 0$, $M_b < 0$). In this case, the convective instability can occur only if the reagent also lowers the interfacial tension σ and its activity reaches a certain value, which increases with decreasing values of d. If, on the other hand, the diffusion coefficient of the reagent is less than that of the product ($d \gtrsim 1.06$), then the system is unstable for any negative values of the activity of B ($M_b > 0$), but is stable for sufficiently large positive values of the activity.

The family of neutral stability curves shown in Fig. 2 demonstrates the effect of the Marangoni number M_c on the nature of stability of the system if the diffusion coefficient of the reagent is greater than that of the product ($d = 0.8$). In this case, increasing the positive activity of the product ($M_c < 0$) enhances the onset of instability, while the negative activity ($M_c > 0$) and the positive activity of the reagent ($M_b < 0$) stabilize the system.

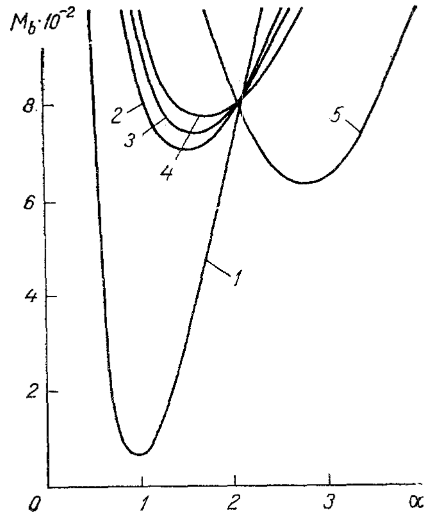

Fig. 3

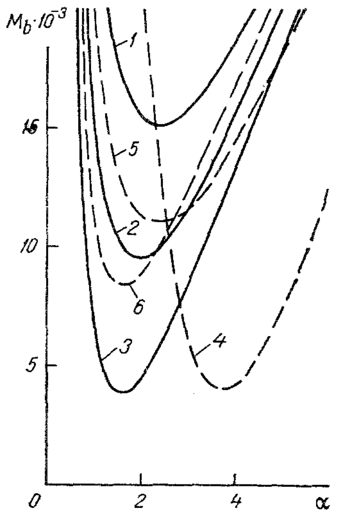

Fig. 4

Fig. 3. Neutral stability curves for $h = 1.5$, $a^* = 1.3$, $\mu = 1.8$, $d = 2$, $d_1 = 5/6$, $T = 1$; 1) $M_c = 10^3$; 2) 10^2; 3) 0; 4) -10^2, 5) -10^3.

Fig. 4. Neutral stability curves for $h = 1.5$, $a^* = 1.3$, $\mu = 1.8$, $d_1 = 5.6$, $T = 10^2$: 1) $d = 2$, $M_c = -10^2$; 2) 2 and 10; 3) 2 and 10^2; 4) 0.8 and -10^3; 5) 0.8 and -10^2; 6) 0.8 and 10^2.

If the diffusion coefficient of the product is considerably greater than that of the reagent, the nature of the stability curves changes qualitatively. As seen from the curves in Fig. 3, for $d = 2$ even the negative activity of the product enhances the onset of instability, while the positive activity can increase the stability threshold of the system. It should be noted, however, that the stabilizing influence occurs only in a certain range of values of M_c, and with an increase in the positive activity, and the corresponding decrease in M_c, the situation changes to the opposite one. For example, for $M_c = -10^4$, the critical value of the Marangoni number for the reagent M_b^* becomes negative and equals $-7.2 \cdot 10^3$, while for $M_c = -10^5$, we have $M_b^* = 1.1 \cdot 10^5$.

Comparison of curves in Figs. 3 and 4 (curves 1-3) illustrates the effect of the dimensionless rate constant of the reaction T on the convective stability threshold for $d > 1$. An increase in the rate constant of the chemical reaction leads to an increase in the critical values of the Marangoni number and, therefore, stabilizes the system. For $M_c = -10^3$, we have $M_b^* = 3.9 \cdot 10^4$ and for $M_c = 10^3$, we obtain $M_b^* = -7.1 \cdot 10^4$. Curves 4-6 in Fig. 4 illustrate that increasing the value of T has a stabilizing influence on the system even for $d < 1$.

It follows from the results obtained above that the effect of the ratio of diffusion coefficients on the Marangoni instability is in qualitative agreement with the main results on the theory by Sternling and Scriven [5], which was developed for the case of a single surface-active component, transported across the interface in the absence of chemical reactions. On the other hand, the presence of two components with surface activity, involved in a chemical reaction, can considerably diversify the regimes of concentration—capillary instability in a two-layer system.

In conclusion, we note that the onset of convective instability of the surface separating two phases leads to a sharp increase in the rate of mass transfer. We described a mechanism of hydrochemical interaction and found conditions under which such an instability may be due to chemical reactions. Thus, one possible method of enhancing transfer processes in industrial gas—liquid systems is to use chemisorbents, extraction agents, and solvents, with physicochemical properties leading to interfacial instability. Another method is to introduce appropriate reagents and surface-active substances.

APPENDIX

The following are all the nonzero elements of the matrix $\|a(p, q)\|$ in Eq. (17):

$$a(1,\ 1) = 2\operatorname{sh}\alpha,\ a(1,\ 2) = a(9,\ 7) = a(11,\ 11) = e_1,$$

$$a(1,\ 3) = a(9,\ 8) = a(11,\ 12) = e_2,\ a(2,\ 1) = 2\alpha\operatorname{ch}\alpha,$$

$$a(2,\ 2) = (1+\alpha)e_1,\ a(2,\ 3) = (1-\alpha)e_2,\ a(3,\ 4) = 2\operatorname{sh}(h\alpha),$$

$$a(3,\ 5) = he_4,\ a(3,\ 6) = he_3,\ a(4,\ 4) = 2\alpha\operatorname{ch}(h\alpha),$$

$$a(4,\ 5) = (1-\alpha h)e_4,\ a(4,\ 6) = (1+\alpha h)e_3,$$

$$a(5,\ 1) = -a(5,\ 4) = 2\alpha,\ a(5,\ 2) = a(5,\ 3) = -a(5,\ 5) =$$

$$= -a(5,\ 6) = 1,\ a(6,\ 1) = -(3/4)il,$$

$$a(6,\ 2) = 2a(6,\ 3) = (d_1/3\alpha)a(6,\ 4) = il/4\alpha,\ a(6,7) = -a(6,8) = \alpha/d_1,$$

$$2a(6,\ 5) = 4a(6,\ 6) = -da(7,\ 2) = -2da(7,\ 3) =$$

$$= -(1/3\alpha)a(7,\ 4) = a(7,\ 5) = 2a(7,\ 6) = da(8,\ 2) = -il/4d_1\alpha,$$

$$a(6,\ 9) = -a(6,\ 10) = a(7,\ 9) = -a(7,\ 10) = -(1/2)a(12,\ 5) = \alpha,$$

$$a(7,\ 11) = -a(7,\ 12) = a(8,\ 11) = -a(8,\ 12) = \alpha d,$$

$$a(8,\ 1) = (3/4)il/dd_1 - i\mathrm{T}\,ld_1e_5/4\alpha,$$

$$a(8,\ 4) = -2a(8,\ 6) = i\mathrm{T}(1-l)\,l/4d_1\alpha,\ a(8,\ 3) = -il/8dd_1\alpha +$$

$$+ i\mathrm{T}\,ld_1e_5/8\alpha^2,\ a(8,\ 7) = a(8,\ 8) = \mathrm{T}\,e_5,$$

$$a(8,\ 9) = a(8,\ 10) = \mathrm{T}(1-l),\ a(9,\ 1) = -ild_1[\operatorname{ch}\alpha + (1/4\alpha)e_2],$$

$$a(9,\ 2) = (i/4)\,ld_1e_1(1/\alpha - 1),\ a(10,\ 9) = e_4,\ a(10,\ 10) = e_3,$$

$$a(9,\ 3) = (i/4)\,ld_1e_2(1 + 1/\alpha + 1/2\alpha^2),$$

$$a(10,\ 4) = -i(l/d_1)[h\operatorname{ch}(\alpha h) + e_3/4\alpha],$$

$$a(10,\ 6) = -(le_3/4d_1)(h^2 - h/\alpha + 1/2\alpha^2),$$

$$a(11,\ 1) = i(l/d^2d_1)(\operatorname{ch}\alpha + d_1e_2/4d\alpha),$$

$$a(11,\ 2) = i(le_1/4d^2d_1)(1 - 1/\alpha),$$

$$a(11,\ 3) = -i(le_2/4d^2d_1)(1 + 1/\alpha + 1/2\alpha^2),\ a(12,\ 2) = 2\mu\alpha,$$

$$a(12,\ 1) = \mu l\,\mathrm{M}_c/4dd_1,\ a(12,\ 3) = -2\mu\alpha - \mu l\,\mathrm{M}_c/8dd_1\alpha,$$

$$a(12,\ 4) = l\,\mathrm{M}_b/4hd_1,\ a(12,\ 6) = 2\alpha - l\,\mathrm{M}_b/8hd_1\alpha,$$

$$a(12,\ 9) = (12,\ 10) = -i\alpha\,\mathrm{M}_b/h,\ a(12,\ 11) = a(12,\ 12) = -\mu d\,\mathrm{M}_c\alpha,$$

$$e_1 = \exp\alpha,\ e_2 = \exp(-\alpha),$$

$$e_3 = \exp(h\alpha),\ e_4 = \exp(-h\alpha),$$

$$e_5 = b_0/a_0 - lh/d_1.$$

NOTATION

t, time; h_1 and h_2, widths of the liquid layers; ν, μ, kinematic and dynamic viscosities; a, b, and c, concentrations of components A, B, and C; a_0 and b_0, concentrations at the bounding surfaces; D_a, D_b, and D_c, diffusion coefficients of components A, B, and C; σ, surface tension at the liquid–liquid interface; f_b and f_c, coefficients of surface activity of components B and C; M_b and M_c, Marangoni numbers for the reagent B and the product C, respectively; T, dimensionless rate constant of the reaction.

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NATURAL CONVECTION IN A LONG RECTANGULAR CAVITY

K. P. Morgunov, T. Yu. Morgunova, and V. A. Misyura

UDC 536.25

We discuss an approximate analytical method of calculating the parameters of the motion of a gas in a long cavity induced by the presence of a heated vertical wall. Assuming the flow is plane-parallel and the longitudinal temperature gradient in the central region of the flow is constant, we obtain analytical expressions for the velocity and temperature profiles. We use the law of conservation of energy in integral form to match the solution in the central region with the end regions near the walls, and thereby obtain the flow parameters without considering the structure of the flow in the end regions.

Introduction. The structure of the flow in a closed cavity containing a gas is determined by the temperature boundary conditions on the cavity walls. Previous studies of natural convection in cavities have been concerned mainly with small to moderate values of the ratio of the horizontal dimension L of the cavity to its vertical dimension H and comparatively little attention has been paid to flow in long horizontal cavities in the presence of a temperature gradient along the axis of the cavity. In [1-3] a two-dimensional rectangular closed cavity was considered whose length L was much larger than its height H, while the vertical walls were maintained at different constant temperatures. It was assumed that at a certain distance from the heated wall the parameters describing the flow vary much more rapidly in the transverse direction than along the cavity axis. With this assumption a relatively simple solution of the Navier–Stokes equations in the Boussinesq approximation can be obtained. We will assume that a solution of this type is correct for gas flow in a region sufficiently far from the vertical walls; this region is called the central flow region. The basic problem is to explain the effect of the conditions on the vertical walls of the cavity on the form of the solution in the central region.

We discuss below an approach to the matching of the flow in the central region to the flow near the vertical walls. The method is based on the integral conservation laws of continuum mechanics.

We consider a two-dimensional rectangular closed cavity containing a gas. The length L of the cavity is much larger than the height H (Fig. 1). The horizontal walls of the cavity are assumed to be adiabatic. One of the vertical walls is isothermal with temperature T_0. Two types of conditions are considered for the other vertical wall: the wall is isothermal with temperature T_H, greater than T_0; the specific heat flux q through the wall is specified.

The equations describing the steady laminar flow of a viscous incompressible liquid or gas in a horizontal cavity are, in dimensionless variables:

$$\frac{\partial u}{\partial x}+\frac{\partial v}{\partial y}=0; \tag{1}$$

$$\frac{1}{\mathrm{Pr}}\left(u\frac{\partial u}{\partial x}+v\frac{\partial u}{\partial y}\right)=-\frac{\partial p}{\partial x}+\frac{\partial^2 u}{\partial x^2}+\frac{\partial^2 u}{\partial y^2}\,; \tag{2}$$

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