Computational Thermal Sciences, 11(1-2):29–39 (2019)

NUMERICAL MASS TRANSFER STUDIES IN CASE OF CONVECTIVE FLOWS OCCURRENCE IN ISOTHERMAL TERNARY GAS MIXTURES

Vladimir Kossov,[1,] Dauren Zhakebayev,[2] & Olga Fedorenko[3]*

[1]*Abai Kazakh National Pedagogical University, Almaty, Kazakhstan*

[2]*Al-Farabi Kazakh National University, Almaty, Kazakhstan*

[3]*Institute of Experimental and Theoretical Physics at Al-Farabi Kazakh National University, Almaty, Kazakhstan*

*Address all correspondence to: Vladimir Kossov, Abai Kazakh National Pedagogical University, Almaty, Kazakhstan; Tel.: +7 777 160 6156; Fax: +7 727 291 1022, E-mail: kosov_vlad_nik@list.ru

Original Manuscript Submitted: 10/22/2017; Final Draft Received: 3/21/2018

Isothermal diffusion mixing in multicomponent gas mixtures at various pressures is studied numerically in He + Ar – N_2 and CH_4 + Ar – N_2. It is shown that in systems where the diffusion coefficients differ significantly from each other, nonlinear distributions of the concentrations of the components arise with increasing pressure, which leads to a nonmonotonous density distribution of the gas mixture in the computational domain. The resulting nonlinearity is the cause of the formation of convective structured flows. The transition from the diffusion to the convective regime is characterized by a significant increase in the average mixing rate. The dynamics of the development of convective currents and their structure is investigated.

KEY WORDS: *diffusion, convection, mixtures, pressure, instability, structures*

1. INTRODUCTION

There are wide varieties of mixing regimes in multicomponent gas mixtures (Taylor and Krishna, 1993). In fact, practically, it is not taken into account that the process of molecular diffusion can lead to an instability of the mechanical equilibrium of the mixture with the subsequent occurrence of natural convection (Linden et al., 1994; Kaminskii and Obvintseva, 2007), which appreciably intensifies the total mass transfer. Moreover, the occurrence and development of concentration convection occurs not only in the framework of the traditional concepts of the Rayleigh thermal problems (Joseph, 1976; Nield and Bejan, 2006), but also for situations in which motions arise under stable stratification of the mixture (Kosov et al., 1997; Zhavrin et al., 2016).

In experiments (Zhavrin and Kosov, 1988) on the study of diffusion in ternary gas mixtures, it was found that due to the difference in the diffusion activity of the components in such systems convective instability appears. The intensity of total mass transfer in this case increases by tens and hundreds of times in comparison with diffusion mixing and depends on such characteristics as pressure (Zhavrin et al., 2000), initial mixture composition (Kosov et al., 2017), temperature (Seleznev et al., 2013), channel geometric characteristics, and angle of inclination (Ankusheva et al., 2010). Investigating the diffusion of a mixture of solution vapors under normal pressure into an inert gas (Dil'man et al., 2005), convective currents not typical for diffusion were recorded. In such cases the experiments (Lotkhov et al., 2003) have shown the possibility of occurrence of a synergistic effect associated with a significant increase of speed of the mixing components of the system. Summarizing the results of experimental studies on multicomponent mixing, it is possible to conclude that the diffusion regime changes to a convective one.

The apparent paradoxical origin of convection in diffusion can be explained within the framework of stability theory (Joseph, 1976; Gershuni and Zhukhovitskii, 1976). Extending the approach of the theory of stability to the

NOMENCLATURE

A_i	dimensionless concentration gradient (-)
D^*_{ij}	"practical" coefficients of three-component diffusion (m^2/s)
c_i	i-th component concentration (-)
d	diameter (m)
$\vec{j}_i$	diffusion flux vector (m/s)
n	number density (m^{-3})
m_i	molecular mass (kg)
p	pressure (Pa)
r	radius (m)
$\vec{u}$	weight-average velocity vector (m/s)
$\vec{v}$	number-average velocity vector (m/s)

Greek Symbols

α	dimensionless parameter defined by the ratio of experimental values of concentrations c_{exp} to the concentrations c_{theor} calculated by Stefan-Maxwell equations (-)
β_i	coefficient of thermal expansion (m/K)
$\vec{\gamma}$	unit vector (-)
η	shear viscosity (Pa·s)
ξ	bulk viscosity (Pa·s)

Subscripts and Superscripts

i, j	numbering of components in multicomponent system
exp	experiment
$theor$	theory

case of isothermal mixing of ternary gas mixtures, it was shown (Kosov et al., 2000) that the change in the "diffusion-convection" regimes is associated with the occurrence of increasing vibration perturbations in the system, followed by the appearance of structured flows. Further research (Kossov et al., 2017) on the study of the characteristic features of the change in the kinetic regimes during diffusion in vertical channels of various geometries showed that convective instability manifests itself in such systems, leading to the appearance of concentration gravitational convection leading to a significant increase in the mixing speed of the components of the system.

At the same time, it should be noted that the method used in Gershuni and Zhukhovitskii (1976) and Kossov et al. (2017), which studies the stability of dynamical systems, includes a linear analysis in itself. Possibilities of it are limited to the class of stationary problems; i.e., systems with a linear distribution of the density along the height of the diffusion channel at the initial instant of time are studied. Therefore, the use of numerical methods for solving hydrodynamic equations in the case of isothermal ternary gas mixtures, taking into account free and forced convection, gives the possibility for more detailed studying of the features of conditions for the onset of convective instability, depending on the initial conditions and the physicochemical parameters of the system, and to investigate the dynamics of convective currents development and their structure.

In this paper, we propose a design model for studying the formation of structured flows in isothermal gas mixtures at different pressures over time through 2D modeling in a vertical cylindrical channel of finite dimensions. A comparison is made between the experimental and theoretical results.

2. MATHEMATICAL MODEL OF DIFFUSION INSTABILITY AND NUMERICAL 2D ALGORITHM

2.1 Basic Equations

The macroscopic motion of an isothermal three-component gas mixture is described by the general system of equations involving the Navier-Stokes equations, conservation equations of the particle number of mixture, and components in the Boussinesq approximation. Taking into consideration the conditions of independent diffusion during which $\sum_{i=1}^{3} \vec{j}_i = 0$ and $\sum_{i=1}^{3} c_i = 1$ for an isothermal gas mixture, the system of equation by analogy with Kossov

et al. (2017) can be written as

$$\begin{aligned}
\rho\left[\frac{\partial\vec{u}}{\partial t}+(\vec{u}\,\nabla)\,\vec{u}\right] &= -\nabla p+\eta\Delta\vec{u}+\left(\frac{\eta}{3}+\xi\right)\nabla\mathrm{div}\vec{u}+\rho\vec{g},\\
\frac{\partial n}{\partial t} &= -\mathrm{div}(n\vec{v}),\\
\frac{\partial c_1}{\partial t}+\vec{v}\,\nabla c_1 &= \mathrm{div}[D_{11}^{*}\nabla c_1+D_{12}^{*}\nabla c_2],\\
\frac{\partial c_2}{\partial t}+\vec{v}\,\nabla c_2 &= \mathrm{div}[D_{21}^{*}\nabla c_1+D_{22}^{*}\nabla c_2].
\end{aligned} \tag{1}$$

Here $\vec{u}$ is the weight-average velocity vector; $\vec{v}$ is the number-average velocity vector; ρ is the density; p is the pressure; η and ξ are the coefficients of shear and bulk viscosity; $\vec{g}$ is the gravitational acceleration vector; n is the number density; t is the time; c_i is the ith component concentration; $\vec{j}_i$ is the diffusion flux density of the ith component; D_{ij}^{*} is the practical diffusion coefficients, which are determined by the mutual diffusion coefficients D_{ij} (Bird et al., 2002):

$$\begin{aligned}
D_{11}^{*} &= \frac{D_{13}\left[c_1D_{32}+(c_2+c_3)\,D_{12}\right]}{D},\\
D_{12}^{*} &= -\frac{c_1D_{23}\,(D_{12}-D_{13})}{D},\\
D_{22}^{*} &= \frac{D_{23}\left[c_2D_{13}+(c_1+c_3)\,D_{12}\right]}{D},\\
D_{21}^{*} &= -\frac{c_2D_{13}\,(D_{12}-D_{23})}{D},\\
D &= c_1D_{23}+c_2D_{13}+c_3D_{12}.
\end{aligned}$$

The relationship between the total flux of the ith component and the gaseous mixture velocity can be written as

$$\vec{v}=\sum\vec{j}_i/c_0,\quad \vec{u}=\sum m_i\vec{j}_i/\rho,$$

where m_i is the molecular mass of the ith component, $\rho=\sum m_ic_i$, $c_0=\sum_{i=1}^{n}c_i$.

Equation (1) shall be supplemented by the equation of the medium state

$$\rho=\rho(c_1,c_2,p),\quad T=const. \tag{2}$$

To solve Eqs. (1) and (2) it is necessary to determine the relationship between the weight-average $\vec{u}$ and number-average $\vec{v}$ velocities. We will consider that the gas molecules of the ith component are dispersed ($i\in K_{dispersed}$) and the rest of the particles are carried ($i\in K_{carrier}$). Carried phases move with the same velocity as $v^{carrier}$:

$$v_i=v^{carrier},\quad i\in K_{carrier}. \tag{3}$$

The Stokes law determines the velocity difference between the dispersed and carried phases:

$$v_i=v^{carrier}+u_i^{settling},\quad i\in K_{carrier}. \tag{4}$$

The settling velocity of the ith component $u_i^{settling}$ under the laminar flow regime takes the form

$$u_i^{settling}=\frac{\left(\rho_i^0-\rho\right)\cdot d_i^2}{18\eta}g, \tag{5}$$

where d_i is the diameter of the ith component of the gas. In the case of the turbulent flow regime the settling velocity $u_i^{settling}$ can be calculated as

$$u_i^{settling} = \sqrt{\frac{4\left(\rho_i^0 - \rho\right) \cdot d_i^2}{3\rho C}} g, \tag{6}$$

where the aerodynamic drag coefficient C is found from the following formulae: $C = 24/\mathrm{Re}$ at $10^{-4} < \mathrm{Re} < 2$ (laminar regime), $C = 18.5/\mathrm{Re}^{0.6}$ at $2 < \mathrm{Re} < 500$ (transient regime), and $C = 0.44$ at $500 < \mathrm{Re} < 10^5$ (turbulent regime). Ostrovsky (2000) has suggested one of the successful approximations of the Reynolds number.

The semiempirical dependence (Aerov and Todes, 1968) can be recommended for the approximate calculations of the settling velocity of the ith component $u_i^{settling}$. This dependence is valid for all flow regimes and has the following form:

$$C = 8\left(\frac{3}{\mathrm{Re}} + \frac{0.45}{\mathrm{Re}^{4/9}} + 0.042\right), \quad \mathrm{Re} = \frac{\mathrm{Ar}}{18 + 0.61\sqrt{\mathrm{Ar}}},$$

where $\mathrm{Ar} = \left(d_i^3 \rho \left(\rho_i^0 - \rho\right) g\right)/\eta^2$ is the Archimedes number.

The weight-average velocity can be presented as

$$u = \sum_{i=1}^{n} c_i v_i \tag{7}$$

Substituting expressions (3) and (4) in formula (7) yields

$$u = v^{carrier} + \sum_{i \in K_{dispersed}} c_i u_i^{settling}. \tag{8}$$

Formula (8) gives the velocity of the carried phases in the following form:

$$v^{carrier} = u - \sum_{i \in K_{dispersed}} c_i u_i^{settling}. \tag{9}$$

Velocities of the ith component are calculated explicitly from Eq. (4) as

$$v_i = v^{carrier} + u_i^{settling}, \quad i \in K_{carrier}. \tag{10}$$

If we take into consideration the diameters of the gas molecules [gas-kinetic diameter of the studied gas molecules are as follows: $d(\mathrm{N_2}) = 0.37$ nm, $d(\mathrm{Ar}) = 0.36$ nm, $d(\mathrm{He}) = 0.215$ nm, $d(\mathrm{CH_4}) = 0.444$ nm] then the settling velocity of the ith component is $u_i^{settling} \approx 10^{-15}$. Hence, according to formulae (9) and (10) the velocity of the ith component is equal to the weight-average velocity, i.e., $v_i = u$.

Using the method of small disturbances and taking into account that $v_i = u$ the system (1) takes the form

$$\begin{aligned}
\frac{\partial \vec{u}}{\partial t} + \vec{u}\,\nabla\vec{u} &= -\frac{1}{\rho_0}\nabla p + \nu\nabla^2\vec{u} + g\left(\beta_1 c_1 + \beta_2 c_2\right)\vec{\gamma}, \\
\mathrm{div}\vec{u} &= 0, \\
\frac{\partial c_1}{\partial t} + \vec{u}\,\nabla c_1 &= D_{11}^*\nabla^2 c_1 + D_{12}^*\nabla^2 c_2, \\
\frac{\partial c_2}{\partial t} + \vec{u}\,\nabla c_2 &= D_{21}^*\nabla^2 c_1 + D_{22}^*\nabla^2 c_2,
\end{aligned} \tag{11}$$

where $\nu = \eta/\rho$ is the kinematic viscosity, and β_i is the concentration expansion coefficient.

We will write this system in the dimensionless form normalizing by the scales of length $x_1^* = x/H$, $x_2^* = z/H$; time $\tau = t\nu/H^2$; velocity $u_1^* = uH/D_{22}^*$, $u_2^* = wH/D_{22}^*$; and pressure $p^* = pH^2/\rho_0\nu D_{22}^*$ (ρ_0 is a characteristic

medium density corresponding to the mean values of the concentration), concentrations of the ith component $c_1^* = c_1/A_1H$, $c_2^* = c_2/A_2H$ (omitting the dimensionless sign *) as follows:

$$\begin{aligned}
&\frac{\partial u_1}{\partial \tau} + \frac{1}{\mathrm{Pr}_{22}} u_1 \frac{\partial u_1}{\partial x_1} + \frac{1}{\mathrm{Pr}_{22}} u_2 \frac{\partial u_1}{\partial x_2} = -\frac{\partial p}{\partial x_1} + \frac{\partial^2 u_1}{\partial x_1^2} + \frac{\partial^2 u_1}{\partial x_2^2} + \mathrm{Ra}_1 c_1,\\
&\frac{\partial u_2}{\partial \tau} + \frac{1}{\mathrm{Pr}_{22}} u_1 \frac{\partial u_2}{\partial x_1} + \frac{1}{\mathrm{Pr}_{22}} u_2 \frac{\partial u_2}{\partial x_2} = -\frac{\partial p}{\partial x_1} + \frac{\partial^2 u_2}{\partial x_1^2} + \frac{\partial^2 u_2}{\partial x_2^2} + \mathrm{Ra}_2 c_2,\\
&\frac{\partial c_1}{\partial \tau} + \frac{1}{\mathrm{Pr}_{22}} u_1 \frac{\partial c_1}{\partial x_1} + \frac{1}{\mathrm{Pr}_{22}} u_2 \frac{\partial c_1}{\partial x_2} = \frac{1}{\mathrm{Pr}_{11}} \frac{\partial^2 c_1}{\partial x_1^2} + \frac{1}{\mathrm{Pr}_{11}} \frac{\partial^2 c_1}{\partial x_2^2} + \frac{1}{\mathrm{Pr}_{12}} \frac{\partial^2 c_2}{\partial x_1^2} + \frac{1}{\mathrm{Pr}_{12}} \frac{\partial^2 c_2}{\partial x_2^2},\\
&\frac{\partial c_2}{\partial \tau} + \frac{1}{\mathrm{Pr}_{22}} u_1 \frac{\partial c_2}{\partial x_1} + \frac{1}{\mathrm{Pr}_{22}} u_2 \frac{\partial c_2}{\partial x_2} = \frac{1}{\mathrm{Pr}_{21}} \frac{\partial^2 c_1}{\partial x_1^2} + \frac{1}{\mathrm{Pr}_{21}} \frac{\partial^2 c_1}{\partial x_2^2} + \frac{1}{\mathrm{Pr}_{22}} \frac{\partial^2 c_2}{\partial x_1^2} + \frac{1}{\mathrm{Pr}_{22}} \frac{\partial^2 c_2}{\partial x_2^2},\\
&\frac{\partial u_1}{\partial x_1} + \frac{\partial u_2}{\partial x_2} = 0.
\end{aligned} \tag{12}$$

Here $\mathrm{Pr}_{ii} = \nu/D_{ii}^*$ is the diffusion Prandtl number; $\mathrm{Ra}_1 = g\beta_1 A_1 H^4/D_{22}^*\nu$, $\mathrm{Ra}_2 = g\beta_2 A_2 H^4/D_{22}^*\nu$ are the partial Rayleigh numbers ($\mathrm{Ra} = \mathrm{Gr} \cdot \mathrm{Pr}$, where Gr is the Grashof number).

The initial conditions and the boundary conditions for the system of equations (12) are shown in Table 1.

2.2 Numerical Algorithm

As seen from the experimental data (Kossov et al., 2017; Kosov et al., 2017), mixing is carried out in the channel, which has, as a rule, a cylindrical shape. Heavy- and light-density components of a binary mixture are in the upper part of the channel, and the gas with intermediate density is in the lower part of the channel. To simplify the tasks we consider a two-dimensional cross-section area of cylindrical sphere $H \times d$ in the Cartesian coordinate system (x, z), where H is the height of the cylindrical channel, and $d = 2r$ is the diameter [Fig. 1(a)]. To register the isoconcentrational lines characterizing the emergence and development of convection, it is sufficient to consider the part of this area ($H/d \gg 1$), where the diffusion of components takes place [Fig. 1(b)] ignoring the other areas of the two-dimensional area.

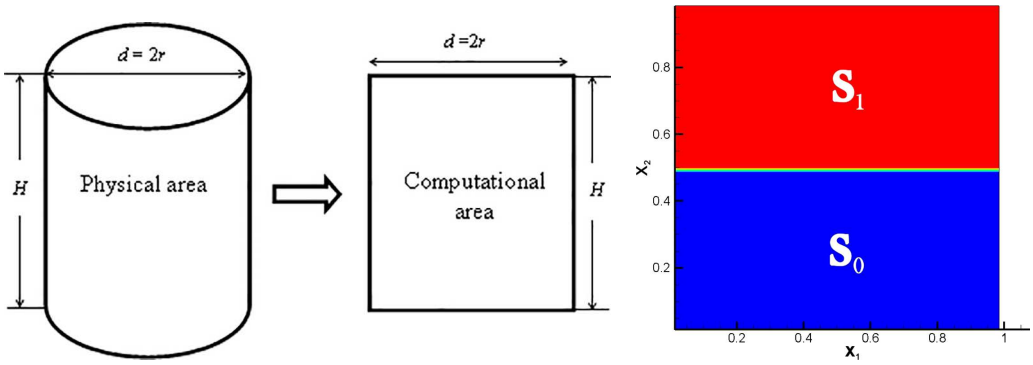
For the numerical solution of the equations system (12) we use the splitting scheme on physical parameters. The spatial derivatives are approximated on the uniform spatial grid. The time derivatives are approximated by differences ahead with the first order.

On the first stage, the transference of the number of motion is done due to the convection and diffusion. The intermediate velocity field is solved by the five-point sweep method specified by Navon (1987) with the fourth order of accuracy in space and the third order of accuracy with respect to time using the explicit scheme of Adams-Bashforth for convective terms and the implicit scheme of Crank-Nicolson for the diffusion members defined by (Kim and Moin, 1985)

$$\frac{\overline{u}^* - \overline{u}^n}{\tau} = -\frac{1}{\mathrm{Pr}_{22}} \overline{u}^n \nabla \overline{u}^* + \Delta \overline{u}^* + \mathrm{Ra}_1 c_1 + \mathrm{Ra}_2 c_2. \tag{13}$$

TABLE 1: Initial and boundary conditions

Initial Conditions		**Boundary Conditions**
$u_i(x_1, x_2, \tau = 0) = 0, \quad i = 1, 2$		$u_i(x_1, x_2, \tau) = 0, \quad i = 1, 2$
$c_1\big\vert_{\tau=0,(x_1,x_2)\in S_1} = X_1;$	$c_1\big\vert_{\tau=0,(x_1,x_2)\in S_0} = 0$	$\dfrac{\partial c_i}{\partial n} = 0, \quad i = 1, 2$
$c_2\big\vert_{\tau=0,(x_1,x_2)\in S_1} = X_2;$	$c_2\big\vert_{\tau=0,(x_1,x_2)\in S_0} = 0$	—
$c_3\big\vert_{\tau=0,(x_1,x_2)\in S_1} = 0;$	$c_3\big\vert_{\tau=0,(x_1,x_2)\in S_0} = 0$	—



(a) (b)

FIG. 1: Modeling of the studied process. (a) Display of areas; (b) the initial conditions for the three-component mixture He + Ar – N_2

On the second stage, based on the found intermediate velocity field, there is the pressure field. The intermediate velocity field is found using the fractional step method. By analogy with Abdibekova et al. (2014) we have used the sweep method at each stage of the sweep fractional step method to find the stage significance of the intermediate field speed:

$$\Delta p = \frac{\nabla \overline{u}^*}{\tau}. \tag{14}$$

On the third stage, it is assumed that transference is done only due to the pressure gradient, where the final velocity field is recalculated:

$$\frac{\overline{u}^{n+1} - \overline{u}^*}{\tau} = -\nabla p. \tag{15}$$

On the fourth stage the concentration of mixture components is calculated on the basis of the five-point sweep method using the Adams-Bashforth scheme taking into account the found velocity fields:

$$\frac{\overline{c}_1^{n+1} - \overline{c}_1^n}{\tau} = -\frac{1}{\mathrm{Pr}_{22}} \left(\overline{u}^{n+1}\nabla\right) \overline{c}_1^* + \frac{1}{\mathrm{Pr}_{11}} \Delta \overline{c}_1^* + \frac{1}{\mathrm{Pr}_{12}} \Delta \overline{c}_2^*, \tag{16}$$

$$\frac{\overline{c}_2^{n+1} - \overline{c}_2^n}{\tau} = -\frac{1}{\mathrm{Pr}_{22}} \left(\overline{u}^{n+1}\nabla\right) \overline{c}_2^* + \frac{1}{\mathrm{Pr}_{21}} \Delta \overline{c}_1^* + \frac{1}{\mathrm{Pr}_{22}} \Delta \overline{c}_2^*. \tag{17}$$

The results of numerical studies, obtained by the described approach, were compared with quantitative values of the experimental data obtained in the previous paragraph.

3. THE RESULTS OF NUMERICAL MODELING

For the numerical study, the following three-component gas mixtures were chosen: 0.65 CH_4 (1) + 0.35 Ar (2) – N_2 (3) and 0.4 He (1) + 0.6 Ar (2) – N_2 (3). Experimental studies (Kosov et al., 2017) for the system 0.4 He (1) + 0.6 Ar (2) – N_2 (3) showed that the transition from a stable diffusion process to concentration gravitational convection occurs at a pressure of 1.07 MPa. Analysis of the experimental data for the system 0.65 CH_4 (1) + 0.35 Ar (2) – N_2 (3) (Zhavrin et al., 2016) indicates that a stable diffusion process takes place in the system at all experimental pressures. The calculations were carried out on a uniform rectangular staggered grid with the number of nodes 250 × 250 along the axes x_1, x_2, respectively. The time step was selected to be 0.001. The main assumption in modeling is to limit the two-dimensional flows. The value of the Rayleigh number was varied in the range $12.88 < \mathrm{Ra}_1 < 77.27$, $12.14 < \mathrm{Ra}_2 < 72.86$; the values of the Prandtl number were $\mathrm{Pr}_{11} = 16.56$, $\mathrm{Pr}_{22} = 29.10$.

Figure 2 gives the velocity profile in section $x_2 = 0.5$ at different pressures. For the systems 0.65 CH_4 (1) + 0.35 Ar (2) – N_2 (3) in which the coefficients $D_{12} \approx D_{13} \approx D_{23}$, the velocity profiles are unchanged at different pressures.

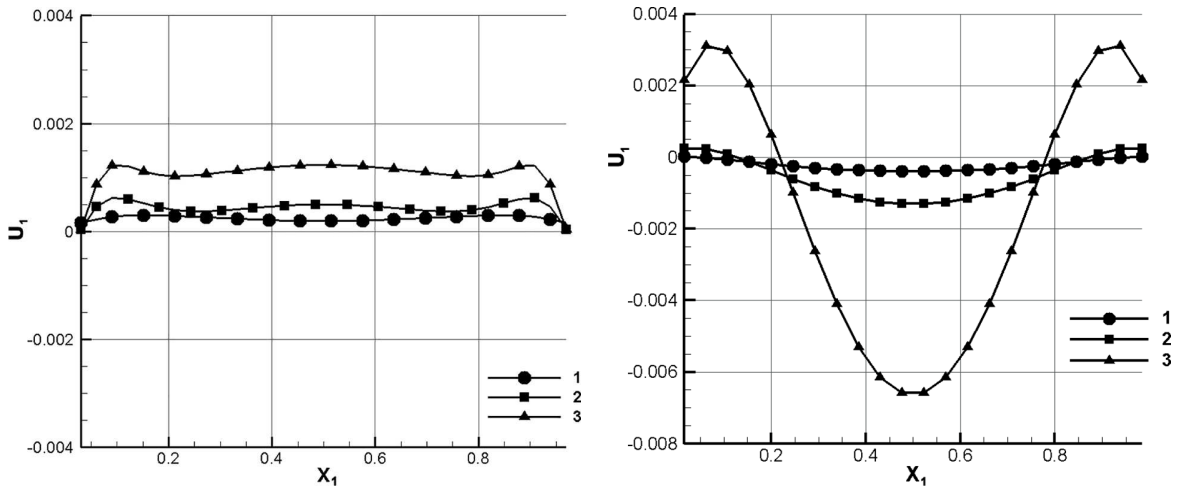


FIG. 2: Vertical velocity profiles in the middle of computational area at time $t = 2$ s for the three-component systems at different pressures; (a) 0.65 CH_4 (1) + 0.35 Ar (2) – N_2 (3), $T = 298.0$ K, (1) $p = 0.5$ MPa at $Ra_1 = 12.88$, $Ra_2 = 12.14$; (2) $p = 1.5$ MPa at $Ra_1 = 38.64$, $Ra_2 = 36.43$; (3) $p = 3.0$ MPa for $Ra_1 = 77.27$, $Ra_2 = 72.86$; (b) 0.4 He (1) + 0.6 Ar (2) – N_2 (3), $T = 298.0$ K, (1) $p = 0.5$ MPa, $Ra_1 = 4.06$, $Ra_2 = 8.92$; (2) $p = 1.5$ MPa, $Ra_1 = 12.20$, $Ra_2 = 26.77$; (3) $p = 2.0$ MPa, $Ra_1 = 16.27$, $Ra_2 = 35.70$

For the system 0.4 He (1) + 0.6 Ar (2) – N_2 (3) where D_{ij} significantly differ from each other with increasing pressure, the velocity profile becomes nonlinear.

The velocity profile at pressure 2.0 MPa is significantly nonlinear; it shows that in the system outgoing and rising convective flows might take place, which is in agreement with the experimental data shown in Zhavrin et al. (2016) and Kosov et al. (2017). Figure 3 shows isoconcentration lines characterizing the type of transport at different instants of time. In the system 0.65 CH_4 (1) + 0.35 Ar (2) – N_2 (3) the mass transfer occurs by diffusion [Figs. 3(a) and 3(b)]. At various isoconcentrations bending lines are virtually absent. For the system 0.4 He (1) + 0.6 Ar (2) – N_2 (3) quite another picture is observed. Prior to pressure $p \sim 0.5$ MPa ($Ra_1 = 4.07$, $Ra_2 = 8.92$) the picture of isoconcentration lines is similar to those shown in Figs. 3(a) and 3(b). The diffusive transfer is carried out in the system. Further pressure increase brings instability to the system. Isoconcentrational lines are curved [Fig. 3(d)] and the transfer from the diffusion mode to convective takes place. Thus, the main feature of the change of the mode "diffusion – convection" is the manifestation of the curvature of isoconcentrational lines. The critical value of the pressure of the difference problem on the 250×250 grid lies between 0.9 and 1.5 MPa, which corresponds to the experimental data described for the system 0.4 He (1) + 0.6 Ar (2) – N_2 (3) in Kosov et al. (2017).

Convection did not develop in calculations with the numbers $Ra_1 < 7.3199$ and $Ra_2 < 16.0646$. Convection was recorded at $p = 1$ MPa for $Ra_1 = 8.13$ and $Ra_2 = 17.85$. The critical values of the Rayleigh number (Ra_{cr}) of the difference problem on the 250×250 grid lie between $7.3199 \le Ra_1 \le 12.1999$ and $16.0646 \le Ra_2 \le 26.7744$, and less than the theoretical value Ra_{theor} of 67.95 for the vertical cylindrical channel, obtained under the linear stability theory (Gershuni and Zhukhovitskii, 1976). The reason for this mismatch is the small number of mesh nodes that occur on the convective structure characteristic of the critical flow, the size of which is determined by the critical wave number $\gamma_{cr} = 2.871$ (here, two nodes per shaft), and as a result there is high mesh viscosity. However, the solutions obtained on coarse grids should still satisfy the basic integral dependences derived from the differential equations, with the only difference being that the critical value of the Rayleigh number of the difference problem has a value different from the differential one, obtained in the framework of the linear stability theory.

Figure 4 shows the fields of concentrations at pressure $p = 2.5$ MPa for $Ra_1 = 20.33$, $Ra_2 = 44.62$, and various times. Calculation results show that at the starting stage the flow occurs with small speeds. As a result of the mixing process progress with time, the mixing process intensifies, which leads to a substantial distortion of the

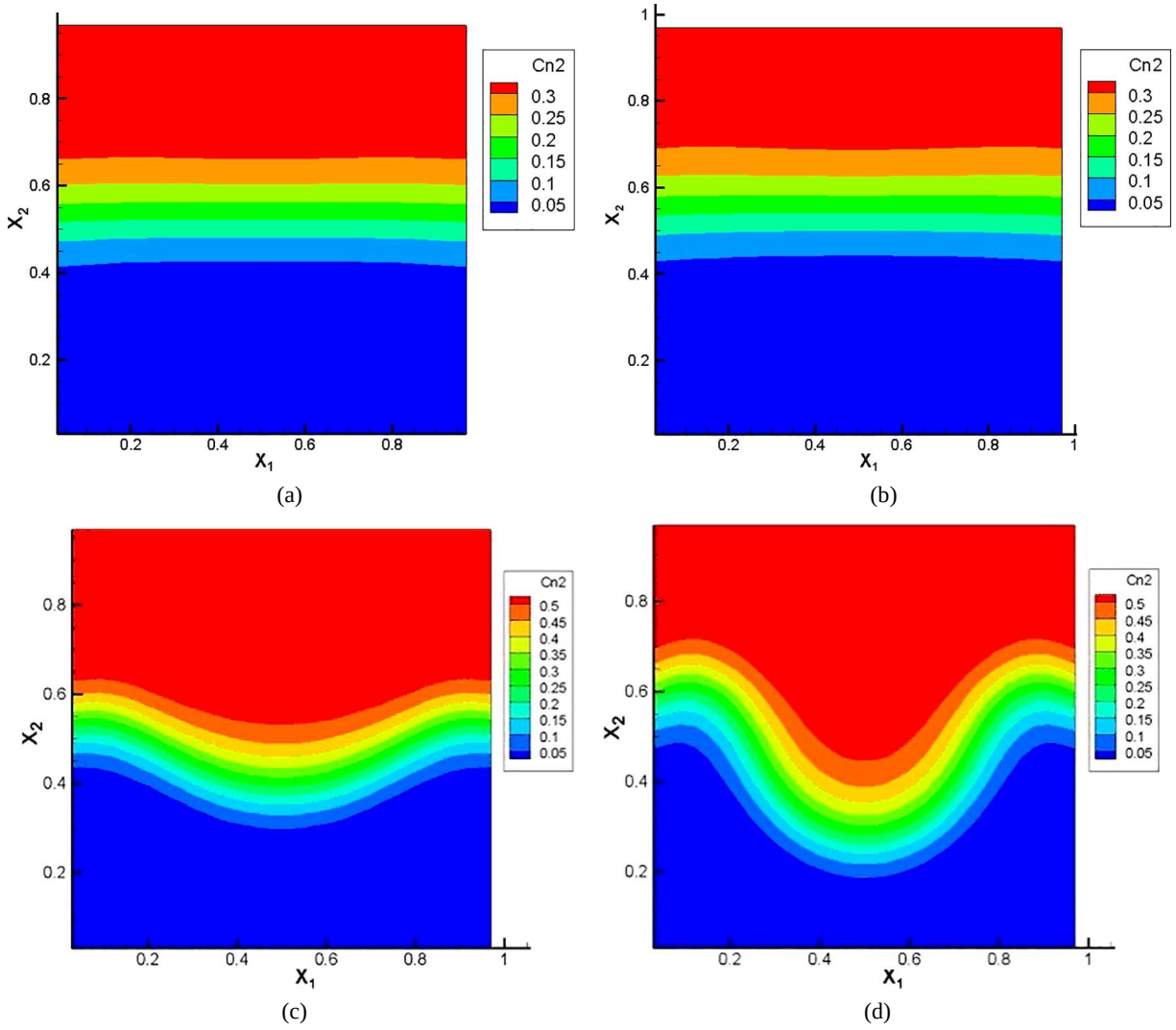


FIG. 3: Concentration fields at various pressures for the system 0.65 CH_4 (1) + 0.35 Ar (2) – N_2 (3), $T = 298.0$ K: (a) $p = 1.5$ MPa at $Ra_1 = 38.64$, $Ra_2 = 36.43$, $t = 13.5$ s; (b) $p = 2.0$ MPa at $Ra_1 = 51.51$, $Ra_2 = 48.57$, $t = 18$ s; and for the system 0.4 He (1) + 0.6 Ar (2) – N_2 (3): (c) $p = 1.5$ MPa at $Ra_1 = 12.20$, $Ra_2 = 26.77$, $t = 13.5$ s; (d) $p = 2.0$ MPa at $Ra_1 = 16.27$, $Ra_2 = 35.70$, $t = 18$ s

isoconcentration lines and an increase in the transfer rate [Figs. 4(c) and 4(d)]. Loss of time of mechanical stability of mixture for the given system is tens of seconds, which is comparable with the experimental data shown in Kosov et al. (2015). Then there was observed the rapid development of convective currents. Convective flow is realized in the form of large scaled structures with their subsequent annihilation. Further, a similar revival of structured convection currents takes place, which leads to the pulsation of the medium on the surface of the flow. Periodically the ("drip") blending mode is clearly fixed on the experimental data shown in Kosov et al. (2015). At the final stage of mixing, the convection is attenuated and multicomponent transfer is carried out in a diffusion mode.

Thus the comparison results show that the used model and the method of calculation allows us to determine the parameters of regime change "diffusion-concentration convection" and to obtain reliable data on the concentration fields and the speed transfer in the regime of advanced convective instability in isothermal ternary gas mixtures.

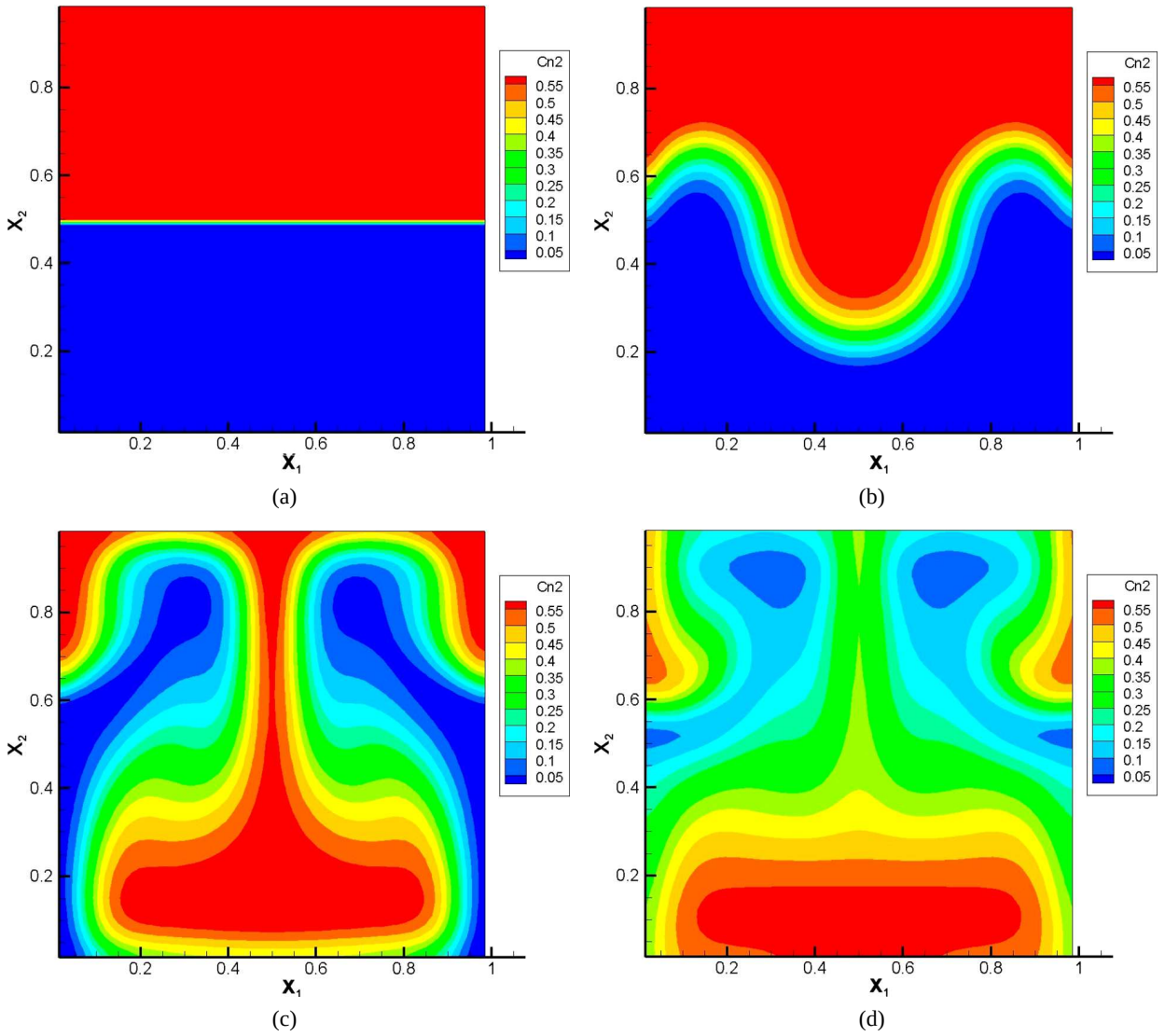


FIG. 4: Concentration fields for the argon at different times of mixing in the system 0.4 He (1) + 0.6 Ar (2) – N_2 (3), $p = 2.5$ MPa for $Ra_1 = 20.33$, $Ra_2 = 44.62$, $T = 298.0$ K: (a) $t = 0$ s; (b) $t = 10.55$ s; (c) $t = 20.30$ s; (d) $t = 30.05$ s

4. CONCLUSIONS

The stability of mechanical equilibrium in isothermal gas mixtures is studied numerically. The mechanical equilibrium in the mixture is disturbed at certain pressures and the system undergoes a transition between the two mixing regimes—isothermal diffusion to gravitational concentration convection. A mathematical model is proposed for numerical modeling of complex mass transfer in isothermal three-component gas mixtures at various pressures. The pressure influence on the systems where the appearance of convection is associated with the instability of mechanical equilibrium of the mixture is numerically studied. The pressure values associated with the diffusion-to-convection transition are obtained. The results of the calculation for determining the diffusion regions and gravitational concentration convection areas in the ternary gaseous mixture as a function of pressure are in satisfactory agreement with the experimental data.

ACKNOWLEDGMENTS

The work was supported by the Science Committee of the Ministry of Education and Science of the Republic of Kazakhstan (Project No. AP05130712).

REFERENCES

Abdibekova, A.U., Zhakebayev, D.B., and Zhumagulov, B.T., The Decay of MHD Turbulence Depending on the Conducting Properties of Environment, *Magnetohydrodynamics*, vol. **50**, no. 2, pp. 121–138, 2014.

Aerov, M.E. and Todes, O.M., *Hydraulic and Thermal Foundations for the Operation of Devices with a Stationary and Boiling Granular Layer*, Leningrad: Khimiya, pp. 25–125, 1968.

Ankusheva, N.B., Kossov, V.N., and Seleznev, V.D., Effect of Diffusion Channel Inclination on Stability of Mechanical Equilibrium in Isothermal Binary Gas Mixtures, *J. Appl. Mech. Tech. Phys.*, vol. **51**, no. 1, pp. 62–64, 2010.

Bird, R.B., Stewart, W.E., and Lightfoot, E.N., *Transport Phenomena*, New York, NY: John Wiley & Sons, Inc., pp. 513–538, 2002.

Dil'man, V.V., Lipatov, D.A., Lotkhov, V.A., and Kaminskii, V.A., Instability in Unsteady-State Evaporation of Binary Solutions into an Inert Gas, *Theor. Found. Chem. Eng.*, vol. **39**, no. 6, pp. 566–572, 2005.

Gershuni, G.Z. and Zhukhovitskii, E.M., *Convective Stability of Incompressible Fluids*, Jerusalem: Keter, pp. 7–137, 1976.

Joseph, D.D., *Stability of Fluid Motions I, II*, Berlin-Heidelberg/New York, NY: Springer-Verlag, 1976.

Kaminskii, V.A. and Obvintseva, N.Yu., Evaporation Regimes of Binary Solutions, *Theor. Found. Chem. Eng*., vol. **41**, no. 5, pp. 512–518, 2007.

Kim, J. and Moin, P., Application of a Fractional-Step Method to Incompressible Navier-Stokes Equations, *J. Comput. Phys.*, vol. **59**, no. 2, pp. 308–323, 1985.

Kosov, V.N., Seleznev, V.D., and Zhavrin, Yu.I., Separation of Components during Isothermal Mixing of Ternary Gas Systems under Free Convection Conditions, *Tech. Phys*., vol. **42**, no. 10, pp. 1236–1237, 1997.

Kosov, V.N., Seleznev, V.D., and Zhavrin, Yu., I., Oscillatory and Monotonic Instability at the Boundary of the Transition 'Molecular Diffusion-Concentration Convection' in Ternary Gas Mixtures, *J. Eng. Phys. Thermophys.*, vol. **73**, no. 2, pp. 303–310, 2000.

Kosov, V.N., Fedorenko, O.V., and Zhavrin, Yu.I., Convective Instability in Isothermal Ternary Gas Mixtures at Various Pressures, in *Proc. of 7th Baltic Heat Transf. Conf.*, Tallinn, University of Technology, pp. 207–212, 2015.

Kosov, V.N., Kul'zhanov, D.U., Zhavrin, Y.I., and Fedorenko, O.V., Emergence of Convective Flows during Diffusional Mass Transfer in Ternary Gas Systems: The Effect of Component Concentrations, *Russ. J. Phys. Chem. A*, vol. **91**, no. 6, pp. 984–989, 2017.

Kossov, V., Krasikov, S., and Fedorenko, O., Diffusion and Convective Instability in Multicomponent Gas Mixtures at Different Pressures, *Europe. Phys. J.: Spec. Top.*, vol. **226**, no. 6, pp. 1177–1187, 2017.

Linden, P.F., Redondo, J.M., and Youngs, D.I., Molecular Mixing in Rayleigh-Taylor Instability, *J. Fluid Mech.*, vol. **265**, pp. 97–124, 1994.

Lotkhov, V.A., Dil'man, V.V., Vasilevskii, D.I., and Lipatov, D.A., Dynamics of Unsteady-State Evaporation of Binary Mixtures in Closed Systems, *Theor. Found. Chem. Eng*., vol. **37**, no. 2, pp. 131–136, 2003.

Navon, M., Pent: A Periodic Penta-Diagonal Systems Solver, *Commun. Appl. Numer. Methods*, vol. **3**, no. 1, pp. 63–69, 1987.

Nield, D.A. and Bejan, A., *Convection in Porous Media*, New York, NY: Springer, pp. 189–255, 2006.

Ostrovsky, G.M., *Applied Mechanics of Inhomogeneous Medium*, St. Petersburg, Russia: Nauka, pp. 167–214, 2000.

Seleznev, V.D., Kosov, V.N., Poyarkov, I.V., Fedorenko, O.V., and Beketaeva, M.T., Double Diffusion in Ar-N_2 Binary Gas System at the Constant Value of Temperature Gradient, *Acta Phys. Pol., A*, vol. **123**, no. 1, pp. 62–66, 2013.

Taylor, R. and Krishna, R., *Multicomponent Mass Transfer*, New York, NY: John Wiley & Sons, Inc., pp. 95–123, 1993.

Zhavrin, Yu.I. and Kosov, V.N., Effect of Temperature on Diffusional Instability, *J. Eng. Phys. Thermophys. (Moscow, Russ. Fed.)*, vol. **55**, no. 1, pp. 774–778, 1988.

Zhavrin, Yu.I., Kosov, V.N., Fedorenko, O.V., and Akzholova, A.A., Some Features of the Multicomponent Gas Transfer in the Convective Instability of Gas Mixture, *Theor. Found. Chem. Eng.*, vol. **50**, no. 3, pp. 171–177, 2016.

Zhavrin, Yu.I., Kosov, V.N., Kul'zhanov, D.U., and Karataeva, K.K., Effect of the Pressure on the Type of Mixing in a Three-component Gas Mixture Containing a Component Possessing the Properties of a Real Gas, *Tech. Phys. Lett.*, vol. **26**, no. 12, pp. 1108–1109, 2000.