THERMODIFFUSION SEPARATION OF A LIQUID MIXTURE UNDER DEVELOPED CONVECTION CONDITIONS

K. G. Kostarev and A. F. Pshenichnikov UDC 536.25

Gravitational convection which develops in nonisothermal cavities determines the nonisothermal cavities determines the form of concentration fields formed by thermodiffusion to a significant degree. The significant effect of convection on thermodiffusion separation appears even in horizontal layers heated from above [1]. The convection which develops in such layers due to the slightly nonisothermal state of their boundaries or a slight incline can decrease thermodiffusion separation across the layer and simultaneously produce longitudinal concentration changes several times larger than normal [2, 3]. In this case separation depends mainly on the Rayleigh diffusion number (the product of the Grashof number Gr and the Schmidt number Sc) and the dimensionless thermodiffusion parameter [4, 5]. For $Sc \cdot Gr \sim 10^5$ concentration separation reaches a medium. Further intensification of convection should lead to a smearing of the concentration fields, although the question of solution concentration inhomogeneity for $Gr \geq 10^4$ remains open. In the present study this question will be investigated experimentally, using the example of thermal convection in the gap between two horizontal coaxial cylinders. The inner cylinder has the higher temperature. Thermal convection of a one-component liquid in such a cavity has been studied thoroughly [6]. Its characteristic feature is the presence beneath the inner cylinder of a stagnation zone with very slow ("creeping") motion. This zone has stable temperature stratification, like a horizontal layer heated from above, but in contrast to the latter case the convective motion inside the zone is not independent, but "penetrates" from the region of developed convection. Thus, the temperature and concentration fields in the stagnation zone are determined by the character and intensity of the convective motion in the middle portion of the cavity.

The experimental model consisted of two coaxial brass cylinders with faces bounded by plane semitransparent mirrors. The radius of the internal cylinder was 5 mm, with a cylindrical gap width of 20 mm and length of 28 mm. The internal cylinder was provided with an electrical heater, while the temperature of the external cylinder was controlled by a jet ultrathermostabilization unit. Alignment screws were used to orient the mirrors at a slight angle to each other to form the working volume of a two-mirror autocollimating interferometer, allowing study of thermal and concentration fields [7, 8]. The interferogram of the model filled with a homogeneous concentration isothermal liquid was a system of straight vertical bands of equal widths. The appearance of temperature or concentration inhomogeneity led to a shift in the bands, proportional to the magnitude of the inhomogeneity. The temperature head between the cylinders was measured by copper–constantin thermocouples, connected to a type ShCh-300 digital voltmeter. The temperature of the surrounding medium was measured by a mercury thermometer with 0.1 K scale divisions.

The working liquids used were aqueous solutions of sodium sulfate, which are distinguished by a high value of the Soret coefficient and low diffusion coefficient [9, 10]. Before the start of each experiment the model was filled with fresh solution and hermetically sealed. The temperature of the external cylinder was set equal to that of the surrounding medium T_0 ($T_0 = 295 \pm 1$ K). After switch-on of the heater, thermal convection developed in the cavity and in 2-3 h steady-state temperature and concentration distribution was established. Entry into the steady-state regime was determined from the interference pattern.

The shift in the interference bands observed during the experiments is controlled by the change in the index of refraction of the liquid, upon which temperature and concentration act simultaneously, so that obtaining information on the concentration field is difficult. This

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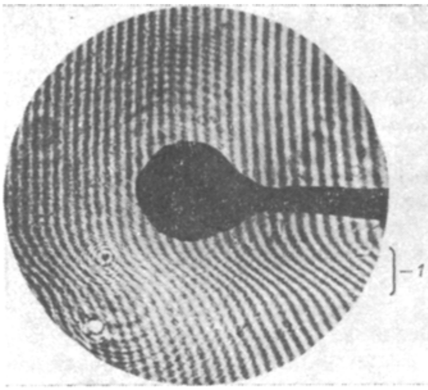

Fig. 1

difficulty was overcome by recording the concentration field under isothermal conditions, for which purpose the internal cylinder heater was switched off, thermal convection ceased, and the slow process of equalization of concentration perturbations began. Since the relaxation time of temperature perturbations is two to three orders of magnitude shorter than the relaxation time for concentration perturbations, the equalization process occurred under essentially isothermal conditions. The thermal perturbation relaxation time in the experiments did not exceed 25 min. The concentration field which existed at the moment of heater switch-off was reconstructed by extrapolating results obtained at various moments in time under isothermal conditions.

The method described cannot be used to study rapidly damping fine scale concentration perturbations, although integral characteristics can be measured quite reliably. Uncertainty in concentration measurement is related mainly to convective motion during the time of the transient process and proved insignificant for the cavity form chosen. The experiments performed revealed that marked concentration gradients develop only in the stagnation zone, where convective motion is suppressed by stable liquid stratification, with respect to both temperature and concentration.

Figure 1 shows a typical inteferogram of the concentration field in the gap filled with an aqueous sodium sulfate solution. Separation occurred under the action of thermodiffusion with a temperature head between the cylinders of $\Delta T = 15$ K. The Grashof number, defined in terms of the annular gap width, was equal to $6.9 \cdot 10^5$ with mean (over volume) concentration $C_0 = 0.11$. Here and below by concentration we understand the mass fraction of dissolved material. The photograph was made 45 min after heater switch-off. The absence of inclination in the bands above the cylinder indicates complete mixing of the solution by the convective motion. The concentration changes here are so small ($\Delta C \leq 8 \cdot 10^{-6}$) that they cannot be observed. Below the cylinder the band orientation changes; a transition layer appears – the zone with the highest concentration gradient in the cavity. In Fig. 1 this layer is denoted by the digit 1. The concentration distribution along the vertical diameter is shown in Fig. 2 ($C_0 = 0.157$, $t = 30$ min). The concentration value in the upper part of the cavity is taken as the origin of the coordinates. Curves 1-3 correspond to $\Delta T = 15.4$, 8.3, 2.4 K. Motion is practically absent in the lower part of the cavity and the concentration distribution over height is close to linear.

For the future we will characterize the concentration fields by the maximum concentration change within the cavity ΔC_1, the concentration change in the transition layer ΔC_2 (Fig. 2), and the height of the concentration zone h – the vertical distance from the low point of the cavity to the middle of the transition layer. All these parameters change slowly with time, which permits use of extrapolation, and estimation of the error connected with the transition to measurements in the isothermal regime. Thus for h this error does not exceed 10%.

Figure 3 shows ΔC_2 as a function of the temperature difference ΔT between the cylinders for $C_0 = 0.157$ and 0.066 (lines 1 and 2). A similar dependence is characteristic of the maximum ΔC_1 (analysis of the results shows that $\Delta C_1 \approx 3\Delta C_2$). As is evident from Fig. 3, for both solutions concentration separation is found only at certain temperature difference values ($\Delta T = \Delta T_*$). For $\Delta T < \Delta T_*$ convective motion encompasses the entire region and marked concentration changes are absent. At $\Delta T > \Delta T_*$ a stagnation zone develops in the lower portion

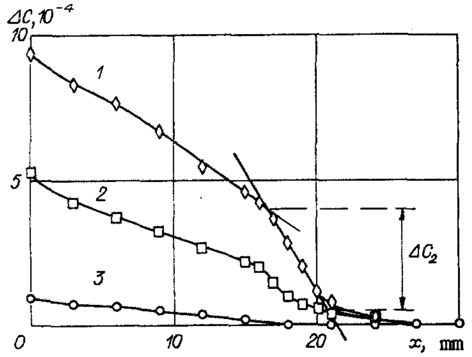

Fig. 2

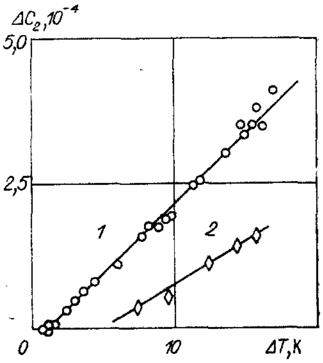

Fig. 3

of the cavity with a significant concentration change. As has already been noted, a stagnation zone is also formed in the gap between two cylinders for circulation of a one-component liquid [6], if $Gr > 10^3$. With such a zone stable temperature stratification of the liquid occurs, and a temperature boundary layer is formed at its upper boundary. The height of the stagnation zone is usually only 2-4 mm less than the gap width b (in the present study b = 20 mm). Thermodiffusion separation leads to additional suppression of convection and accelerates formation of the stagnation zone. Extremely weak motion with velocities $u \le 10^{-3}$ mm/sec occurs within this zone due to viscous friction. The motion is transferred from the developed convection zone and can be observed only by the distortions which it introduces into the concentration field formed by thermodiffusion.

The height of the concentration zone h determines the volume of liquid enriched by dissolved matter, and generally speaking, may be significantly less than b. This fact is a consequence of the large difference in the numerical values of the diffusion coefficients and thermal diffusivities of the liquid solutions. The creeping motion in the stagnation zone has a greater effect on the concentration field than on the temperature field. With increase in the thermodiffusion parameter $\gamma = S(1 - C_0)C_0\Delta T$ (where S is the Soret coefficient) liquid stratification intensifies in the stagnation zone and convective motion therein is suppressed. This leads to an increase in h, such that for $C_0 \ge 0.14$ and $\Delta T \ge 4$ K the former practically coincides with the height of the stagnation zone. We note that the parameter γ has the sense of a concentration change which develops due to thermodiffusion under convection-free conditions. As an example, Fig. 4 shows the dependence of h on the temperature difference between the cylinders. Curve 1 corresponds to $C_0 = 0.157$, curve 2, to 0.066. The dependence is nonlinear due to the crisis character of concentration zone formation. With increase in ΔT the intensity of the thermodiffusion effect increases while there is a simultaneous localization of the convective motion in the upper portion of the cavity. The dependence $h = f(C_0)$ is monotonic, and close to linear for low concentrations (Fig. 5, ΔT = 15 K).

The maximum concentration separation obtained in the experiments at ΔT = 15.7 K and C_0 = 0.157 comprised $9.3 \cdot 10^{-4}$ for the same concentration and temperature difference, but in a convection-free position (for example, in a planar horizontal layer with heating from above) significantly higher separation can be achieved ($\gamma = 0.018$). It is evident by comparison that weak liquid motion in the stagnation zone leads to a decrease in concentration separation by approximately an order of magnitude.

The intensity of the thermodiffusion effect increases with concentration ($C_0 \le 0.5$), so that the dependencies of ΔC_1 and ΔC_2 on C_0 were studied for a constant temperature difference (ΔT = 15 K) between the cylinders (Fig. 6). The dependence of ΔC_1 on C_0 is close to linear (curve 1). The dependence of the concentration change in the transition layer (curve 2) is of a more complex form, although on the whole they both increase monotonically with increase in γ.

Analysis of the results obtained permits the conclusion that thermodiffusion separation of liquid solutions under developed convection conditions ($Gr \ge 10^4$) can be neglected, except for those situations where it is possible for stagnation zones with very slow motion to form within the cavity. The solution concentration within such zones can differ greatly from the mean concentration over the volume. The volume of the concentration zone and the separation

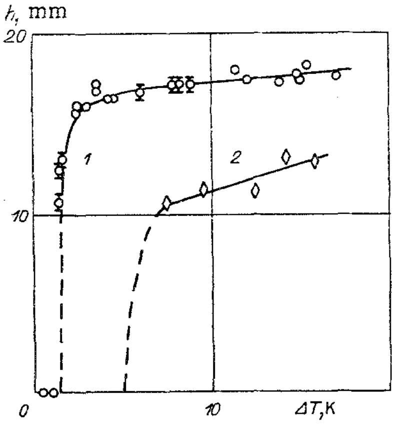

Fig. 4

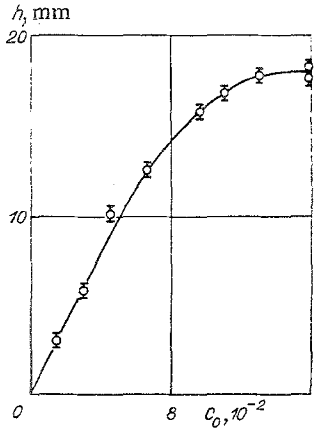

Fig. 5

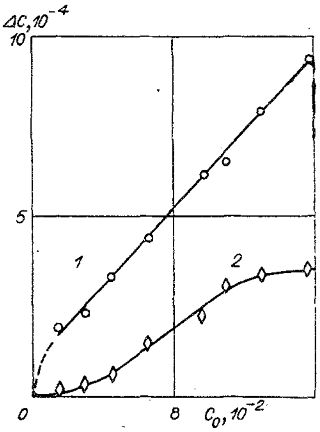

Fig. 6

achieved depend on the intensity of the convective motion and that of the thermodiffusion effect. Although separation is always less than under convection-free conditions.

In conclusion, the authors thank G. Z. Gershuni for his evaluation of the results and most useful observations.

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HYDRODYNAMIC STABILITY OF A CYLINDRICAL REACTION FRONT ASSOCIATED WITH A STRONG INCREASE OF VISCOSITY

G. V. Zhizhin and A. S. Segal'

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Hydrodynamic stability of a plane chemical reaction front in a gas was first considered in [1] neglecting transport effects. The effect of transport processes on the stability of a plane front in viscous gases and in condensed phases was studied in [2-5]. The stability of a curved front was considered in [6] for the example of the propagation of a spherical flame in a gas at rest. Transport effects were assumed to be small in this case and were taken into account phenomenologically in the Markstein approximation [7].

In the present paper we consider, in the linear approximation, the hydrodynamic stability of a stationary cylindrical reaction front in a radial, axisymmetric flow of a condensed medium. The flow is generated with the help of two coaxial, permeable, cylindrical surfaces. It is assumed that the viscosity of the medium is significantly increased by the reaction process (this is typical of polymerization reactions, for example) and hence inertial effects are small and are not taken into account [8, 9]. We study the dependence of the perturbation increment of the stationary states of the front on the parameters of the problem: the ratio of the viscosities of the medium on the front, the ratios of the radii of the boundary surfaces to the radius of the front, and the resistances of the order approximation in the (small) ratio of the viscosities this dependence is obtained analytically. It is shown that the front is absolutely stable in nearly the entire physical region of the parameter space. The front becomes unstable only when it approaches the outer boundary surface and the surface has a small resistance.

We note that it was found in a number of papers (see [8-12], for example) that for channel flow of a reacting medium, whose viscosity increases in the process of the reaction, the reaction front is sharply curved and drawn out near the channel axis (the "rupture" phenomenon). Our study of the hydrodynamic stability of a cylindrical front shows that small distortions of the front are damped for a wide range of the parameters and hence a "rupture" in the radial direction does not occur.

1. We assume the thickness of the front to be small compared to the distance between the surfaces bounding the flow and therefore treat it as a surface of discontinuity propagating with a constant velocity U with respect to the reactions medium (the local Michelson law [13]). The density of the medium is taken to be constant.

With these assumptions, the motion of the medium in front of and behind the front is described by the equation of continuity and the Stokes equation

$$\nabla p_{1,2} = \mu_{1,2}\nabla^2\mathbf{V}_{1,2}; \tag{1.1}$$

$$\nabla\cdot\mathbf{V}_{1,2} = 0, \tag{1.2}$$

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