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

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**ABSTRACT** Isothermal multicomponent diffusion in three-component gas mixtures He + Ar – $N_2$ and $CH_4$ + Ar – $N_2$ at different pressures and certain concentrations of components in the binary mixtures is experimentally studied. It is shown that convective instability, which significantly intensifies the multicomponent mass transfer, occurs due to the pressure increase in systems where diffusion coefficients are significantly different from each other. Parameters characterizing the transition of diffusion mixing into the convective one can be determined by the numerical simulation methods. Modelling of the multi-component mixing process is carried out by means of the joint solution of the Navier-Stokes equations, diffusion equations and continuity equation. It is shown that essentially non-linear distributions of the component concentrations that lead to non-linear distribution of the density of the gas mixture, which is responsible for the formation of convective structures, arises at a certain pressure in the system.

## 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 [$m/s$]
$\vec{\mathrm{v}}$ = number-average velocity vector [$m/s$]
$\alpha$ = dimensionless parameter defined by the ratio of experimental values of concentrations $c_{\exp}$ to the concentrations $c_{\mathrm{theor}}$ calculated by Stefan-Maxwell equations [-]
$\beta_i$ = coefficient of thermal expansion [$m/K$]
$\vec{\gamma}$ = unit vector [-]
$\eta$ = shear viscosity [$Pa\ s$]

$\xi$ = bulk viscosity [$Pa\ s$]

**Subscripts and Superscripts**

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

## INTRODUCTION

Taylor and Krishna [1993] have shown that in multicomponent gas mixtures there are wide variety of change of the modes. The intensity of the heat and mass transfer in them is defined by molecular, convective, and often the joint action of these modes. At the same time, practically it is not taken into consideration the fact that the process of molecular diffusion can lead to instability of mechanical equilibrium of mixture, with further occurrence of natural convection studied by Linden et al. [1994] and Kaminskii and Obvintseva [2007] that significantly intensifies the overall mass transfer. While the occurrence and development of the concentration convection takes place not only in the frame of traditional concepts of Rayleigh thermal problems described by Joseph [1976] and Nield and Bejan [2006] but also for the situations where movements occur in a stable stratification of the mixture that shown by Kossov et al. [1997] and Zhavrin et al. [2016]. In experiments on studying the mixing in multicomponent systems carried out by Kossov et al. [2014] and on researching the diffusions of mixture of solvent vapor into inert gas described by Dil'man et al. [2005] were recorded convective flows leading to synergetic effect, associated with a significant increase of speed of the mixing system components. Therefore, issues related to the definition of the mechanism of regime change “diffusion – convection” in multicomponent gas mixtures, the parameters determining the appearance of convective regime is an important tasks for convective heat mass transfer.

The paper presents experimental data on the study of regime change borders “diffusion - concentration convection” in isothermal ternary gas mixtures. Proposed calculated design model on studying the formation of structured flows in isothermal gas mixtures at different pressures over time by 2-D modeling in a vertical cylindrical channel of finite dimensions. The comparison between experimental and theoretical results is being done.

## EXPERIMENTAL RESULTS

The study of gaseous mixtures mixing was carried out by the two flasks method connected with a vertical capillary, which has successfully proved itself in the study of diffusion mass transfer at different pressures and temperatures. For example, this method was used in the experiments carried out by Dunlop and Bignell [1987]. The scheme of device where the mass transfer realizes is shown in Fig. 1 and has been used by Kossov et al. [2014] to study binary diffusion equally diluted by the hydrocarbon gases. Therefore, it is expected that the obtained experimental data under the mixing of binary mixture into third component will be accurate too.

The first part of the separation device is a system for the transportation of gaseous mixtures from the gas holders 20, 21 into the flasks of the diffusion apparatus through the system of valves 1-10, the gauges 12 that measure the pressure in the flasks, the container 13 to equalize the pressure. The gauges are fitted with specially made permeators 11. The second part of the installation is a diffusion apparatus. Subject to the gaseous mixture, two diffusion apparatuses with the different channels and volumes of flasks were used: 1. $V_1 = (214.0 \pm 0.5)\cdot 10^{-6}$ m$^3$, $V_2 = (227.0 \pm 0.5)\cdot 10^{-6}$ m$^3$, the flasks were connected with vertical cylindrical channel of length $L = (165.4 \pm 0.05)\cdot 10^{-3}$ m and diameter $d = (6.10 \pm 0.02)\cdot 10^{-3}$ m (in the experiments with the system 0.65 $CH_4$ (1) + 0.35 Ar (2) –

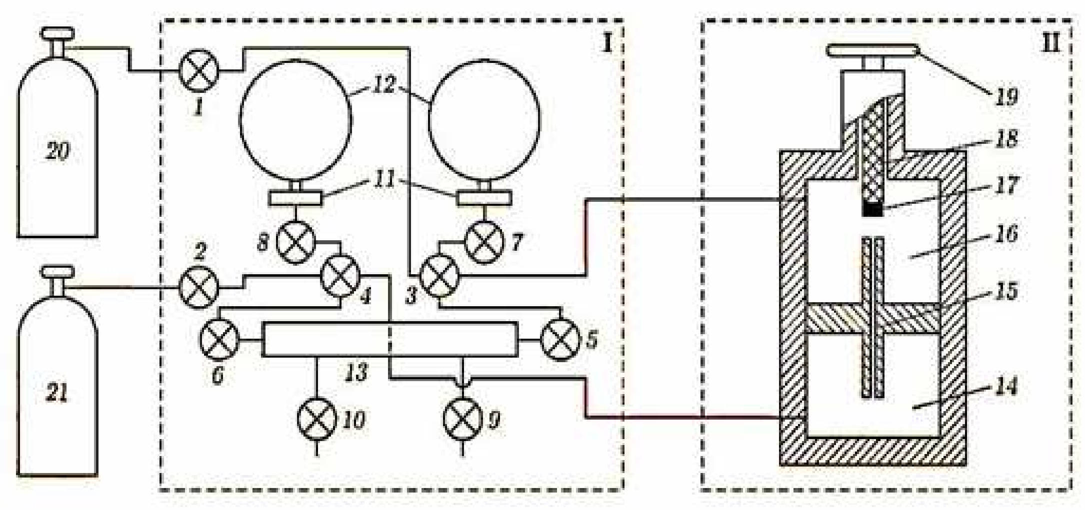
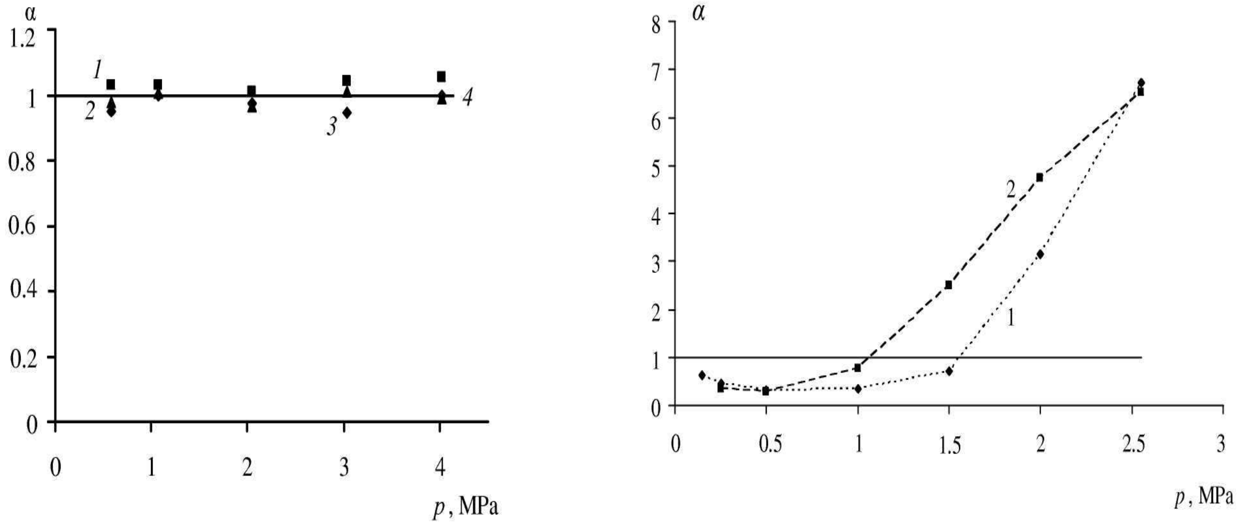


Figure 1. Block diagram of the installation of the two-flask method. The gas preparation block and the two-flask diffusion setup are indicated by I and II, respectively; 1-8 – valves; 9 – valve connected to the backing vacuum pump; 10 – valve connected to the interferometer or chromatograph; 11 – membrane dividers; 12 – reference pressure gages; 13 – equalizing tank; 14 – bottom flask; 15 – diffusion channel; 16 – top flask; 17 – fluoroplastic pellet; 18 – rod; 19 – handwheel; 20, 21 – gas holders.

$N_2$ (3)); 2. $V_1 = (60.0 \pm 0.5)\cdot 10^{-6}$ m$^3$, $V_2 = (62.1 \pm 0.5)\cdot 10^{-6}$ m$^3$, $L = (70.05 \pm 0.05)\cdot 10^{-3}$ m, $d_1 = (3.00 \pm 0.02)\cdot 10^{-3}$ m and $d_2 = (4.00 \pm 0.02)\cdot 10^{-3}$ m (in the experiments with the system 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3)).

For the study were selected the systems 0.65 $CH_4$ (1) + 0.35Ar (2) – $N_2$ (3) and 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3) (here and after lets agree that the figures in front of a chemical element determine the initial concentration of the component in the mixture, and the numbers in parentheses after the chemical element define the numbering of components). The methodology of the experiment has been described previously by Zhavrin et al. [2000] and assumed to fulfill the following procedures. The top and bottom of the flasks of the device were filled with researching gas to a pressure of the experiment. Tightness in filling the flasks ensured overlapping capillary device. At the opening of the channel connecting the flask, the time indicating the beginning of the mixing process is fixed. At the end of the experiment, the channel was blocked and the analyses of gas mixture were started. Concentration analysis of error was within the ranged, from ± 0.3 to ± 0.5%. In the upper flask of the device was binary mixture of methane (or helium) and argon, and in the bottom - nitrogen. The initial composition of researching systems was chosen so that the density of the binary mixture at a predetermined pressure range did not exceed the density of nitrogen. Researching systems have another feature. In the first mixture the interdiffusion coefficients $D_{ij}$ that described mathematically by Bird et al. [2002] are approximately equal ($D_{12} \approx D_{13} \approx D_{23}$), and in the second diffusion coefficients of hydrogen and argon in nitrogen is differ approximately in four times ($D_{12} \approx D_{13} >> D_{23}$).

The pressure dependence of the components' concentrations (molar fraction) with fixed initial composition, time (1 hour), temperature was determined in the experiments with the studied systems according to the method described above. Each experiment was performed under fixed pressure. Pressure was varied from experiment to experiment. One of the bottom lines of the experimental research is a detection technique of the boundary transition from the diffusion mode to



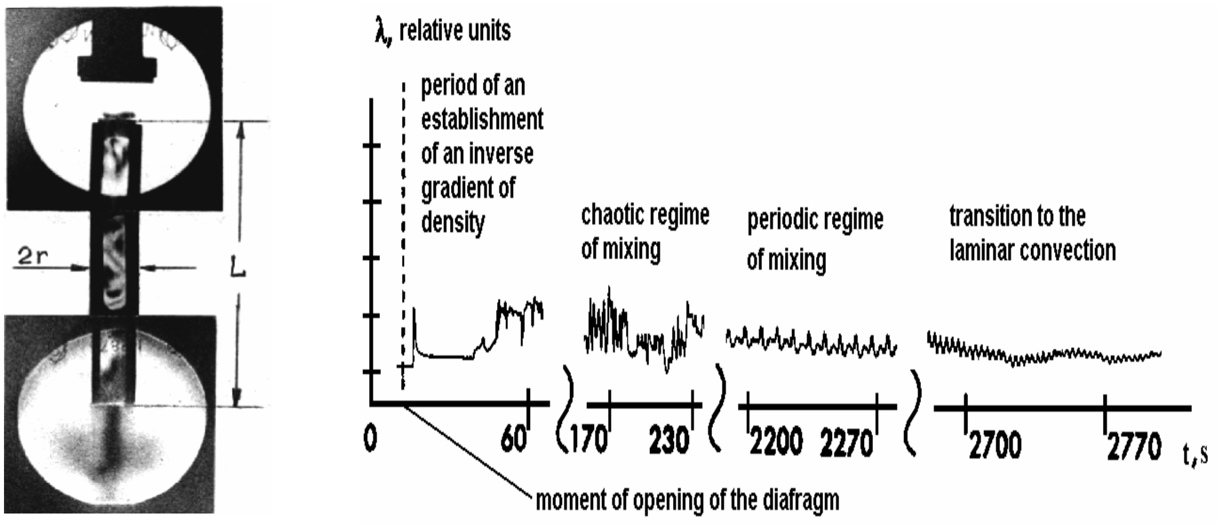
(a) (b)

Figure 2. The experimental data for diffusion and convection in isothermal ternary mixtures. a) The dependence of parameter $\alpha$ on the pressure: 0.65 $CH_4$ + 0.35 Ar – $N_2$, $T$ = 298.0 K, 1 - methane, 2 - Ar, 3 - nitrogen, 4 - calculation assuming diffusion; b) Parameter $\alpha$ at various pressure in the mixture 0.4 He + 0.6 Ar – $N_2$, $T$ = 298.0 K, 1 – $2r$ = 3 mm; 2 – $2r$ = 4 mm.

convection one for the studied system. This can be done by comparing the experimental data (for example, the concentration of diffused components $c_{iexp}$) with the calculated assuming diffusive transport according to Stefan-Maxwell equations specified by Bird et al. [2002], assuming a quasi-stationary diffusion of ternary gas mixture defined by Aleksandrov [2001].

As it seen from Fig.1b for the system 0.65 $CH_4$ (1) + 0,35Ar (2) – $N_2$ (3), where the coefficients $D_{ij}$ are comparable with each other, diffusion is observed. Experienced values of component concentrations within the experimental error coincide with the calculated. Along the entire range of pressures studied parameter $\alpha \approx 1$. In the system 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3) is observed completely different picture (Fig. 1b). At a certain pressure, the parameter $\alpha$ begins significantly exceed the unit. Further pressure increase leads to a sharp rise of the dimensionless parameter $\alpha$, which is not typical for diffusion. In this area of pressure, the experimental values of $c_{i\mathrm{exp}}$ significantly exceed the calculated, obtained assuming diffusion. A similar situation takes place for the case when under fixed conditions of the experiment the radius of cylindrical channel changes. Intensification of mass transfer is associated with the occurrence of convective mechanisms of mixing the components, caused by the instability of the mechanical equilibrium of the gas mixture. The cause of instability of mechanical equilibrium is the inversion of the gradient density, which arises due to the difference of the diffusion coefficients $D_{ij}$. It should be assumed that the arisen convection flows have a complex structure. To determine its characteristic properties the schlieren method specified by Merzkirch [1987] and the method of fast-response transducers described by Brunn [1995] are used. Figure 3 presents the results of research obtained by Kosov et al. [2015] and associated with the determination of the characteristic periods of convective regimes appeared under diffusion in ternary gas mixtures. Figure 3a shows the registered structured flows generated during the instability of mechanical equilibrium in an isothermal ternary gas mixtures. In addition, the complex structure of convective modes can be detected with the help of fast-response transducers determining a local thermal conductivity of gas.

Since the thermal conductivity of the gas mixture depends on the concentrations of components, it gives us the ability to attend to its change over time, and thus, to determine the characteristic



(a) (b)

Figure 3. Concentration convection at the isothermal diffusion in ternary gas mixtures obtained by Kosov et al [2015]. a) the shadow pictures of convective flows; b) the change of thermal conductivity of the mixture over with time, illustrating the regime change of convection.

periods of the observed modes (Fig. 3b). We can assume that the experimental data shown in Fig. 2 b correspond to the flows and the convection modes presented in Fig. 3.

Thus, in multicomponent systems with essential difference of diffusion component coefficients under certain conditions may be revealed convective mixture mechanisms.

## MATHEMATICAL MODEL OF DIFFUSION INSTABILITY AND NUMERICAL 2D-ALGORITHM

**Base 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 Kosov et al. [2014] can be written as

$$\rho\left[\frac{\partial \vec{u}}{\partial t} + (\vec{u}\nabla)\vec{u}\right] = -\nabla p + \eta\Delta\vec{u} + \left(\frac{\eta}{3} + \xi\right)\nabla \mathrm{div}\vec{u} + \rho\vec{g}$$

$$\frac{\partial n}{\partial t} = -\mathrm{div}(n\vec{\mathrm{v}}) \tag{1}$$

$$\frac{\partial c_1}{\partial t} + \vec{\mathrm{v}}\nabla c_1 = \mathrm{div}\left[D_{11}^{*}\nabla c_1 + D_{12}^{*}\nabla c_2\right]$$

$$\frac{\partial c_2}{\partial t}+\vec{\mathrm{v}}\nabla c_2=\operatorname{div}\left[D_{21}^{*}\nabla c_1+D_{22}^{*}\nabla c_2\right]$$

Here $\vec{u}$ is the weight-average velocity vector; $\vec{\mathrm{v}}$ is the number-average velocity vector; $\rho$ is the density; $p$ is the pressure; $\eta$ and $\xi$ are the coefficient 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 $i$-th component concentration; $\vec{j}_i$ is the diffusion flux density of the $i$-th component; $D_{ij}^{*}$ is the practical diffusion coefficients, which are determined by the mutual diffusion coefficients $D_{ij}$

$$D_{11}^{*}=\frac{D_{13}\left[c_1D_{32}+\left(c_2+c_3\right)D_{12}\right]}{D}$$

$$D_{12}^{*}=-\frac{c_1D_{23}\left(D_{12}-D_{13}\right)}{D}$$

$$D_{22}^{*}=\frac{D_{23}\left[c_2D_{13}+\left(c_1+c_3\right)D_{12}\right]}{D}$$

$$D_{21}^{*}=-\frac{c_2D_{13}\left(D_{12}-D_{23}\right)}{D}$$

$$D=c_1D_{23}+c_2D_{13}+c_3D_{12}$$

The relationship between the total flux of the $i$-th component and the gaseous mixture velocity can be written as

$$\vec{\mathrm{v}}=\sum\vec{j}_i/c_0 \qquad \vec{u}=\sum m_i\vec{j}_i/\rho$$

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

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

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

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

$$\mathrm{v}_i=\mathrm{v}^{carrier},\ i\in K_{carrier} \tag{3}$$

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

$$\mathrm{v}_i=\mathrm{v}^{carrier}+u_i^{settling},\ i\in K_{carrier} \tag{4}$$

Settling velocity of the *i*-th component $u_i^{settling}$ under 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 *i*-th component of gas. In the case of turbulent flow regime the settling velocity $u_i^{settling}$ can be calculated as

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

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

The semiempirical dependence of Aerov and Todes [1968] can be recommended for the approximate calculations of the settling velocity of the *i*-th component $u_i^{settling}$. This dependence is valid for all flow regimes and has the following form

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

where $Ar = \frac{d_i^3\, \rho\left(\rho_i^o - \rho\right) g}{\eta^2}$ is the criterion of Archimedes.

The weight-average velocity can be presented as

$$u = \sum_{i=1}^{n} c_i \mathrm{v}_i \tag{7}$$

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

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

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

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

Velocities of the *i*-th component are calculated explicitly from Eq. (4) as

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

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

Using the method of small disturbances and taking into account that $\mathrm{v}_i = u$ the system (1) takes the form

$$\frac{\partial \vec{u}}{\partial t} + \vec{u}\nabla\vec{u} = -\frac{1}{\rho_0}\nabla p + \nu\nabla^2\vec{u} + g\left(\beta_1 c_1 + \beta_2 c_2\right)\vec{\gamma} \tag{11}$$

$$div\vec{u} = 0$$

$$\frac{\partial c_1}{\partial t} + \vec{u}\nabla c_1 = D_{11}^*\nabla^2 c_1 + D_{12}^*\nabla^2 c_2$$

$$\frac{\partial c_2}{\partial t} + \vec{u}\nabla c_2 = D_{21}^*\nabla^2 c_1 + D_{22}^*\nabla^2 c_2$$

where $\nu = \eta/\rho$ is the kinematic viscosity, $\beta_i$ is 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 $p^* = pH^2/\rho_0\nu D_{22}^*$ ($\rho_0$ is a characteristic medium density corresponding to the mean values of the concentration), concentrations of the *i*-th component $c_1^* = c_1/A_1H$, $c_2^* = c_2/A_2H$ (omitting the dimensionless sign *)

$$\frac{\partial u_1}{\partial \tau} + \frac{1}{Pr_{22}}u_1\frac{\partial u_1}{\partial x_1} + \frac{1}{Pr_{22}}u_2\frac{\partial u_1}{\partial x_2} = -\frac{\partial p}{\partial x_1} + \frac{\partial^2 u_1}{\partial x_1^2} + \frac{\partial^2 u_1}{\partial x_2^2} + Ra_1 c_1$$

$$\frac{\partial u_2}{\partial \tau} + \frac{1}{Pr_{22}}u_1\frac{\partial u_2}{\partial x_1} + \frac{1}{Pr_{22}}u_2\frac{\partial u_2}{\partial x_2} = -\frac{\partial p}{\partial x_1} + \frac{\partial^2 u_2}{\partial x_1^2} + \frac{\partial^2 u_2}{\partial x_2^2} + Ra_2 c_2$$

$$\frac{\partial c_1}{\partial \tau} + \frac{1}{Pr_{22}}u_1\frac{\partial c_1}{\partial x_1} + \frac{1}{Pr_{22}}u_2\frac{\partial c_1}{\partial x_2} = \frac{1}{Pr_{11}}\frac{\partial^2 c_1}{\partial x_1^2} + \frac{1}{Pr_{11}}\frac{\partial^2 c_1}{\partial x_2^2} + \frac{1}{Pr_{12}}\frac{\partial^2 c_2}{\partial x_1^2} + \frac{1}{Pr_{12}}\frac{\partial^2 c_2}{\partial x_2^2} \tag{12}$$

$$\frac{\partial c_2}{\partial \tau} + \frac{1}{Pr_{22}}u_1\frac{\partial c_2}{\partial x_1} + \frac{1}{P_{22}}u_2\frac{\partial c_2}{\partial x_2} = \frac{1}{Pr_{21}}\frac{\partial^2 c_1}{\partial x_1^2} + \frac{1}{Pr_{21}}\frac{\partial^2 c_1}{\partial x_2^2} + \frac{1}{Pr_{22}}\frac{\partial^2 c_2}{\partial x_1^2} + \frac{1}{Pr_{22}}\frac{\partial^2 c_2}{\partial x_2^2}$$

$$\frac{\partial u_1}{\partial x_1} + \frac{\partial u_2}{\partial x_2} = 0$$

Here $Pr_{ii} = \nu/D_{ii}^*$ is the diffusion Prandtl number, $Ra_1 = g\beta_1 A_1 H^4/D_{22}^*\nu$, $Ra_2 = g\beta_2 A_2 H^4/D_{22}^*\nu$.

The initial conditions and the boundary conditions for the system of equations (12) are the following:

Initial conditions:

1) For the speed of components: $u_i(x_1, x_2, \tau = 0) = 0,\ i = 1,2$.

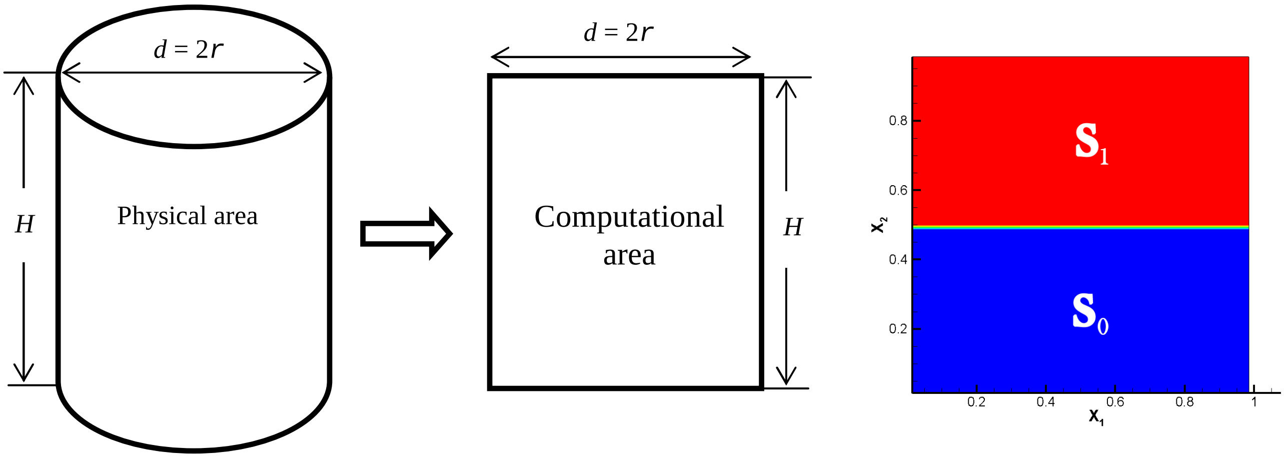
2) The process when the heavy and light gases are located in the upper part of the computational domain and the average gas is at the bottom part is studied. For the concentration in the upper computational area $S_1$ (Fig. 4b): $c_1|_{t=0,(x_1,x_2)\in S_1} = X_1$ is for the light component, $c_2|_{t=0,(x_1,x_2)\in S_1} = X_2$ is for the heavy component, $c_3|_{t=0,(x_1,x_2)\in S_1} = 0$ is for the middle component, where $X_1$, $X_2$ are the mole fractions of gas components.

3) For the concentration in the lower computational area $S_0$ (Fig. 4b): $c_1|_{t=0,(x_1,x_2)\in S_0} = 0$ is for the light component, $c_2|_{t=0,(x_1,x_2)\in S_0} = 0$ is for the heavy component, $c_3|_{t=0,(x_1,x_2)\in S_0} = 1$ is for the middle component.

Boundary conditions:

For the speed of components: $u_i = 0,\ i = 1,2$; for the concentration: $\frac{\partial c_i}{\partial n} = 0,\ i = 1,2$.

*Numerical algoritm*. As it is seen from the experimental data, mixing is carried out in the channel, which has as a rule a cylindrical shape. Heavy and light in density components of a binary mixture are in the upper part of the channel, and the gas with intermediate density in the lower part of the channel. To simplify the tasksit is considered 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. 4a). To register the isoconcentrational lines characterizing the emergence and development of convection, it is sufficient to consider the part of this area ($H/d$ >> 1), where the diffusion of components take place (Fig. 4b) ignoring the other areas of the two-dimensional area.



(a) (b)

Figure 4. Modelling of the studied process. a) display of areas; b) the initial conditions for the three-component mixture He + Ar – $N_2$.

For the numerical solution of the equations system (12) is used 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-Bachfort for convective terms and implicit scheme of Crank- Nicolson for the diffusion members defined by Kim and Moin [1985]

$$\frac{\overline{u}^{*}-\overline{u}^{n}}{\tau}=-\frac{1}{Pr_{22}}\overline{u}^{n}\nabla\overline{u}^{*}+\mathit{\Delta}\overline{u}^{*}+Ra_{1}c_{1}+Ra_{2}c_{2} \tag{13}$$

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

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

On the third stage, it is assumed that transference is done only due to 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 are calculated on the basis of five-point sweep method using the Adam-Bachfort scheme taking into account the found velocity fields

$$\frac{\overline{c_1}^{n+1}-\overline{c_1}^{n}}{\tau}=-\frac{1}{Pr_{22}}\left(\overline{u}^{n+1}\nabla\right)\overline{c_1}^{*}+\frac{1}{Pr_{11}}\Delta\overline{c_1}^{*}+\frac{1}{Pr_{12}}\Delta\overline{c_2}^{*} \tag{16}$$

$$\frac{\overline{c_2}^{n+1}-\overline{c_2}^{n}}{\tau}=-\frac{1}{Pr_{22}}\left(\overline{u}^{n+1}\nabla\right)\overline{c_2}^{*}+\frac{1}{Pr_{21}}\Delta\overline{c_1}^{*}+\frac{1}{Pr_{22}}\Delta\overline{c_2}^{*} \tag{17}$$

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

## THE RESULTS OF NUMERICAL MODELING

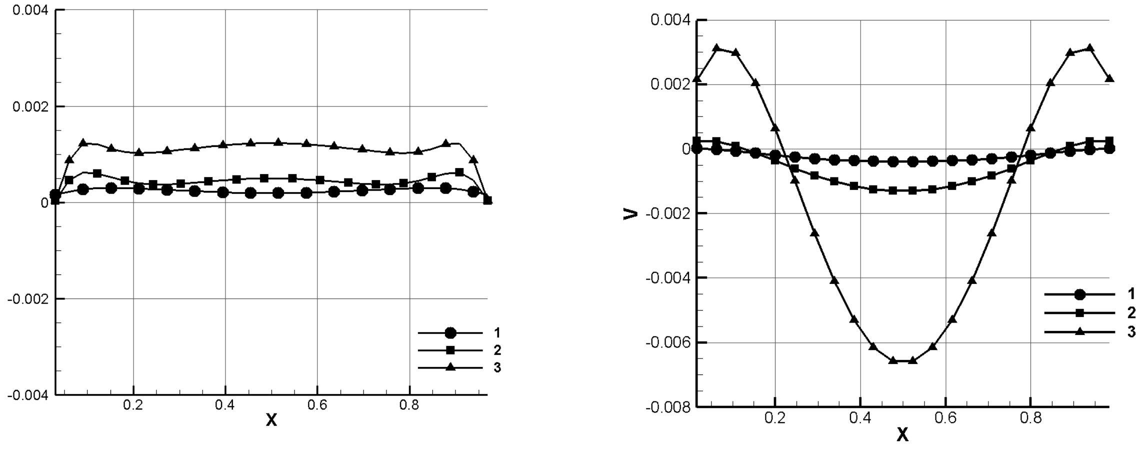
The calculations were carried out on a uniform rectangular staggered grid with the number of nodes 250 × 250 along the axes $x_1$, $x_2$, respectively. The time step was selected to be 0.001. The calculations are done for actual physical parameters of geometrical characteristics of the channel (Table 1). The main assumption in modeling is to limit the two-dimensional flows. At Fig. 5 is given the velocity profile of speed in section $y = 0.5$ 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.

Table 1
Some thermodynamic parameters of the mixing gases. $T$ = 298.1 K; $P$ = 0.101 MPa

| Compo-nents | ρ, (kg/m$^3$) | η, $10^{-5}$ (Pa s) | $D_{12}$, $10^{-4}$ (m$^2$/s) | $D_{13}$, $10^{-4}$ (m$^2$/s) | $D_{23}$, $10^{-4}$ (m$^2$/s) | Molar mass, $10^{-3}$ (kg/mole) |
|---|---|---|---|---|---|---|
| | | | $CH_4$ (1) + Ar (2) – $N_2$ (3) | | | |
| $CH_4$ | 0.644 | 1.105 | | | | 16.042 |
| Ar | 1.623 | 2.263 | 0.220 | 0.210 | 0.204 | 39.944 |
| $N_2$ | 1.138 | 1.779 | | | | 28.016 |
| | | | He (1) + Ar (2) – $N_2$ (3) | | | |
| He | 0.160 | 1.950 | | | | 4.003 |
| Ar | 1.623 | 2.263 | 0.743 | 0.700 | 0.203 | 39.944 |
| $N_2$ | 1.138 | 1.779 | | | | 28.016 |

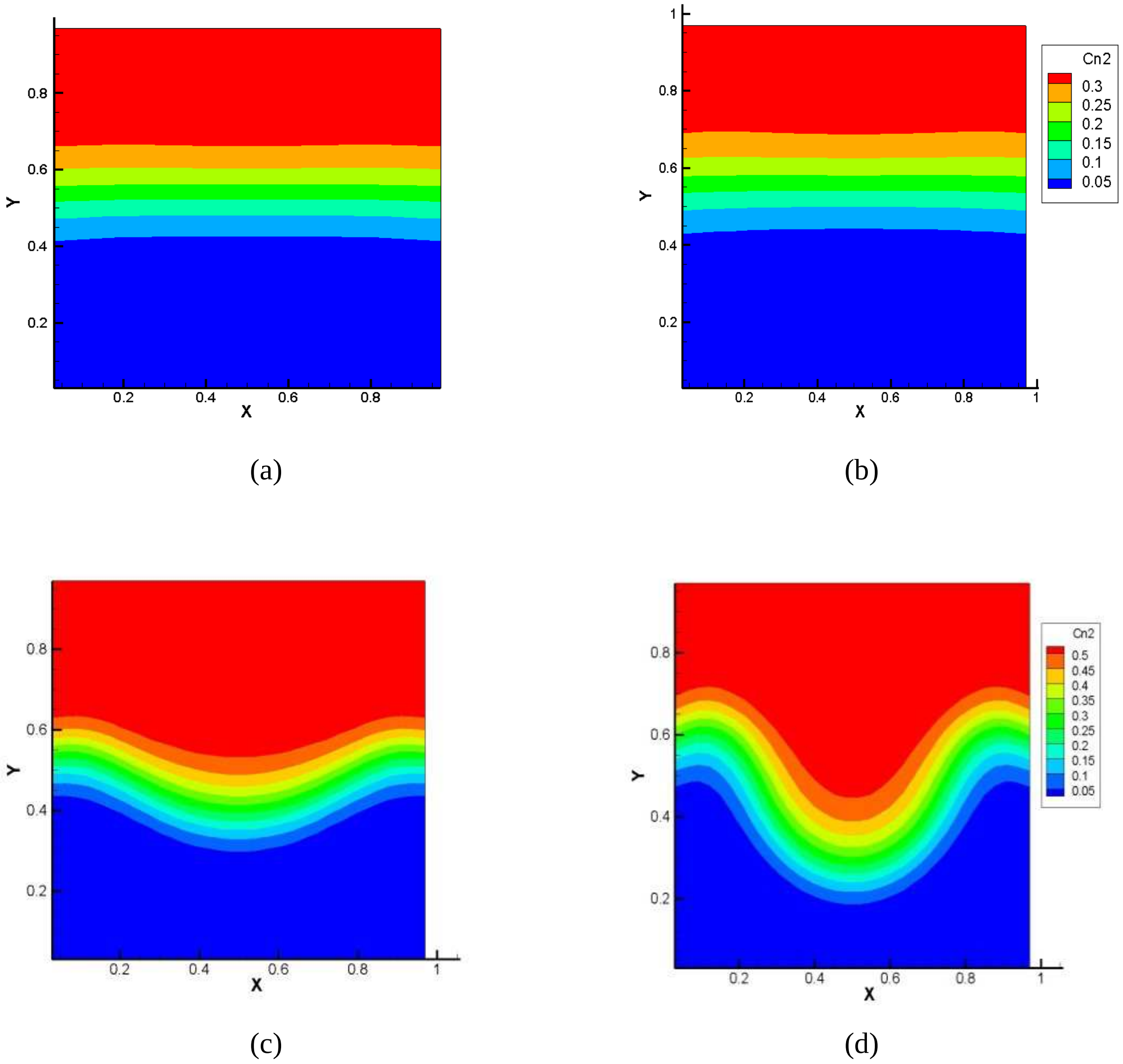
As the experimental data shows at Fig. 2a the diffusion takes place in the system. For the systems 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3) where $D_{ij}$ significantly differ from each other with increasing pressure becomes nonlinear.

The velocity profile at pressure 2.0 MPa significantly nonlinear and shows that in the system might take place an outgoing and rising convective flows, which is in agreement with the experimental data shown in Fig. 5b. In Fig. 6 the concentration lines characterize the mass transfer at different times. In the system 0.65 $CH_4$ (1) + 0.35 Ar (2) – $N_2$ (3) the mass transfer occurs by diffusion (Fig. 6a, b). At various isoconcentration bending lines is virtually absent. For the system 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3) is observed quite another picture. Prior to pressure $P$ ~ 1.0 MPa the picture of isoconcentration lines is similar to those shown in Fig. 6 a, b. The diffusive transfer is carried out in the system. Further pressure increase brings instability in the system. Isoconcentrational lines curved (Fig. 6, d) and takes place the transfer from the diffusion mode to convective. These results coincide with the experimental data shown in Fig. 2b. Thus, the main feature of the change of the mode "diffusion - convection" is the manifestation of the curvature of isoconcentrational lines.



(a) (b)

Figure 5. Vertical velocity profiles in the middle of computational area at time $t$ = 2 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; 2) $p$ = 1.5 MPa; 3) $p$ = 3.0 MPa. b) 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3), $T$ = 298.0 K, 1) $p$ = 0.5 MPa; 2) $p$ = 1.5 MPa; 3) $p$ = 2.0 MPa.



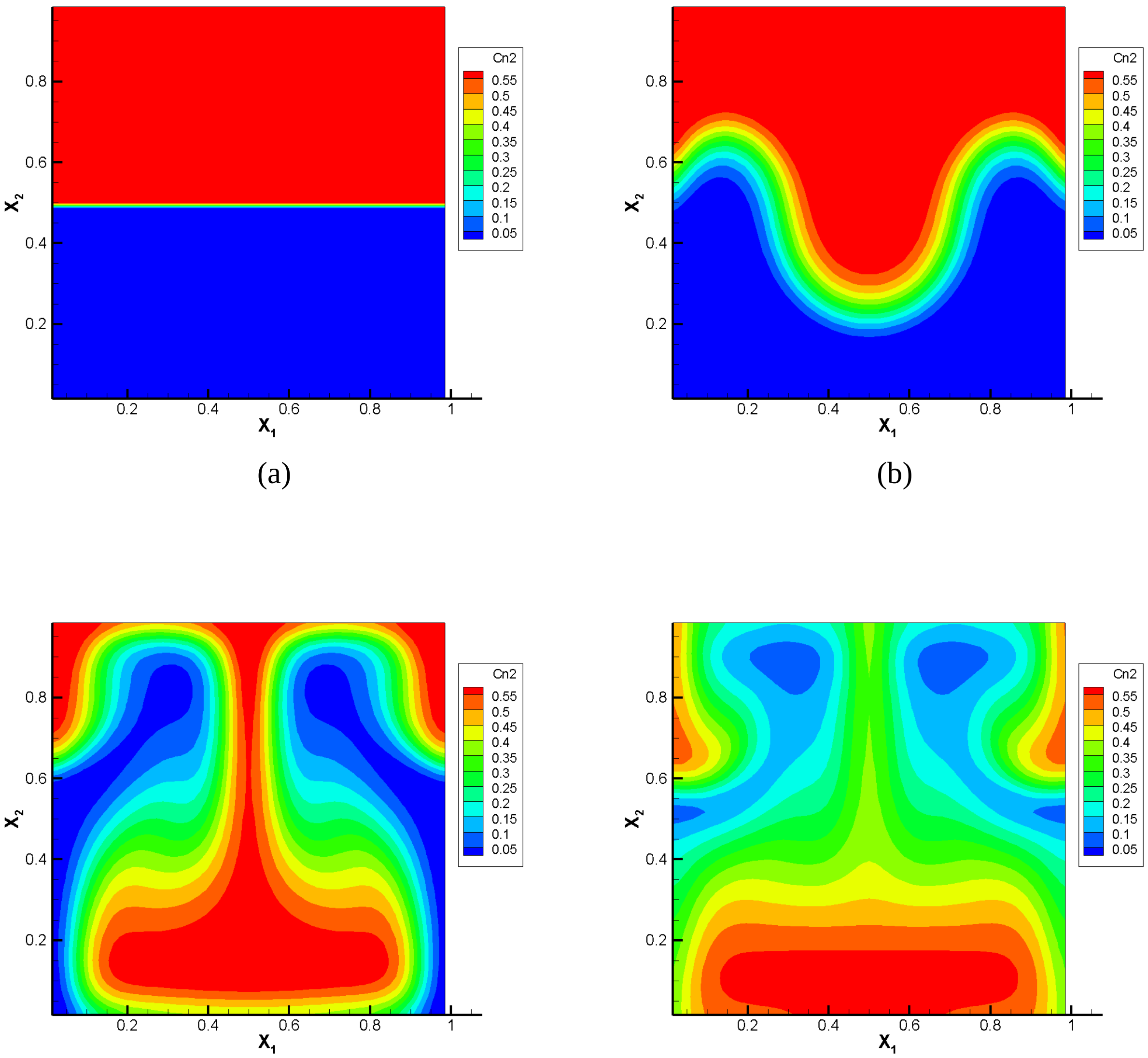
(a) (b)

(c) (d)

Figure 6. 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, $t$ = 13.5 s; b) $P$ = 2.0 MPa, $t$ = 18s; and for the system 0.4 He (1) + 0.6 Ar (2) – $N_2$ (3): c) $P$ = 1.5 MPa, $t$ = 13.5 s; d) $P$ = 2.0 MPa, $t$ = 18 s.

Fig. 7 shows the fields of concentrations at pressure $P$ = 2.5 MPa and various times. Calculation results show that at the starting stage the flow occurs with small speeds.

At the result of intensification of currents there observed a significant curvature of isoconcentrational lines and increase of speed transfer. Loss of time of mechanical stability of mixture for the given system forms tens of seconds, which is comparable with the experimental data shown in Fig. 3b. Then there observed the rapid development of convective currents. Convective flow is realized in the form large scaled structures with their subsequent annihilation. Further, takes place a similar revival of structured convection currents, which leads to the pulsation of medium on the surface of the flow. Periodicaly ("drip") blending mode is clearly fixed on the experimental data shown in Fig. 3. At the final stage of mixing the convection attenuated and multicomponent transfer is carried out in a diffusion mode.



(a) (b)

(c) (d)

Figure 7. Concentration fields for the argon at different times of mixing in the system 0.4 He (1) + 0.6 Ar (2) - N2 (3), *P* = 2.5 MPa, *T* = 298.0: a) *t* = 0 s; b) *t* = 10.55 s; c) *t* = 20.30 s; d) *t* = 30.05 s.

Thus the comparison results show that the used model and the method of calculation allows 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.

## CONCLUSIONS

It is experimentally studied the stability of mechanical equilibrium in isothermal gas mixtures. At a certain value of pressure the mechanical equilibrium in a mixture is disturbed and the system undergoes a transition "isothermal diffusion - convection gravity concentration". There have been offered mathematical model for numerical simulation of complex mass transfer in isothermal ternary gas mixtures at different pressures. Numerically investigated the effect of pressure systems, where the occurrence of convection is due to the instability of the mechanical mixture equilibrium.

Obtained pressure values associated with the transition of "diffusion - convection." The results of calculations to determine the diffusion areas and the gravity concentration convection in a ternary gas mixture, depending on the pressure is in satisfactory agreement with the experimental data.

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