

DEVELOPMENT OF THERMOGRAVITATION CONVECTION IN A TWO-LAYER SYSTEM
 IN THE PRESENCE OF A SURFACE-ACTIVE MATERIAL ON THE BOUNDARY

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UDC 536.25

Convective instability of equilibrium in a system of two horizontal layers of immiscible liquids, caused by the Rayleigh instability mechanism, has been studied within the framework of the linear theory in [1-5]. The present study will investigate the effect of a surface-active material (SAM), deposited on the boundary between the liquids, on the development of thermogravitation convection. Calculations were performed for two types of systems, which in the absence of a SAM show instability of a monotonic or an oscillatory character. A new type of oscillatory equilibrium instability was observed, produced by the effect of the SAM. In some region of parameter values the oscillatory instability may prove to be the more dangerous one. The action of the Marangoni effect on thermogravitation oscillations is considered.

1. Let the space between two solid horizontal plates on which constant but different temperatures are maintained (temperature difference equal to Θ) be filled by two layers of viscous immiscible liquid. The origin of the coordinate system is located on the boundary between the layers; the x axis is directed horizontally, and the y axis vertically upward. The solid boundaries are described by the equations $y = a$ and $y = -a_2$. The dynamic and kinematic viscosity, thermal conductivity, thermal diffusivity, and volume expansion coefficients will be denoted by η_m , ν_m , κ_m , χ_m , β_m ($m = 1$ for the upper liquid, 2 for the lower). The effect of curvature of the separating boundary will not be considered since for thermogravitation convection it is insignificant [6]; the boundary is assumed planar and undeformed ($y = 0$). We assume that the SAM is concentrated on the boundary with a surface concentration $\Gamma(x)$. Reduction in the surface tension coefficient with increase in temperature and SAM concentration is described by the expression $\sigma = \sigma_0 - \alpha T - \alpha_S \Gamma$.

We assume that the SAM concentration is low, so that its molecules form a "surface gas." SAM adsorption and desorption phenomena will not be considered. Transport of the SAM along the boundary is described by the equation [7]

$$\frac{\partial \Gamma}{\partial t} + \frac{\partial}{\partial x} (v_x \Gamma) = D_0 \frac{\partial^2 \Gamma}{\partial x^2} \quad (1.1)$$

where v_x is the liquid velocity on the boundary, D_0 is the SAM surface diffusion coefficient. In equilibrium the SAM concentration on the surface is constant: $\Gamma = \Gamma_0$.

We introduce the notation: $\eta = \eta_1/\eta_2$, $\nu = \nu_1/\nu_2$, $\kappa = \kappa_1/\kappa_2$, $\chi = \chi_1/\chi_2$, $\beta = \beta_1/\beta_2$, $\alpha = a_2/a_1$. For the units of length, time, velocity, temperature, and surface concentration we choose the values a_1 , a_1^2/ν_1 , ν_1/a_1 , Θ , and Γ_0 . The dimensionless temperature gradient dT_0/dy in equilibrium is equal to $A_1 = -s/(1 + \kappa\alpha)$ in the upper liquid and $A_2 = -s\kappa/(1 + \kappa\alpha)$ in the lower, where $s = -1$ for heating from above and $s = 1$ for heating from below.

We will now consider the stability of equilibrium. We impose on the equilibrium state perturbations of the flow function ψ_m' , the temperature T_m' , and the SAM concentration Γ' :

$$\begin{aligned} (\psi_1', T_1', \psi_2', T_2', \Gamma') = & (\psi_1(y), T_1(y), \psi_2(y), T_2(y), \Gamma) \times \\ & \times \exp [ikx - (\lambda + i\omega)t]. \end{aligned} \quad (1.2)$$

Here k is the wave number; $\lambda + i\omega$ is the complex decrement.

The linearized convection equations have the form

$$\begin{aligned}(\lambda + i\omega)D\psi_m &= -d_m D^2\psi_m + ik \text{Gr} b_m T_m, \\ -(\lambda + i\omega) T_m - ik\psi_m A_m &= \frac{c_m}{\text{Pr}} DT_m,\end{aligned}\tag{1.3}$$

where $D = d^2/dy^2 - k^2$, $b_1 = d_1 = c_1 = 1$, $b_2 = 1/\beta$, $d_2 = 1/\nu$, $c_2 = 1/\chi$, $\text{Pr} = \nu_1/\chi_1$ is the Prandtl number; $\text{Gr} = g\beta_1\theta\alpha_1^3/\nu_1^2$ is the Grashof number.

Denoting differentiation with respect to y by a prime, we write the conditions on the solid boundaries:

$$y = 1: \psi_1 = \psi_1' = T_1 = 0, \quad y = -a: \psi_2 = \psi_2' = T_2 = 0;\tag{1.4}$$

and the boundary separating the liquids:

$$y = 0: \psi_1 = \psi_2 = 0, \quad \psi_1' = \psi_2', \quad T_1 = T_2, \quad \kappa T_1' = T_2';\tag{1.5}$$

$$\eta\psi_1'' - ik(\text{Mr} T_1 + B\Gamma) = \psi_2'', \quad \text{Mr} = \frac{\alpha\theta a_1}{\eta_2\nu_1}, \quad B = \frac{\alpha_s\Gamma_0 a_1}{\eta_2\nu_1}.\tag{1.6}$$

After dedimensionalization and linearization we write Eq. (1.1) as

$$(\lambda + i\omega - D_s k^2)\Gamma = ik\psi_1'(0) \quad (D_s = D_0/\nu_1).\tag{1.7}$$

Eliminating Γ from Eqs. (1.6) and (1.7), we obtain the boundary condition

$$y = 0: \eta\psi_1'' - ik\left(\text{Mr} T_1 + \frac{ikB}{\lambda - D_s k^2 + i\omega} \psi_1'\right) = \psi_2''.\tag{1.8}$$

The equilibrium stability condition is defined by the condition $\lambda = 0$.

2. The boundary problem of Eqs. (1.3)-(1.5), (1.8) describes development of convection in a system with SAM on the boundary with simultaneous action of Rayleigh and thermocapillary instability mechanisms. Depending on the ratio of the parameters Mr and Gr , either instability mechanism may be dominant. The effect of a SAM on thermocapillary instability ($\text{Gr} = 0$) was studied in [8-11]. In the present study we will consider the situation in which the Rayleigh mechanism dominates. The boundary problem can be solved by the Runge-Kutta method.

It is known that, in the absence of SAM ($B = 0$) convective equilibrium instability can set in in both monotonic and oscillatory manners [4, 5]. The possibility of oscillatory instability follows from the fact that at $K = \beta\eta\chi/\nu \neq 1$ the boundary problem is non-self-conjugate [12]. Nevertheless, in the absence of SAM monotonic equilibrium is more typical; at present, oscillatory instability has been observed only in the system transformer oil-formic acid over a limited interval of layer thickness ratios a , while for this system, too, the minimum of the neutral curve is realized for monotonic perturbations.

We will study the effect of a SAM on development of thermogravitation convection in the case where the instability is monotonic in the absence of SAM. As an example, we choose the system air-water with the following parameters: $\text{Pr} = 0.758$, $\eta = 0.0182$, $\nu = 15.1$, $\kappa = 0.0396$, $\chi = 138$, $\beta = 17.7$, and $a = 1$. Calculations in this section and Sec. 3 were performed for $D_s = 10^{-3}$.

We will consider a situation in which thermocapillary effects are insignificant ($\text{Mr} \ll \text{Gr}$). Then in boundary condition (1.8) we may take $\text{Mr} = 0$. Calculations show that, for the given system, convective equilibrium instability in the presence of SAM may set in in either a monotonic or an oscillatory manner. We will consider the case of monotonic instability ($\omega = 0$). Figure 1 shows monotonic neutral curves for $B = 0.09, 0.05, 0.02, 0$ (curves 1-4). The dependence of critical Grashof number Gr_* , normalized to wave number, on B is shown in Fig. 2 (curve 1). Since, as is evident from boundary condition (1.8), the characteristic dimension of the problem is the ratio B/D_s , even at $B \sim 10^{-1}$ the neutral curve approaches the limit ($B \rightarrow \infty$). This limit corresponds to replacement of boundary condition (1.8) by $\psi_1'(0) = 0$, which physically corresponds to a solid surface dividing the liquids.

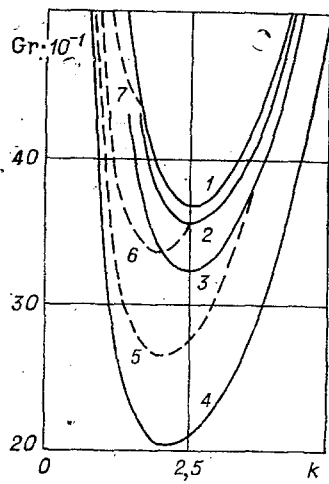


Fig. 1

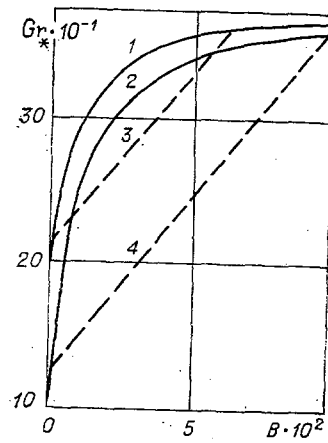


Fig. 2

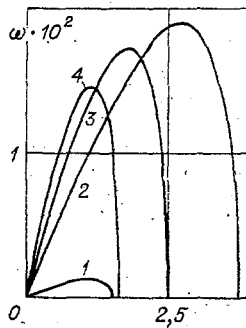


Fig. 3

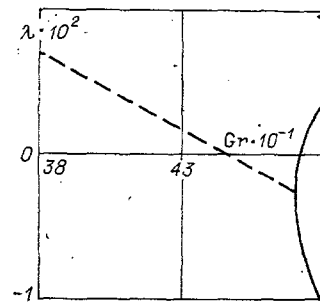


Fig. 4

Inclusion of the SAM leads to the appearance of oscillatory instability. At low B , the oscillations appear in the longwave region $0 < k < k_*(B)$. With increase in B , the termination point of the oscillatory neutral curve shifts in the shortwave direction, and when some value B_1 is exceeded, oscillatory perturbations become more dangerous. With further increase in B , $k_*(B)$ begins to decrease and, finally (at $B > B_2$), monotonic perturbations again become the more dangerous ones. We note that at $B = \infty$ (solid separation surface) the boundary problem is self-conjugate independent of the liquid parameters, so that oscillatory instability is impossible. In Fig. 1 the oscillatory neutral curves for $B = 0.02, 0.05$, and 0.09 are shown by curves 5-7. The function $Gr_*(B)$ for oscillatory instability is shown by curve 3 of Fig. 2. Curves of frequency ω as a function of wave number k for $B = 0.0005, 0.02, 0.05, 0.09$ are shown in Fig. 3 (curves 1-4). It can be shown analytically (by expansion in the parameter k), that the longwave asymptote of the oscillatory neutral curve at $Gr_0(k, B)$ at $B \neq 0$ is independent of B and coincides exactly with the asymptote of the monotonic neutral curve $Gr_m(k, 0)$ at $B = 0$:

$$\lim_{k \rightarrow 0} Gr_0(k, B) k^2 = \lim_{k \rightarrow 0} Gr_m(k, 0) k^2 = \text{const.}$$

The longwave frequency asymptote as $k \rightarrow 0$ has the form

$$\omega = k\sqrt{Bc} + o(k),$$

where the quantity c is a function of the system parameters. It should be stressed that the section of the monotonic neutral curve in the region $k < k_*(B)$ located above the oscillatory neutral curve (Fig. 1) is not the limit of monotonic instability. Figure 4 shows the dependence of the decrement λ on Gr in the region $k < k_*$ ($B = 0.09, k = 1.5$); the dashed curve corresponds to oscillatory perturbations; the solid, to monotonic. With increase in Gr , oscillatory instability develops at $Gr = Gr_0 = 44.5$; for $Gr > Gr_0 = 47.0$ the oscillation frequency vanishes and the system has two types of monotonically increasing perturbations, one of which becomes attenuating at $Gr > Gr_m = 47.1$.

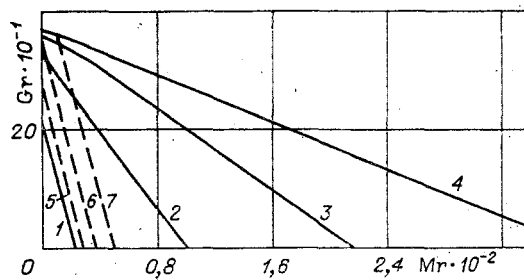


Fig. 5

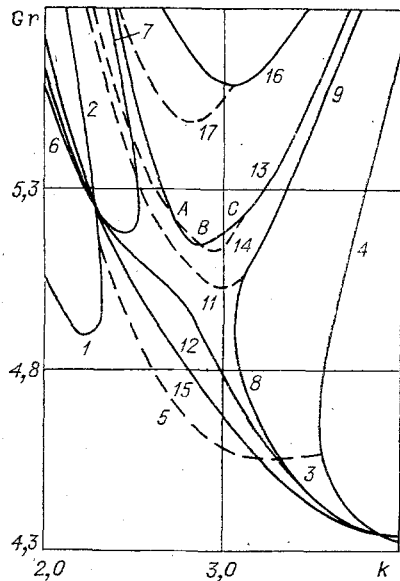


Fig. 6

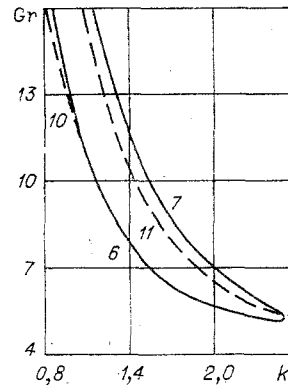


Fig. 7

We will consider the action of thermocapillary effects on the system air-water. In the absence of SAM, with increase in Mr , Gr_* decreases, although instability remains monotonic. In the presence of SAM, as in the case of purely thermogravitation convection ($Mr = 0$), two neutral curves exist - monotonic and oscillatory. Figure 2 shows curves of $Gr_*(B)$ for monotonic (curve 2) and oscillatory (curve 4) instability at $Mr = 10$. The interval of B values in which oscillatory perturbations are more dangerous increases with growth in Mr . Figure 5 shows Gr_* as a function of Mr for fixed values of B for monotonic (solid curves) and oscillatory (dashed curves) neutral curves. Curve 1 corresponds to $B = 0, 2, 5, 0.02, 3, 6, 0.05, 4, 7,$ and 0.09 .

The effect of SAM on loss of equilibrium stability was also studied for the system water-silicone oil (Dow-Corning N 200). The change in the neutral curve pattern with increase in B is similar in character.

3. We will describe the effect of SAM on development of convection in the system transformer oil-formic acid with the following parameters: $Pr = 306.32$, $\eta = 11.123$, $\nu = 15.408$, $\kappa = 0.41$, $\chi = 0.714$, $\beta = 0.672$, $a = 0.6667$.

Convective equilibrium stability has been studied for this system in the absence of SAM in [4, 5]. Outside the wave number interval $2.3 < k < 3.6$ the instability is monotonic in character, and we can distinguish stability limits at which conversion is excited predominantly in the oil layer (Fig. 6, curves 1 and 4) and the acid layer (lines 2 and 3). At the limits of the indicated interval "closing" of the monotonic neutral curves occurs, which leads to the appearance of an oscillatory neutral curve (curve 5). For low B near both monotonic instability boundaries (curves 1 and 2), corresponding to excitation of convection in the oil layer and the acid layer, in the longwave region an oscillatory instability segment develops, caused by the SAM; the mechanism by which it appears does not differ from that described in Sec. 2. With increase in B one of these segments unites with the oscillatory neutral curve which existed in the absence of SAM, forming a single oscillatory neutral

curve which existed in the absence of SAM, forming a single oscillatory neutral curve which intersects the monotonic curve. The pattern of neutral curves takes on the form shown in Figs. 6 and 7 (lines 6-11, $B = 0.05$). With further increase in B , branching of the monotonic neutral curves takes place. We will describe the neutral curve pattern at $B = 0.06$. The lower branches of both monotonic neutral curves combine, forming a single monotonic neutral curve (curve 12). The same happens to the upper branches of the neutral curves (curve 13). The oscillatory neutral curve 14 intersects the boundary of monotonic instability 13 at the points A and B, and merges with it at point C, at which the oscillation frequency vanishes. The region limited above by curve 14 and below by curve 13 (between the points A and B) represents an equilibrium "stability island." With further increase in parameter B , the neutral curve pattern simplifies (lines 15-17, $B = 0.1$).

Thus, the presence of surface-active material on the boundary between the media leads to the appearance of a specific type of oscillatory instability, which under certain conditions may prove to be the most dangerous instability.

In conclusion, the authors express their gratitude to E. M. Zhukhovitskii for his helpful evaluation.

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