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CONVECTIVE FLOWS OF BINARY MIXES IN THIN CHANNELS. THEORY AND EXPERIMENT

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Introduction

Thermal convection in a horizontal fluid layer of binary mix heated from below originates in the case of normal Sore effect as a result of instability with respect to monotonous disturbances. The same instability situation is observed for cavities of different forms that have wide horizontal boundaries. In similar cavities heated from below there is thermodiffusive division of mix components along vertical axis. So there is the opinion that the oscillatory convection in binary mix near the boundary of stability exists for only anomalous thermal diffusion when there is the competition of thermal gravitational mechanism of convection excitation and thermal diffusive one. Our preliminary experimental and theoretical results show that the oscillatory convection in binary mix is possible to be observed near the boundary of stability for normal thermodiffusion in specific conditions. According to the basic assumption explaining experiments the complex oscillatory regimes in binary mixes for positive Sore coefficient are determined by thermodiffusive division of mix components in horizontal plane when the fluid moves predominantly along vertical heat-conducting boundaries [1,2]. Long convective loop in vertical direction (connected channels) and Hele-Shaw cell are examples of the cavities in which the same flows could be observed.

In practice it is often important to take into account the presence of oscillatory regimes in binary liquids near the threshold of convection excitation. Thus, the phenomenon of thermal diffusion is applied for industrial isotopes division in vertical fractionator.

In this paper mechanisms of flow's excitation and various overcritical regimes of thermal convection in connecting channels have been investigated theoretically and experimentally for binary mixes with well-known thermodiffusive properties. Also the influence of oscillatory regimes near the boundary of stability on dopant distribution has been studied. It is found for binary mixes with normal thermal diffusion that similarly to connecting channels specific oscillatory flows take place near the threshold of convection in Hele-Shaw cell.

Experimental technique

The experimental setup (Fig. 1*a*) consists of a rectangular metal bar *1* with massive isothermal heat exchangers *2* in which a liquid circulates in order to control temperature. Thus, uniform temperature distribution across the section and linear with respect to the bar length has been created. In the bar, there are two parallel longitudinal channels with a square section and thickness $2d$ = 3.2 mm, connected at the top and at the bottom by channels of the same cross section. The height of the vertical channels is equal to H = 50 mm. The channels are closed by a transparent plastic plate which made it possible to observe the flow. The mixtures of CCl_4 in decane $C_{11}H_{22}$ and Na_2SO_4 in water have been used as working fluids. The coefficients of concentration density β_c of these mixtures are high; therefore even small concentration gradients create fairly strong non-uniformities of density which cause the onset of convection.

The mixtures were prepared in a glass flask and, before being poured into the channels, were thoroughly mixed for 10 – 15 min by intense shaking. In the experiments the flow rate was recorded by a differential thermocouple (Fig. 1*a*) with an electrode diameter of 0.1 mm installed at the center of the channels with respect to the channel height. Each junction of the thermocouple was 1.5 mm long and reached the channel center. Thus, the junction to some extent averaged the temperature across the channel. In comparing the theoretical results with the thermocouple measurements, an empirical averaging coefficient was used. The junctions of a second thermocouple, inserted into narrow drill holes in the heat exchangers, measured the vertical temperature difference ΔT. The readings of both thermocouples were determined by a V7-21 digital voltmeter and recorded on graph paper by a KSP-4 recorder. As a measure of the flow rate we used the non-dimensional parameter $\Theta = |\vartheta|/\Delta T$, where ϑ is the thermocouple reading and ΔT is the vertical temperature difference between the heat exchangers.

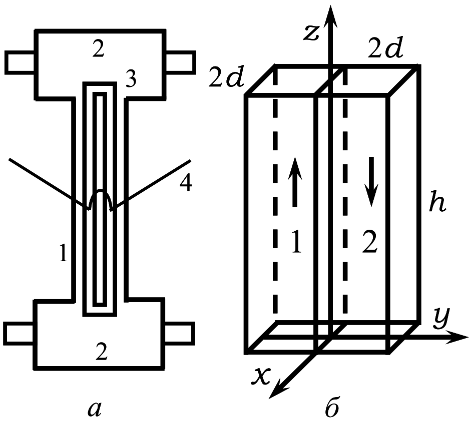

Fig. 1. Experimental setup (*a*): copper bar (*1*), heat exchangers (*2*), channels (*3*), thermocouples (*4*); coordinate system (*b*).

In pure fluids (water, decane and CCl_4) heated from below, convection develops in accordance with the known theoretical and experimental results on the convective instability of one-component Newtonian fluids. For small vertical temperature differences, these fluids are in stable mechanical equilibrium. In this case the non-dimensional parameter Θ is equal to zero. When the critical temperature difference is attained, a monotonous convective circulation flow branches softly from the equilibrium, the fluid ascending in one channel and descending in the other. The flow rate increases with the growth of temperature gradient. The critical temperature difference, at which convective circulation of the decane begins, is ΔT_0 = 1.50 ± 0.05 K, which corresponds to the critical Rayleigh number $\mathrm{Ra_{tc}} \approx 20$. The thermal Rayleigh number has been determined in terms of the temperature gradient $\Delta T/H$ as follows:

$$\mathrm{Ra_t} = \frac{g\beta_t d^4}{\nu\chi}\nabla T$$

where g is the gravity acceleration, β_t, ν, χ are thermal-expansion coefficient, kinematic viscosity and thermal diffusivity. Sometimes, instead of the Rayleigh number it was more convenient to use the supercriticality parameter $\mu_t = \mathrm{Ra_t}/\mathrm{Ra_{tc}}$. Within the limits of experimental error, the curve $\Theta = \Theta(\mu_t)$ was reproduced on both paths for increase and decrease of μ_t. In the experiments, two flow directions occurred with the same probability: one with positive and the other with negative Θ. Also the temperature deviation from a linear distribution measured along the vertical on the channel axis. These measurements were performed using groups of thermocouples with the junctions located along the channel axes.

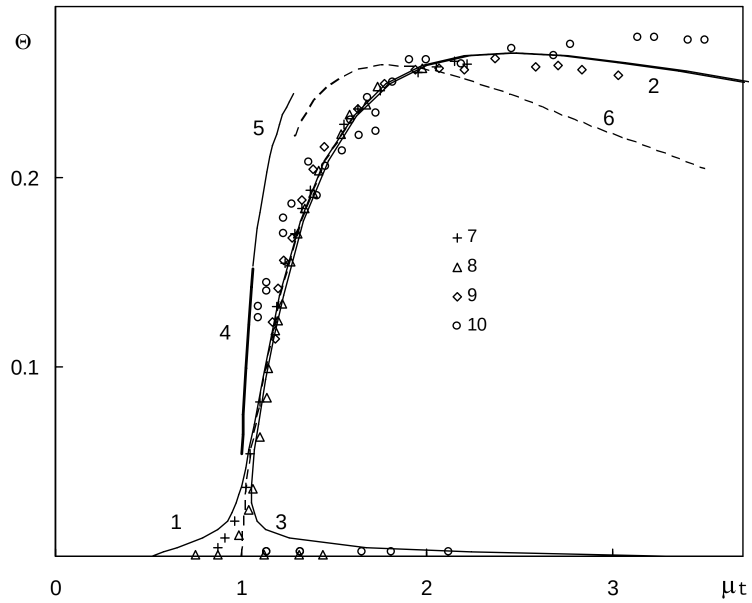

Fig. 2. Amplitude curves for binary-mixture flow in the connected channels: amplitudes of the harmonic and "flop-over" oscillations (*4*), (*5*); stationary-state regimes with and without account for thermodiffusion (*1*), (6) – $\varepsilon > 0$, (*2*) – limiting case $\varepsilon = 0$, (*3*) – $\varepsilon < 0$; experimental data (*7–10*).

When the channels are occupied by a mixture of the working fluids, the results change qualitatively. The oscillatory growth of disturbances begins in the channels when the critical Rayleigh number is attained and, depending on the supercriticality and the initial conditions, ends in either a stationary circulation or in oscillating flow with alternation of the mixture circulation direction. Experimental points for mixtures with different concentrations and theoretical amplitude curves $\Theta = \Theta(\mu_t)$ of different stationary and oscillating regimes are presented in Fig. 2. Experimental points near the abscissa axis correspond to the values of the governing parameter μ_t at which, in different realizations, "hard" transition from mechanical equilibrium to intense convection occurred. For large values of the supercriticality ($\mu_t > 1.3$), as a result of the transient process a stationary convective flow developed usually, whose intensity is shown in Fig. 2. The graph $\Theta = \Theta(\mu_t)$ has a characteristic maximum in experiment with $\Theta_{max} \approx 0.25$ that confirmed by the theory. Curves *1* and *3* correspond to the calculations [3] for stationary convection in binary mixtures with positive and negative values of thermodiffusion coefficient. For small and moderate values μ_t the process of transition from equilibrium ended in oscillations which were accompanied by a periodic change in the direction of mixture flow in the channels. These oscillations with constant amplitude were realized in the right-hand neighborhood of the critical point Ra_{tc} within a very narrow region ($\mu_t \approx 1 - 1.3$). The oscillation period turned out to be very sensitive to small variations of the supercriticality. In narrow interval $\mu_t \approx 1.1 - 1.3$ the oscillation period increased from 3 min to 1 hour. The shape of the oscillations was transformed from near-sinusoidal (Fig. 4) to near-rectangular (Fig. 5). Over non-linear oscillations with near-rectangular shape the system made regular transitions from the state with definite circulation direction to the state with the opposite one.

Thus, in experiment convective instability of equilibrium in binary liquid mixtures is related with the oscillatory growth of the initial disturbances and is accompanied by hysteresis with respect to the Rayleigh number. It is necessary to use the theory of thermoconcentration convection to explain this effect.

Equations system and non-dimensional parameters

Connected channels have rigid, heat conducting boundaries, but in the course of following calculations we neglect the thermal interaction between the left and right channel. Coordinate system with the *z*-axis directed along the channel has been presented in Fig. 1*b*. In this coordinate system $\gamma(0, 0, 1)$ is a unit vector directed vertically upward. Convective loop is heated from below so that, on the vertical channel boundaries, a linear temperature distribution is maintained. It will be shown below that, for this temperature distribution, the binary liquid can be in a state of mechanical equilibrium.

For modeling the convective flows of a binary mixture, we will use the equations for an incompressible fluid obtained in [4] on the basis of the hydrodynamic equations in the Boussinesq approximation:

$$\frac{\partial \boldsymbol{v}}{\partial t}+(\boldsymbol{v}\nabla)\boldsymbol{v}=-\frac{1}{\rho}\nabla p+\nu\Delta\boldsymbol{v}+g\left(\beta_t T+\beta_c C\right)\boldsymbol{\gamma}, \tag{1}$$

$$\frac{\partial T}{\partial t}+(\boldsymbol{v}\nabla)T=\chi\Delta T, \quad \operatorname{div}\boldsymbol{v}=0, \tag{2}$$

$$\frac{\partial C}{\partial t}+(\boldsymbol{v}\nabla)C=D\Delta C+\alpha D\Delta T, \tag{3}$$

Here, $\boldsymbol{v}$, T, p, C are the velocity, temperature, pressure, and heavy-admixture concentration fields and ρ is the mean density of the fluid. The coefficient β_c describes the dependence of the density on the concentration

$$\beta_c=-\frac{1}{\rho}\left(\frac{\partial\rho}{\partial C}\right)_{T,p}.$$

In the case considered, $\beta_c < 0$ because CCl_4 in decane and Na_2SO_4 in water are heavy admixtures. The effects associated with the presence of an admixture are also characterized by the diffusion D and thermodiffusion α coefficients. In the approximation (1) – (3), it is assumed that the diffusion and heat fluxes are related with the concentration and temperature gradients by the formulas:

$$\boldsymbol{J}=-\rho D\left(\nabla C+\alpha\nabla T\right), \quad \boldsymbol{q}=-\kappa\nabla T$$

where κ is the thermal conductivity.

The scales used for non-dimensional variables in equations (1) – (3) are: the channel half-width d for distance, d^2/ν for time, θ for temperature, $\theta\beta_t/\beta_c$ for concentration, and $\rho\nu^2/d^2$ for pressure. In terms of new non-dimensional variables, system (1) – (3) takes the form:

$$\frac{\partial \boldsymbol{v}}{\partial t}+(\boldsymbol{v}\nabla)\boldsymbol{v}=-\nabla p+\Delta\boldsymbol{v}+\frac{\mathrm{Ra}H}{\mathrm{Pr}}(T-C)\boldsymbol{\gamma}, \tag{4}$$

$$\frac{\partial T}{\partial t}+(\boldsymbol{v}\nabla)T=\frac{1}{\mathrm{Pr}}\Delta T, \quad \operatorname{div}\boldsymbol{v}=0, \tag{5}$$

$$\frac{\partial C}{\partial t}+(\boldsymbol{v}\nabla)C=\frac{1}{\mathrm{Sc}}(\Delta C+\varepsilon\Delta T). \tag{6}$$

Equations (4) – (6) contain four non-dimensional parameters, namely:

$$\mathrm{Pr}=\frac{\nu}{\chi}, \quad \mathrm{Sc}=\frac{\nu}{D}, \quad \mathrm{Ra}=\frac{g\beta_t\theta d^3}{\nu\chi}, \quad \varepsilon=\frac{\alpha\beta_c}{\beta_t}$$

Three parameters are the Prandtl, Schmidt, and Rayleigh numbers. Additional non-dimensional parameter in the problem ε characterizes the thermal diffusion in the mixture ($\alpha = k_T/T$, where k_T is the thermodiffusion ratio).

In the calculations, on the vertical channel boundaries we specified the no-slip condition $\boldsymbol{v} = 0$. The channel walls were assumed to be perfectly heat-conducting. Accordingly, on the vertical boundaries of the calculation domain the temperature disturbances were zero. Moreover, on the impermeable rigid walls the normal component of the diffusion flux density J_n vanishes. The boundary condition for non-dimensional diffusion flux density has the form:

$$\frac{\partial C}{\partial \boldsymbol{n}}+\varepsilon\frac{\partial T}{\partial \boldsymbol{n}}=0 \tag{7}$$

Also the condition of zero flux through the cross section of two channels has been imposed:

$$\iint_S \left(v_z{}^{(1)}+v_z{}^{(2)}\right)dxdy=0.$$

Here, the superscript corresponds to the channel number (Fig. 1*b*).

Mechanical equilibrium state

At a certain value of the temperature gradient, mechanical equilibrium state exists that is characterized by the absence of fluid motion (zero velocity):

$$\frac{\partial}{\partial t}=0, \quad \boldsymbol{v}=0, \quad p=p_0, \quad T=T_0, \quad C=C_0.$$

Here T_0, p_0, and C_0 are the equilibrium temperature, pressure, and admixture concentration. Applying the **curl** operator to equation (4), for a binary mixture in the state of mechanical equilibrium we obtain the system of equations

$$[\nabla T_0\times\gamma]-[\nabla C_0\times\gamma]=0, \tag{8}$$

$$\Delta T_0=0, \quad \Delta C_0=0. \tag{9}$$

In what follows, the specific case has been analyzed:

$$\nabla T_0 = -\frac{1}{H}\gamma .$$

This temperature gradient corresponds to the linear temperature distribution $T_0 = -z/H$ and heating from below. In this case, the Laplace equation for the temperature (9) is satisfied identically. The other equations of system (8) – (9) make it possible to find the equilibrium admixture distribution in the channels formed as a result of thermal diffusion. With account for boundary condition (7), on the upper and lower channel boundaries we obtain the linear vertical heavy-admixture concentration distribution $C_0 = \varepsilon z/H$.

Method of solution

In the experiments, the channel's height was greater than the width one $H >> d$. This made it possible to use the straight-trajectory approximation $\boldsymbol{v}(0, 0, u(x, y, t))$ in the calculations. Integrating equation (4) along the channels over a closed contour, we eliminate the pressure gradient. As a result, following equation takes place:

$$\frac{\partial u}{\partial t} = \Delta u + \frac{\mathrm{Ra}}{2\mathrm{Pr}}\int_0^H \left(T^{(1)} - T^{(2)}\right)dz - \frac{\mathrm{Ra}}{2\,\mathrm{Pr}}\int_0^H \left(C^{(1)} - C^{(2)}\right)dz, \tag{10}$$

where the superscripts correspond to the left and right channels, respectively. Thus, the straight-trajectory approximation results in the linearization of the Navier-Stokes equation. Then, the equations (5) and (6) are solved together with (10) using a combination of the Galerkin – Kantorovich procedure and finite-differences method. The experimental measurements show that the temperature has definite vertical distribution and Fourier analysis indicates that this distribution can be approximated by two trigonometric functions

$$T = T_1(x,y,t)\sin\left(\frac{\pi z}{H}\right) + T_2(x,y,t)\cos\left(\frac{\pi z}{H}\right).$$

It is convenient to introduce the new variable $F = C + \varepsilon T$. Taking into account the structure of the equations, the field $F(x, y, z, t)$ can be represented in the form of the expansion

$$F = F_1(x,y,t) + F_2(x,y,t)\cos\left(\frac{\pi z}{H}\right) + F_3(x,y,t)\cos\left(\frac{2\pi z}{H}\right)$$

Substituting the expansions of T and F in the original equations (5) – (6) and (10), after application of the Galerkin – Kantorovich procedure we obtain the amplitude equations for u, T_1, T_2, F_1, F_2, F_3 which are solved numerically using a finite-difference method. The algorithm was designed in accordance with the explicit solution scheme. The time derivatives and the derivatives with respect to the spatial coordinates were approximated by one-sided and central differences. The working number of grid points per channel section was 33×33. The calculations were performed using the time-relaxation method.

Discussion

In accordance with the experiments, the calculations were performed for channels with non-dimensional height $H = 30.5$. Taking into account the equal status of the channels, the results have been presented only for the left channel. In a homogeneous liquid, as the critical Rayleigh number is exceeded, convection takes place "softly". Depending on the initial disturbance shape, both upward and downward flow may develop in the channel. The situation changes radically if an admixture is present in the liquid. Since in the experiments the Schmidt number was much greater than the Prandtl number, in the calculations these parameters were taken equal to Pr = 7, Sc = 60. The thermodiffusion parameter $\varepsilon = 0.1$ corresponds to normal thermal diffusion. It follows from Fig. 2 that, in the binary liquid, development of intensive convection is “hard”, with the threshold being determined by an increase in the oscillatory disturbances. For a small supercriticality, a disturbance introduced into the fluid grows rapidly and then an oscillation regime with a certain amplitude and frequency is established. In the calculations, a sinusoidal-oscillation regime is observed on the range Ra ≈ 28 – 32. The amplitude of temperature oscillations is shown in Fig. 4. The oscillation period in dependence on the supercriticality increases. With increase in Ra, the oscillations cease being sinusoidal threshold-wise, the system goes over to the “flop-over” oscillation regime observed on the range Ra ≈ 32 – 50, see Fig. 5. With increase in μ_t, the channel flow becomes more intensive. When the concentration effects cease to play the key role, for Ra > 50, as a result stationary flow begins to be established. Also the theory predicts origination of stationary flow for $\mu_t < 1$ in the case of positive thermodiffusion coefficient. The graph with temperature amplitude “below threshold” in dependent on time is presented in Fig. 3. This stationary regime is not observed in experiment because of two reasons: first, the time of approach to it is too greater then the time of real experiment; second, the rate of this flow is negligibly small.

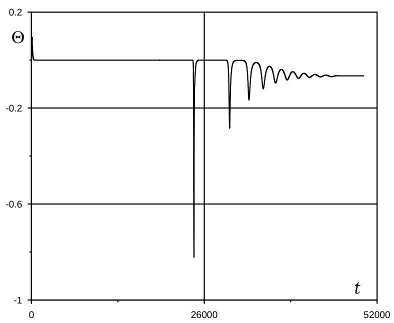

Fig. 3. The approach to the slow stationary flow over stochastic ejections

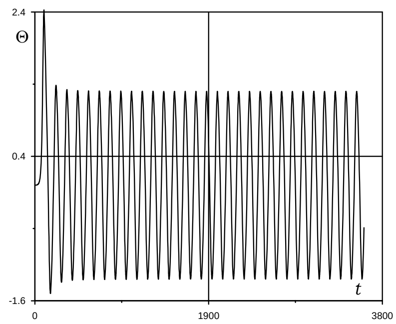

Fig. 4. Harmonic oscillations near the threshold for Ra = 29.5

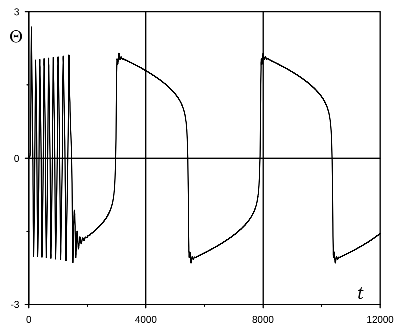

Fig. 5. Regime of periodic alternation of circulation direction

The calculations performed for positive and negative values of the coefficient ε satisfactorily describe the experimental results, which makes it possible to draw certain conclusions concerning the diffusion properties of the admixture. The mechanism responsible for the effects observed is mainly attributable to the thermodiffusion separation of the mixture which is due to the horizontal temperature gradients $\nabla_x T$, $\nabla_y T \sim \Theta/d = 3$ K/cm rather than to the weak vertical gradients $\nabla_z T = \Delta T/h \sim 0.3$ K/cm with a characteristic component separation time $h^2/D \sim 103$ hours. The horizontal gradients occur only in the circulating fluid. The separation time across the channel is $d^2/D \sim 1$ hour, which coincides in order of magnitude with the time of circulation of the fluid around the loop, i.e. a liquid particle is able to change its composition during the motion in each of the channels. For a fairly slow circulation, there is a feedback effect of the concentration non-uniformities generated by thermal diffusion on the convective flow.

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