Journal of Engineering Physics and Thermophysics, Vol. 75, No. 4, 2002

EXPERIMENTAL DETERMINATION OF THE BOUNDARY OF MONOTONIC DISTURBANCES UPON INSTABILITY OF THE MECHANICAL EQUILIBRIUM IN THREE-COMPONENT GAS MIXTURES

Yu. I. Zhavrin,[a] **V. N. Kosov,**[b]
D. U. Kul'zhanov,[c] **and K. K. Karataeva**[c]

UDC 533.15:536.25

Instability of the mechanical equilibrium of ternary gas mixtures, in which the density from above is larger than from below, is investigated by the two-flask method under isothermal conditions at different pressures. A comparison of the experimental data with the results calculated within the framework of the linear analysis of stability has shown fair agreement in determination of the boundaries of instability.

An experimental study of the diffusion in ternary gas mixtures in a vertical channel with isothermal diffusion has revealed that under certain conditions (pressure, temperature, geometric sizes of a capillary) instability of the mechanical equilibrium of the mixture develops followed by free concentration convection in the gravity field [1, 2]. The reasons for the occurrence of the gravitational convection can be explained within the framework of the linear theory of stability [3], the application of which for the case of an isothermal mixture has revealed different types of diffusional mixing [4, 5]. However, for some regimes of mixing experimental data are virtually absent. In this case, theoretical predictions of the position of the regions of stability (instability) need experimental verification. In the present work, the authors report experimental data on determination of the boundaries of disturbance of the mechanical equilibrium for diffusion of a heavier, with respect to density, binary mixture placed in the upper section of the channel toward the third smaller-density component.

Procedure and Experimental Results. A change of the diffusion regime for free gravitational convection was observed in a two-flask apparatus (Fig. 1) with flask volumes of $\approx 55.6\ \text{cm}^3$, with the capillary of diameter $d = 0.4$ cm and length of $L = 6.4$ cm at a temperature of $T = 298$ K. Experiments were carried out at different pressures. A study was made of the diffusion of a binary mixture of helium and argon, placed into flask I, in nitrogen, which was in flask II.

The composition of the initial components in the binary mixture was chosen such that at any pressure in the experiment the density of the mixture in the upper flask was larger than the nitrogen density: $\nabla\rho_{\text{mix}} > 0$.

The initial composition of the binary mixture was analyzed by an ITR-1 interferometer with an error of 0.1% and by a chromatograph with an error of 0.3%. In all cases, the experiment lasted 30 min. On cessation of each experiment the composition of the gas mixtures was analyzed in both flasks.

The binary transition diffusion-concentration convection can be traced if the parameter $\alpha_i = Q_i^{\text{exp}}/Q_i^{\text{theor}}$ (i = 1, 2, 3) is represented graphically as a function of pressure. Here Q_i^{theor} is the diffusion flux calculated by the Stefan–Maxwell equations in the assumption of diffusion [6], Q_i^{exp} is the experimental flux found from experimental values of the concentration and the time of mixing of the component i. The typical dependences of thus-obtained relative values of α_i on pressure for argon in the $0.3\text{He} + 0.7\text{Ar} - \text{N}_2$ and $0.1\text{He} + 0.9\text{Ar} - \text{N}_2$ systems (the gas concentrations are given in molar fractions) are depicted in Fig. 2. Similar dependences are also observed for the other components of the mixture.

A change of the diffusion-concentration convection regimes occurs when α_i exceeds unity. The critical value of pressure determining the transition from the diffusion region to the convection one is substantially smaller for the systems with a positive density gradient than for the ternary mixtures where under similar conditions the instability of the mechanical equilibrium is manifested for the case of a negative density gradient [1, 2]. An increase in the concen-

[a]Kazakh Al'-Farabi State National University, Almaty; [b]Abai Almaty State University; [c]Atyrausk Institute of Petroleum and Gas. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 75, No. 4, pp. 80–83, July–August, 2002. Original article submitted July 11, 2001; revision submitted December 17, 2001.

 1062-0125/02/7504-0868$27.00

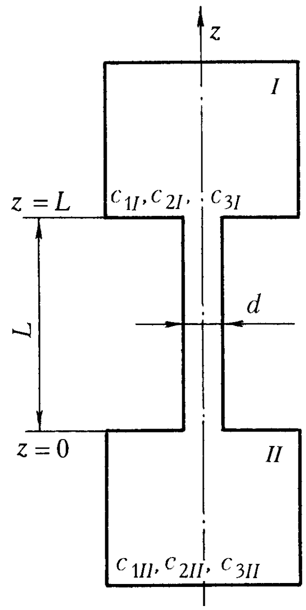

Fig. 1. Two-flask apparatus. Geometry of the problem.

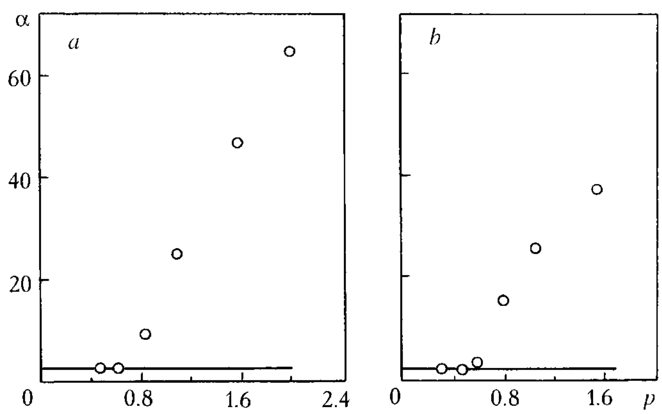

Fig. 2. Dependence of α of argon on pressure in mixing of the binary helium-argon mixture with nitrogen (T = 298 K); a) 0.3He + 0.7Ar; b) 0.1He + 0.9Ar. p, MPa.

tration of the heavy component in the mixture leads in the region of pressures from 1.2 to 1.5 MPa to a decrease in the critical pressure determining the unstable regime of mixing.

Comparison of the Experimental Results and Theory. Following [3–5], we will repeat the reasoning behind the occurrence of the instability of the mechanical equilibrium of the three-component gas mixture under the conditions of isothermal diffusion in a plane vertical channel. Macroscopic motion of the isothermal gas mixture with account for the condition of independent diffusion at which $\sum_{i=1}^{3} \mathbf{j}_i = 0$ and $\sum_{i=1}^{3} c_i = 1$ is described by the following system of equations:

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u}\nabla)\,\mathbf{u} \right] = -\nabla p + \eta \nabla^2 \mathbf{u} + \left(\frac{\eta}{3} + \xi \right) \nabla\, \mathrm{div}\, \mathbf{u} + \rho\, \mathbf{g}\,, \quad \frac{\partial \rho}{\partial t} + \mathrm{div}\,(\rho\, \mathbf{u}) = 0\,,$$

$$\rho \left(\frac{\partial c_i}{\partial t} + \mathbf{u}\, \nabla c_i \right) = -\,\mathrm{div}\, \mathbf{j}_i\,, \quad \mathbf{j}_1 = -\,\rho\, (D_{11}^{*} \nabla c_1 + D_{12}^{*} \nabla c_2)\,, \quad \mathbf{j}_2 = -\,\rho\, (D_{21}^{*} \nabla c_1 + D_{22}^{*} \nabla c_2)\,. \tag{1}$$

The "practical" diffusion coefficients D_{ij}^* are related to the diffusion coefficients of the binary gas mixtures D_{ij} by the expressions

$$D_{11}^* = \frac{D_{13}\,[c_1 D_{32} + (c_2 + c_3)\,D_{12}]}{c_1 D_{23} + c_2 D_{13} + c_3 D_{12}}\,, \quad D_{12}^* = -\,\frac{c_1 D_{23}\,(D_{12} - D_{13})}{c_1 D_{23} + c_2 D_{13} + c_3 D_{12}}\,,$$

$$D_{22}^* = \frac{D_{23}\,[c_2 D_{13} + (c_1 + c_3)\,D_{12}]}{c_1 D_{23} + c_2 D_{13} + c_3 D_{12}}\,, \quad D_{21}^* = -\,\frac{c_2 D_{13}\,(D_{12} - D_{23})}{c_1 D_{23} + c_2 D_{13} + c_3 D_{12}}\,.$$

System (1) is supplemented with the equation of state of a medium

$$\rho = \rho\,(c_1, c_2, p)\,, \quad T = \mathrm{const}\,,$$

that makes it possible to relate the thermodynamic parameters in (1).

Account for the smallness of unsteady disturbances of the mechanical equilibrium, the assumption about the linear distribution of the concentration of components in the channel, the neglect of the squared disturbance terms, the choice of scale units of measurement (distance being the linear dimension of cavity d, time d^2/ν, velocity D_{22}^*/d, concentration of the ith component $A_i d$, pressure $\rho_0 \nu D_{22}^*/d^2$) allow one to pass from (1) to the disturbance equations [5]

$$\mathrm{Pr}_{22}\,\frac{\partial c_1}{\partial t} - u = \tau_{11}\,\frac{\partial^2 c_1}{\partial x^2} + \frac{A_2}{A_1}\,\tau_{12}\,\frac{\partial^2 c_2}{\partial x^2}\,, \quad \mathrm{Pr}_{22}\,\frac{\partial c_2}{\partial t} - u = \frac{A_1}{A_2}\,\tau_{21}\,\frac{\partial^2 c_1}{\partial x^2} + \frac{\partial^2 c_2}{\partial x^2}\,, \quad \frac{\partial u}{\partial t} = \frac{\partial^2 u}{\partial x^2} + \mathrm{Ra}_1\,\tau_{11} c_1 + \mathrm{Ra}_2\,c_2\,, \tag{2}$$

where $\mathrm{Pr}_{ii} = \nu/D_{ii}^*$; $\nu = \eta/\rho$; $\mathrm{Ra}_i = g\beta_i A_i d^4/\nu D_{ii}^*$; $\beta_i = -\dfrac{1}{\rho_0}\left(\dfrac{\partial\rho}{\partial c_i}\right)_{p,T,c_j \neq c_i}$; $\tau_{ij} = D_{ij}^*/D_{22}^*$; $u = u_z$; $A_i\boldsymbol{\gamma} = -\nabla c_{i0}$; $\boldsymbol{\gamma}$ – is the unit vector directed vertically upwards, the index 0 pertains to average values.

A solution of (2) has the form

$$\left\{c_1, c_2, u\right\} = \left\{c_1^0, c_2^0, u^0\right\} \sin\left[(s + 1)\,\frac{\pi}{2}\,x\right] \exp\,[-\,\lambda t]\,, \tag{3}$$

where s = 1, 3, 5, ... are the odd modes of disturbances. The boundary conditions assume disappearance of the velocity and disturbances of the concentrations of the components c_i on the vertical planes restricting a layer of the gas mixture:

$$u = c_1 = c_2 = 0\,, \quad x = \pm\,1\,. \tag{4}$$

Substitution of (3) into (2), account for (4), and vanishing of the real part of the disturbance decrement λ, yield the equation of monotonic disturbances

$$\tau_{11}\left(1 - \frac{A_2}{A_1}\,\tau_{12}\right)\mathrm{Ra}_1 + \left(\tau_{11} - \frac{A_1}{A_2}\,\tau_{21}\right)\mathrm{Ra}_2 = \left[(s + 1)\,\frac{\pi}{2}\right]^4 [\tau_{11} - \tau_{12}\tau_{21}]\,. \tag{5}$$

As noted in [4, 5], if for the binary mixtures the region of stable diffusion is determined by fulfillment of the condition $\mathrm{Ra} \le \mathrm{Ra}_{\mathrm{cr}}$, for the ternary mixtures the corresponding region should be sought on the two-dimensional plane of partial Rayleigh numbers (Fig. 3).

Below line *MM* obtained within the framework of (15) the mechanical equilibrium is stable while above this line, it is unstable. Since the mixture density is related to the concentrations of the components c_i by the expressions

$$\rho = n\,(c_1 m_1 + c_2 m_2 + c_3 m_3) = n\,[c_1\,(m_1 - m_3) + c_2\,(m_2 - m_3) + m_3] = n\,(c_1 \Delta m_1 + c_2 \Delta m_2 + m_3)\,,$$

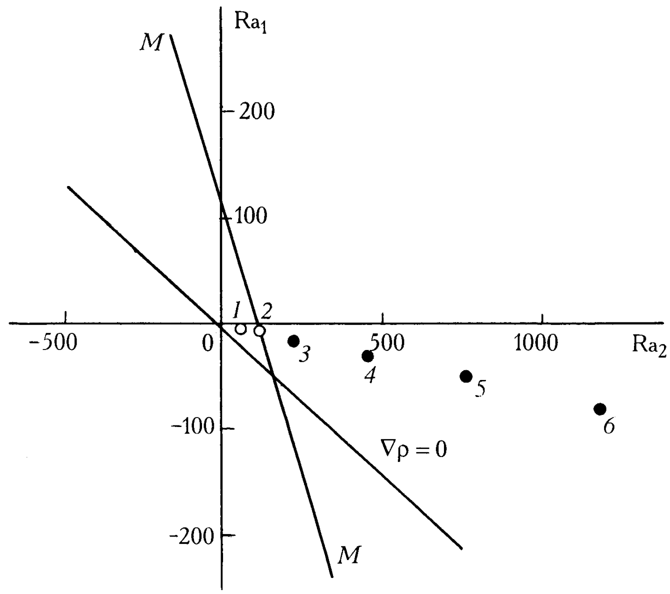

Fig. 3. Regions of diffusion and monotonically increasing disturbances. The system is $1.0He + 0.9Ar - N_2$, T = 298 K. The experimental points correspond to the following pressures: 1) 0.33; 2) 0.46 MPa (stable diffusion); 3) 0.58; 4) 0.83; 5) 1.07; 6) 1.56 MPa (unstable diffusion).

the condition of vanishing of the density gradient of the mixture is of the form

$$\nabla\rho = n\,[(dc_1/dz)\,\Delta m_1 + (dc_2/dz)\,\Delta m_2] = 0\,. \tag{6}$$

In terms of the Rayleigh numbers, expression (6) is written as

$$\tau_{11}\,\mathrm{Ra}_1 = -\,\mathrm{Ra}_2\,. \tag{7}$$

In Fig. 3, condition (7) determines the line passing through the origin of the coordinates. Above this line, the density gradient is positive. Analyzing Fig. 3, we can easily find the regions of diffusion and increasing monotonic disturbances above instability line *MM*. If the conditions of the experiment are chosen in such a way that, for instance, the system is in the diffusion region, then changing one of the parameters determining the Rayleigh number, e.g., pressure, it is possible to pass to the region of gravitational concentration convection. This procedure is depicted in Fig. 3, where the experimental points corresponding to stable diffusion (open circles) and convection (filled circles) are obtained by substituting the experimental concentration values of the $0.1He + 0.9Ar - N_2$ mixture into the expressions for partial Rayleigh numbers in the form [5]

$$\mathrm{Ra}_1 = \frac{gnd^4\Delta m_1\Delta c_1}{\rho\nu D_{11}^{*}L}\,,\quad \mathrm{Ra}_2 = \frac{gnd^4\Delta m_2\Delta c_2}{\rho\nu D_{22}^{*}L}\,,$$

$$n = \frac{p}{kT}\,,\quad \Delta c_1 = c_{1I} - c_{1II}\,,\quad \Delta c_2 = c_{2I} - c_{2II}\,,\quad \Delta m_1 = m_1 - m_3\,,\quad \Delta m_2 = m_2 - m_3\,,$$

where index 1 corresponds to helium, 2 to argon, and 3 to nitrogen. From Fig. 3 it is seen that the experiment confirms the theoretical assumptions about the existence of the region of stable and unstable mechanical equilibrium in the ternary gas mixtures when the density gradient is positive. A similar picture is also observed for the ternary $0.3He + 0.7Ar - N_2$ system.

Thus, it is shown that on isothermal diffusional mixing of a heavier-density binary mixture with a third (light) component there exist regions of diffusion and gravitational concentration convection. The experiments with the

$He + Ar - N_2$ mixture prove the presence of these two regions. The location of the stability boundaries and monotonic disturbances is described well by the linear theory.

The authors thank N. B. Ankusheva for her help in processing of the experimental results.

NOTATION

c_i, concentration of the ith component; D_{ij}^* and D_{ij}, "practical" diffusion coefficient and mutual diffusion coefficient of the gases i and j, respectively; **g**, gravitational acceleration; $\mathbf{j}_i$, density of the diffusion flux of the ith component; k, Boltzmann constant; L, length of the diffusion channel; m, molecule mass; n, numerical density of the mixture; Pr_{ii}, Prandtl number; p, pressure; Ra_i, Rayleigh number; d, characteristic scale, diameter of the diffusion channel; T, temperature; t, time; **u**, velocity; α, parameter determining the diffusion-concentration convection transition; η, coefficient of shear viscosity; λ, time decrement of disturbances; ν, kinematic viscosity of the mixture; ξ, coefficient of volume viscosity; ρ and ρ_0, density and mean density; τ_{ij}, parameter determining the ratio between the "practical" diffusion coefficients.

REFERENCES

1. Yu. M. Zhavrin, N. D. Kosov, S. M. Belov, and S. B. Tarasov, *Zh. Tekh. Fiz.*, **54**, No. 5, 943–947 (1984).
2. Yu. M. Zhavrin and N. D. Kosov, *Inzh.-Fiz. Zh.*, **55**, No. 1, 92–97 (1988).
3. G. Z. Gershuni and E. M. Zhukhovistkii, *Convective Stability of an Incompressible Fluid* [in Russian], Moscow (1972).
4. V. N. Kosov, V. D. Seleznev, and Yu. I. Zhavrin, *Teplofiz. Aéromekh.*, **7** , No. 1, 3–10 (2000).
5. V. N. Kosov, V. D. Seleznev, and Yu. I. Zhavrin, *Inzh.-Fiz. Zh.*, **73**, No. 2, 313–320 (2000).
6. Yu. I. Zhavrin, N. D. Kosov, and Z. I. Novosad, in: *Diffusion in Gases and Liquids* [in Russian], Alma-Ata (1974), pp. 24–29.