Convective Instability and Appearance of Structured Flows for Diffusion in Multicomponent Gas Mixtures

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Abstract: The main objective of this article is to investigate the behavior of gaseous systems with two and more independent gradients or thermodynamic forces exhibiting complicated behavior, when the convective flows occur. The existence of structural formations in these systems is shown by the schlieren method and the fast-response transducers. The linear analysis of stability can explain reasons of the appearance of convective instability in multicomponent gas mixtures.

Key words: diffusion; instability of mechanical equilibrium; theory of stability; partial Rayleigh numbers

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1 Introduction

Instability of mechanical equilibrium in isothermal binary gas mixture may occur only in case when a heavier gas in density is situated on the top (Fig. 1(a)). Then convection occurs under the influence of buoyancy force, i. e. a heavier gas moves downwards, and a lighter one rises. Convection continues until a lighter gas occupies the upper position. If lighter gas were on top then convection would not occur[1].

Adding the third component to the mixture (Fig. 1(b)), as well as placement of mixture in non-homogeneous temperature field, under certain conditions may lead to instability of mechanical equilibrium and occurrence of convection[2]. The reason for this effect is that as opposed to binary isothermal system convection streams in isothermal ternary or non-isothermal binary system are caused not by one concentration gradient but two gradients and the second gradient would be either concentration or thermal[1].

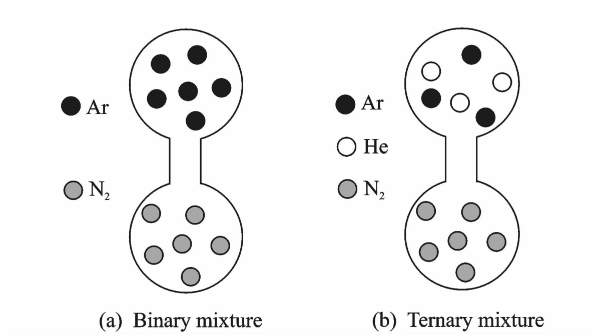

Fig. 1 Scheme of diffusion cell

2 Experimental Data

The experiments on instability were carried out using a two-flask method[3-4] (Figs. 1(a, b)). Two flasks of the same volume were connected through a vertical channel. The pressure in the flasks was maintained the same. Temperature conditions were dependent on the tasks of the experiment. Methodology of experiments was similar to classic one. Capillary that was connecting to two flasks was opened. Some time later, it was closed and the mixture composition in both flasks was registered.

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Received concentrations during the experiment c_{exp} were normalized to the calculated one c_{theor} under the assumption of diffusion.

2.1 Isothermal ternary mixture

There was a binary mixture of light and heavy component in the upper flask. The gas with the average density was placed in the lower flask (Fig. 2). Concentrations of light and heavy components were selected in such a way that mixture in the upper flask always was of a lower density than gas in the lower flask ($\nabla\rho<0$).

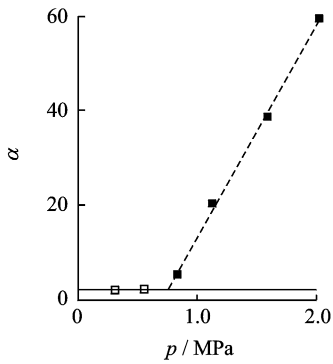

Fig. 2 Dependences of parameter α for argon on pressure in ternary mixture $0.35He + 0.65Ar - N_2$, $T = 293.0$ K

Typical dependences of the parameter $\alpha = c_{\mathrm{exp}}/c_{\mathrm{theor}}$ on pressure for the system $0.35He + 0.65Ar - N_2$ are given in Fig. 2 and for the system $0.4Ar + 0.6N_2 - 0.6Ar + 0.4N_2$ are presented on Fig. 3.

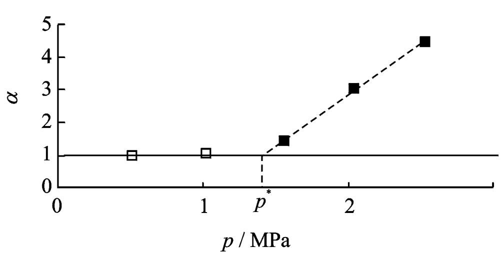

Fig. 3 Dependences of parameter α for argon on pressure in binary mixture $0.4Ar + 0.6N_2 - 0.6Ar + 0.4N_2$ (temperature of the upper flask is 288.0 K, lower flask is 343.0 K)

Hereinafter we are going to indicate points that correspond to diffusion with the open marks "□", and convections with the dark marks "■". Numbers going before chemical elements define molar fraction of the component. It follows from Fig. 2 that, at certain pressure parameter α is higher than unity, i.e. mechanical equilibrium of the mixture become instable. Concentration gravitational convection occurs. The experiments performed previously[2-3] showed that transfer from diffusion to convection is defined by the following parameters: pressure, temperature, mixture viscosity, its original composition, difference in the diffusion coefficients of components, geometry of the channel, and its orientation in relation to the vertical, rotation.

For visual registration of the transition boundaries between the diffusion and the convection schlieren method was applied. Experiments registered presence of convective structured flows of different intensity (Fig. 4).

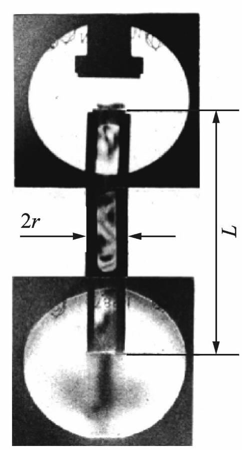

Fig. 4 Convection structures in isothermal ternary gas mixtures

Change of convection regimes can be registered with the fast-response transducers that define the local thermal conductivity of gas also. Because the thermal conductivity of gas mixture depends on the concentration of components, this gives an opportunity to track their changes in time and thus identify the significant periods of observed structures (Fig. 5).

Study of convection in isothermal ternary gas systems, in the range of parameters that considerably exceed critical one $\alpha \gg 1$, revealed effects connected with the nonlinear dependence of intensity of instable process on pressure (Fig. 6), radi-

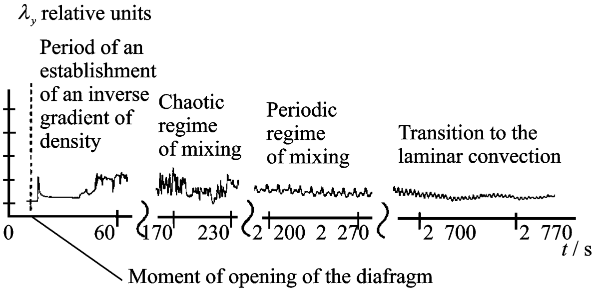

Fig. 5 Convection structures in isothermal ternary gas mixtures, and change of mixture thermal conductivity that illustrates change of convection structures

us of diffuser channel, and angle of inclination of channel in relation to verticality (Fig. 7), and repeated change of regimes between the diffusion and convection [2-3].

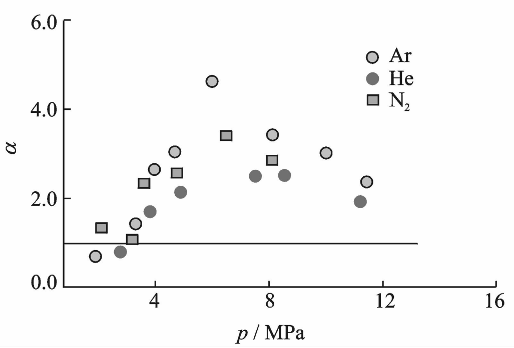

Fig. 6 Parameter α at various pressures for different components in the mixture 0.47He + 0.53Ar—N_2, T=293.0 K

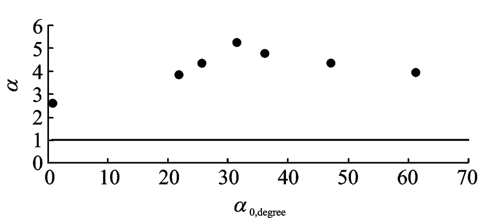

Fig. 7 Parameter α at various angles of inclination of channel α in relation to verticality for helium in the mixture 0.47He + 0.53Ar — N_2, T = 293.0 K, p=2.54 MPa

2.2 Binary mixture

Density of gas (or mixture of gases) in the upper flask is higher than the density of component in the lower flask (Fig. 3). Temperature of the lower flask is higher than temperature of the upper one. On Fig. 3 there are typical dependencies of the parameter $\alpha = c_{\text{exp}}/c_{\text{theor}}$ on pressure for the system 0.4Ar + 0.6N_2 — 0.6Ar + 0.4N_2. From figure one can see that at some pressure $p_* \approx 1.4$ MPa parameter α is higher than unity. Mechanical equilibrium of the mixture becomes instable and convection occurs. The previous research showed that such transfer is defined by the following critical parameters: pressure, temperature difference, geometrical characteristics of the channel and its orientation in relation to verticality, rotation of diffusion cell[4].

3 Theoretical Analysis of Stability of Mechanical Balance

It is possible to predict transition boundaries between the diffusion and the convection with the help of convection stability analysis[1,5]. System of hydrodynamic equations for general case of ternary mixture in non-homogeneous temperature field is defined in a following way [1,6,7]

$$\rho\left[\frac{\partial \boldsymbol{u}}{\partial t} + (\boldsymbol{u}\nabla)\boldsymbol{u}\right] =$$

$$-\nabla p + \eta \nabla^2 \boldsymbol{u} + \left(\frac{\eta}{3} + \xi\right) \nabla \operatorname{div} \boldsymbol{u} + \rho \boldsymbol{g}$$

$$\frac{\partial \rho}{\partial t} + \operatorname{div}(\rho \boldsymbol{u}) = 0$$

$$\rho\left(\frac{\partial c_i}{\partial t} + \boldsymbol{u}\nabla c_i\right) = \operatorname{div} \boldsymbol{j}_i \qquad (1)$$

$$\rho T\left(\frac{\partial S}{\partial t} + \boldsymbol{u}\nabla S\right) = -\operatorname{div}\boldsymbol{q} + \mu \sum_i \operatorname{div} \boldsymbol{j}_i$$

$$\boldsymbol{j}_1 = -\rho(D_{11}^* \nabla c_1 + D_{12}^* \nabla c_2)$$

$$\boldsymbol{j}_2 = -\rho(D_{21}^* \nabla c_1 + D_{22}^* \nabla c_2)$$

$$\sum_{i=1}^{3} \boldsymbol{j}_i = 0,\ \sum_{i=1}^{3} c_i = 1$$

where ρ is the density, $\boldsymbol{u}$ the velocity, t the time, p the pressure, η and ζ are the coefficients of shear and bulk viscosity, $\boldsymbol{g}$ is the free fall acceleration, c_i the concentration of i-th component, S the entropy, T the temperature, $\boldsymbol{q}$ the heat flow, $\boldsymbol{j}$ the diffusion flow, μ the effective chemical potential, and $D_{ij}{}^*$ the matrix coefficient of multicomponent diffusion.

Eqs. (1) are supplemented by the environmental state equation $\rho = \rho(c_i, p, T)$.

We simplify Eqs. (1) in a following way:

(1) Apply the method of small parameter;

(2) Make system of the equation linear;

(3) Choose the following scale units of measurement: d is the distance, d^2/ν the time, D_{22}^*/d the velocity, $A_i d$ the concentration, $\rho_0 \nu D_{22}^*/d^2$ the pressure, and Bd the temperature;

(4) Taking into account that the temperature difference is small then the impact of cross effects is negligible.

The equation system (1) can be solved both for the plane layer and for the cylindrical channel.

For binary gas systems in non-homogeneous temperature field the equations of perturbation have the following form

$$\frac{\partial u}{\partial t} = \Delta u + Ra_c c + Ra_T T$$
$$-\lambda \Pr T = \Delta T + u$$
$$-\lambda \Pr_c c = \Delta c + u \qquad (2)$$

where $\Pr_c = \nu/D_{12}$ is the concentration Prandtl number, $\Pr = \nu/\chi$ the heat Prandtl number, $\nu = \eta/\rho_0$ the kinematic viscosity of the mixture, χ the thermal diffusivity, $Ra_c = g\beta A d^4/\nu D_{12}$ the concentration Rayleigh number, $Ra_T = g\beta_T B d^4/\nu\chi$ the heat Rayleigh number, and

$$\beta = -\frac{1}{\rho_0}\left(\frac{\partial \rho}{\partial c}\right)_{p,T}, \quad \beta_T = -\frac{1}{\rho_0}\left(\frac{\partial \rho}{\partial T}\right)_{p,c}$$

The solution of Eqs. (2) for the cylindrical channel of unlimited height by analogy with Ref. [2] results in the definition of boundary lines of monotonous (MM) and oscillatory (KK) instabilities

$$Ra_c + Ra_T = 67.95$$
$$\Pr_c^2(\Pr + 1)Ra_T + \Pr^2(\Pr_c + 1)Ra_c =$$
$$67.95(\Pr + \Pr_c)(\Pr + 1)(\Pr_c + 1) \qquad (3)$$

For ternary mixtures the equations of perturbations in isothermal conditions can be written as[12]

$$\Pr_{22}\frac{\partial c_1}{\partial t} - u = \tau_{11}\Delta c_1 + \frac{A_2}{A_1}\tau_{12}\Delta c_2$$
$$\Pr_{22}\frac{\partial c_2}{\partial t} - u = \frac{A_1}{A_2}\tau_{21}\Delta c_1 + \Delta c_2$$
$$\frac{\partial u}{\partial t} = \Delta u + Ra_1\tau_{11}c_1 + Ra_2 c_2 \qquad (4)$$

where $\Pr_{ii} = \nu/D_{ij}^*$ is the Prandtl number, $Ra_i = g\beta_i A d^4/\nu D_{ij}^*$ the Rayleigh number, $\tau_{ij} = D_{ij}^*/D_{22}^*$, $A_i\boldsymbol{\gamma} = -\nabla c_{i0}$, $\boldsymbol{\gamma}$ the unit vector.

The solution of Eqs. (4) for the plane vertical channel at the boundary conditions, presupposing the vanishing of the velocity perturbations and the perturbations of the component concentrations on vertical planes bounding the layer of gas mixture allowed obtaining the boundary line of monotonous perturbations in the following form

$$\tau_{11}\left(1 - \frac{A_2}{A_1}\tau_{12}\right)Ra_1 + \left(\tau_{11} - \frac{A_1}{A_2}\tau_{21}\right)Ra_2 =$$
$$\left[(n+1)\frac{\pi}{2}\right]^4(\tau_{11} - \tau_{12}\tau_{21}) \qquad (5)$$

Boundary lines (Eqs. (3, 5)) indicated on Figs. 8, 9 determine the diffusion and convection mixing regions for isothermal and non-isothermal cases in the plane vertical channel, as well as the experimental data shown on Figs. 2, 3 recovered through the Rayleigh numbers.

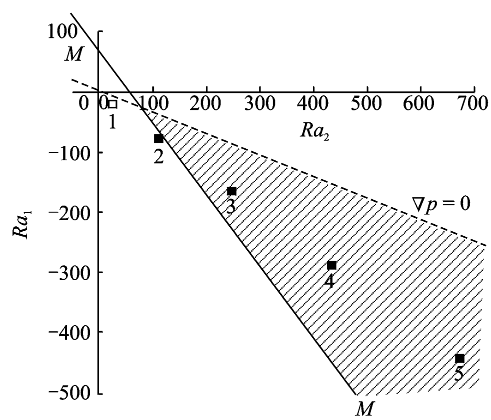

Fig. 8 Boundary line of monotonous instability MM, and line of equal density and experiment data for the system 0.35He + 0.65 Ar − N_2, T = 293.0 K (Points correspond to the pressure values: 1—0.584, 2—1.074, 3—1.565, 4—2.055, 5—2.546 MPa)

Analysis of Fig. 8 permits identifying the region with the negative density gradient, but lying above the instability line MM. On Fig. 8 this region is shaded. If experiment conditions will be adjusted in such a way that the system is in this region, then there should be convection observed, registered experimentally on Figs. 2, 3.

Fig. 9 shows in the coordinates (Ra_c, Ra_T) a mutual relationship of the line MM and the experimental data for non-isothermal mixture 0.4 Ar + 0.6 N_2 − 0.6 Ar + 0.4 N_2.

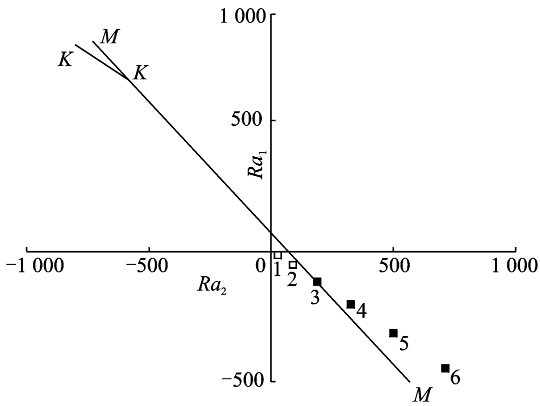

Fig. 9 Boundary line of monotonous instability of mechanical equilibrium *MM* and experimental data for the system 0.4 Ar+0.6N_2−0.6 Ar+0.4 N_2 (Temperature of the upper flask is 288.0 K, one of the lower flask is 343.0 K, and points correspond to the pressure values: 1—0.584, 2—1.074, 3—1.565, 4—2.055, 5—2.546, 6—3.036 MPa)

4 Conclusions

Until convection in the gas systems is conditioned by single gradient of concentration or temperature, the primary reason for its occurrence is the condition when density in the upper section is higher than density in the lower one. When second gradient appears the picture is changed. Now convection may occur at any direction of the density gradient. Theory of stability adequately describes experimental data in both cases when the second gradient is thermal and when it is concentration.

Acknowledgements

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