Turbulence, Heat and Mass Transfer 8

Numerical modelling of convective diffusion in three-component gas mixtures

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Abstract – This paper considers the numerical modeling of convective diffusion in three-component gas mixtures. The experimental results on the study of unstable diffusion process at different pressures is carried out based on the numerical solution of unsteady filtered Navier-Stokes equation, the continuity equation and equation for the concentration. Comparison of the results of numerical simulations with experimental data for the system 0.4688 He + 0.5312 Ar - N2 showed that the intensity of convective mass transfer increases at a certain critical value of pressure.

1. Introduction

The analysis of the occurrence of instability in isothermal mutual diffusion showed that the description is completely analogous to the conventional thermal convection, when the system has only one constant thermodynamic force causing convection (diffusion performs role of the thermal conductivity). In the case of the existence of two forces ∇C and ∇T at the same time we will get qualitatively new effects [1], that the convective unstable states are possible in negative direction of the density gradient (at the bottom of the mixture more dense). Thus, the emergence of concentration convection in the gas mixture contained in the gravity field is possible in the case of inhomogeneous distribution of composition (density) in the channel of a diffusion cell. Based on experimental studies the occurrence of diffusion instability in three-component gas mixtures subject to a number of necessary conditions is shown in [2]:

1) Binary gas mixture (1 + 2) is located at the top, pure gas (3) - at the bottom, $\rho_2 > \rho_3 > \rho_1$; $\rho_{(1+2)} < \rho_3$; $D_{13} > D_{23}$;

2) Binary gas mixture (1+2) is located at the bottom, pure gas (3) – at the top, $\rho_2 > \rho_3 > \rho_1$; $\rho_{(1+2)} > \rho_3$; $D_{13} > D_{23}$;

3) Binary gas mixture (1 + 2) is located at the top, binary gas mixture (3+2) – at the bottom, $\rho_2 > \rho_3 > \rho_1$; $\rho_{(3+2)} > \rho_{(1+2)}$; $D_{12} > D_{32}$;

4) Binary gas mixture (1+2) and pure gas (3) may be disposed either above or below, $\rho_2 > \rho_3 > \rho_1$; $\rho_{(1+2)} = \rho_3$; $D_{13} > D_{23}$; in this version the unstable process is possible at any orientation mixtures, but only for different parameters, particularly pressure.

Necessary conditions for the onset of convection in the case of diffusive mixing should be complemented by the following sufficient conditions:

1) The gas mixture must consist of components that have diffusion coefficients, which differ several times (e.g., D_{He-Ar} is bigger over about three times than D_{Ar-N_2});

2) The instability occurs at certain concentration ranges of components;

3) The pressure effect is significant;

4) A diameter of the diffusion channel should not be less than a certain size;

5) The temperature influences on the occurrence of instability;

6) In some three - component gas mixtures the instability occurs independently from the initial orientation of the components in the diffusion apparatus;

7) The occurrence of the unstable process is more possible with decreasing diffusing mixture viscosity.

The effects caused by the presence of two thermodynamic forces can significantly become complicated by the presence of cross-effects. In this case, we have two reasons for occurrence of thermal convection concentration: heterogeneity of both the temperature and the concentration. A phenomenon, which leads to the loss of stability in such systems, has been called "double-diffusive convection" [3].

Since the isothermal diffusion in the triple mixtures is also characterized by the presence of two independent partial concentration gradients, then it seems urgent to analysis of the most characteristic moments arising in the study of phenomena the convection of class "double-diffusive" or convective diffusion.

The physical meaning of the paradox of instability (convective diffusion) in the three-component gas mixtures can be represented in the following way. The element of medium, which shifted randomly upward, goes into the mixture with a lower density, according to another composition. Due to differences in the coefficients of mutual diffusion of components the transverse diffusion in the first instance tends to equalize the concentration of the light component, its insufficiency is quickly compensated, and the element, which has been displaced, becomes lighter of environment, continues float up, creating instability. A similar situation for non-isothermal case in a binary liquid mixture has been described by several authors [1, 3, 4].

Predict the region of thermodynamic parameters of mechanical equilibrium instability with diffusion can be within the stability theory [4 - 7]. It should be noted that in the study of non-stationary processes, this approach may not accurately determine the critical conditions of "diffusion-convection" transition process. Studying the dynamics of convective flows and evaluation of multicomponent mass transfer kinetics in the developed instability is not possible within the framework of the instability theory. Such problem can be solved by numerical methods of mathematical modelling. The intermediate velocity field is obtained by using fractional step method in combination with TDMA algorithm. At the second step the Poisson equation for pressure is solved using the obtained intermediate velocity field. At the third step is assumed that the transfer of mass is carried out only by the pressure gradient. At each stage of the fractional step method is used the TDMA algorithm to find landmark values of the intermediate values of the velocity field. At the last stage of splitting scheme, the finite field of velocity adjusted for pressure is obtained [8 - 10].

This scheme was approved for the system $0.4722\,He + 0.5278\,Ar - N_2$. For this system the velocity profiles were obtained at different concentrations and experiences values of pressure that remain flat at the pressure below the critical value, at which the transition occurs "diffusion - convection", but at the critical pressure and concentrations velocity profiles of the components the front is not flat , which indicates the presence of upstream and downstream flow.

The purpose of this paper is to simulate numerically the formation mechanism of convective flows in the diffusion of three-component gas mixtures.

2. Numerical model

Numerical simulation of the problem is based on the solution of unsteady Navier-Stokes equation with the continuity and concentration equations (1):

$$\begin{cases} \dfrac{\partial \vec{u}_i}{\partial t} = -\nabla p + \nabla^2 \vec{u} + (R_1 c_1 \tau_{11} + R_2 c_2)\vec{\gamma}, \\ \dfrac{\partial c_1}{\partial t} + \vec{v}\nabla c_1 = \dfrac{1}{\mathrm{Pr}_{11}} \nabla^2 c_1 + \dfrac{A_2}{A_1}\dfrac{1}{\mathrm{Pr}_{12}} \nabla^2 c_2, \\ \dfrac{\partial c_2}{\partial t} + \vec{v}\nabla c_2 = \dfrac{A_1}{A_2}\dfrac{1}{\mathrm{Pr}_{21}} \nabla^2 c_1 + \dfrac{1}{\mathrm{Pr}_{22}} \nabla^2 c_2, \\ div\, \vec{v} = 0. \end{cases} \tag{1}$$

where D^*_{ij} - practical diffusion coefficient, $\mathrm{Pr}_{ii} = \nu / D^*_{ii}$ - diffusion Prandtl number, $R_i = g\beta_i A_i d^4 / \nu D^*_{ii}$ - partial Rayleigh number, $\tau_{ij} = D^*_{ij} / D^*_{22}$ - parameter of the relationship between practical diffusion coefficients.

Boundary conditions consist of the following boundary conditions (2):

$$\vec{u} = 0, \ \frac{\partial c_i}{\partial n} = 0, \tag{2}$$

where n - normal to the boundary.

3. Numerical algorithm

Splitting scheme by physical parameters is used for the numerical solution of the gas motion in the cylindrical region. It is proposed the following physical interpretation of the given splitting scheme.

$$\begin{aligned}
&1. \quad \frac{\overline{u}^{*} - \overline{u}^{n}}{\tau} = -\overline{u}^{n}\nabla\overline{u}^{*} + \Delta\overline{u}^{*} + \tau_{11} Ra_1 C_1 + Ra_2 C_2, \\
&2. \quad \Delta p = \frac{\nabla\overline{u}^{*}}{\tau}, \\
&3. \quad \frac{\overline{u}^{n+1} - \overline{u}^{*}}{\tau} = -\nabla p \\
&4. \quad \frac{\overline{C}_1^{n+1} - \overline{C}_1^{n}}{\tau} = -\left(\overline{u}^{n+1}\nabla\right)\overline{C}_1^{*} + \frac{1}{\mathrm{Pr}_{11}}\Delta\overline{C}_1^{*} + \frac{1}{\mathrm{Pr}_{12}}\Delta\overline{C}_2^{*} \\
&5. \quad \frac{\overline{C}_2^{n+1} - \overline{C}_2^{n}}{\tau} = -\left(\overline{u}^{n+1}\nabla\right)\overline{C}_2^{*} + \frac{1}{\mathrm{Pr}_{21}}\Delta\overline{C}_1^{*} + \frac{1}{\mathrm{Pr}_{22}}\Delta\overline{C}_2^{*}
\end{aligned} \tag{3}$$

At the first stage of the numerical splitting scheme, the transfer of momentum carried out only by convection and diffusion. Compact scheme of high order accuracy is used to approximate the convective and diffusive terms of the equation. The intermediate velocity field is calculated by the method of fractional steps, using the sweep method.

At the second stage, the pressure is calculated by using already founded intermediate velocity field.

At the third step, we assume that the transfer is carried out only by the pressure gradient. At the final step, the concentration is calculated by using finite velocity fields.

3.1 Calculation of the intermediate velocity field

The intermediate velocity field calculated by using the method of fractional steps. Sweep method for finding values of the intermediate velocity field used at the each step of the method of fractional steps.

Consider the method of fractional steps for the horizontal component of the velocity u_1 in the grid point $(i+1/2, j)$.

$$\frac{\partial \overline{u_1}}{\partial \tau} + \bar{u}_1 \frac{\partial \overline{u_1}}{\partial x_1} + \overline{u_2} \frac{\partial \overline{u_1}}{\partial x_2} = \frac{\partial^2 \overline{u_1}}{\partial x_1^2} + \frac{\partial^2 \overline{u_1}}{\partial x_2^2} \tag{4}$$

At the first stage velocity u_1 is looked for in the direction of coordinates x_1:

$$\frac{\bar{u}_{1\,i+\frac{1}{2},j}^{\,n+\frac{1}{2}} - \bar{u}_{1\,i+\frac{1}{2},j}^{\,n}}{\tau} = \frac{1}{2}\left[\Lambda_1 \bar{u}_{1\,i+\frac{1}{2},j}^{\,n+\frac{1}{2}} + \Lambda_2 \bar{u}_{1\,i+\frac{1}{2},j}^{\,n}\right] \tag{5}$$

where

$$\Lambda_1 \bar{u}_{1\,i+\frac{1}{2},j} = -u_1 \left.\frac{\partial \overline{u_1}}{\partial x_1}\right|_{i+\frac{1}{2},j} + \left.\frac{\partial}{\partial x_1}\left(\frac{\partial \overline{u_1}}{\partial x_1}\right)\right|_{i+\frac{1}{2},j},$$

$$\Lambda_2 \bar{u}_{1\,i+\frac{1}{2},j} = -u_2 \left.\frac{\partial \overline{u_1}}{\partial x_2}\right|_{i+\frac{1}{2},j} + \left.\frac{\partial}{\partial x_2}\left(\frac{\partial \overline{u_1}}{\partial x_2}\right)\right|_{i+\frac{1}{2},j}.$$

This equation is solved by the sweep method and for result we get $\bar{u}_{1\,i+\frac{1}{2},j}^{\,n+\frac{1}{2}}$.

$$a_i \bar{u}_{1\,i+1,j}^{\,n+\frac{1}{2}} - b_i \bar{u}_{1\,i,j}^{\,n+\frac{1}{2}} + c_i \bar{u}_{1\,i-1,j}^{\,n+\frac{1}{2}} = -d_i, \tag{6}$$

where

$$a_i = \frac{1}{2}\left[-\bar{u}_{1i,j}^{\,n} \frac{1}{2\Delta x_1} + \frac{1}{\Delta x_1^2} \right],$$

$$b_i = \frac{1}{\tau} + \frac{1}{\Delta x_1^2},$$

$$c_i = \frac{1}{2}\left[\bar{u}_{1i,j}^{\,n} \frac{1}{2\Delta x_1} + \frac{1}{\Delta x_1^2} \right],$$

$$d_i = \frac{\bar{u}_{1i,j}^{\,n}}{\tau} + \frac{1}{2}\Lambda_2 \bar{u}_{1\,i+\frac{1}{2},j}^{\,n}.$$

At the second stage velocity u_1 is looked for in the direction of coordinates x_2:

$$\frac{\bar{u}_{1\,i+\frac{1}{2},j}^{\,n+1} - \bar{u}_{1\,i+\frac{1}{2},j}^{\,n+\frac{1}{2}}}{\tau} = \frac{1}{2}\left[\Lambda_1 \bar{u}_{1\,i+\frac{1}{2},j}^{\,n+\frac{1}{2}} + \Lambda_2 \bar{u}_{1\,i+\frac{1}{2},j}^{\,n+1} \right]. \tag{7}$$

This equation is solved by the sweep method and for result we get $\bar{u}_{1\,i+\frac{1}{2},j}^{\,n+1}$.

$$a_j \bar{u}_{1i,j+1}^{\,n+1} - b_j \bar{u}_{1i,j}^{\,n+1} + c_j \bar{u}_{1i,j-1}^{\,n+1} = -d_j. \tag{8}$$

where

$$a_j = \frac{1}{2}\left[-\bar{u}_{2i,j}^{\,n} \frac{1}{2\Delta x_2} + \frac{1}{\Delta x_2^2} \right],$$

$$b_j = \frac{1}{\tau} + \frac{1}{\Delta x_2^2},$$

$$c_j = \frac{1}{2}\left[\bar{u}_{2i,j}^{\,n} \frac{1}{2\Delta x_2} + \frac{1}{\Delta x_2^2} \right],$$

$$d_j = \frac{\bar{u}_{1i,j}^{\,n+\frac{1}{2}}}{\tau} + \frac{1}{2}\Lambda_1 \bar{u}_{1\,i+\frac{1}{2},j}^{\,n+\frac{1}{2}}.$$

Further, in a similar manner the horizontal velocity components at $(i, j + 1/2)$ point of the grid are obtained.

At the second stage, the poisson's equation, obtained from the continuity equation in view of the velocity field of the first stage.

At the third stage, the resulting pressure field is used to recalculate the final velocity field.

At the final stage, the obtained velocity field is used for solving the component concentrations equations.

3.2 Calculation of the concentration

Consider the method of fractional steps for the concentration $\overline{C_1}$ in the grid point $(i + 1/2, j)$.

$$\frac{\partial \overline{C_1}}{\partial \tau} + \bar{u}_1 \frac{\partial \overline{C_1}}{\partial x_1} + \bar{u}_2 \frac{\partial \overline{C_1}}{\partial x_2} = \frac{1}{\mathrm{Pr}_{11}} \left(\frac{\partial^2 \overline{C_1}}{\partial x_1^2} + \frac{\partial^2 \overline{C_1}}{\partial x_2^2} \right) + \frac{1}{\mathrm{Pr}_{12}} \left(\frac{\partial^2 \overline{C_2}}{\partial x_1^2} + \frac{\partial^2 \overline{C_2}}{\partial x_2^2} \right). \quad (9)$$

In the first stage concentration is looked for in the direction of coordinates x_1:

$$\frac{\overline{C_1}_{i+\frac{1}{2},j}^{n+\frac{1}{2}} - \overline{C_1}_{i+\frac{1}{2},j}^{n}}{\tau} = \frac{1}{2} \left[\Lambda_1 \overline{C_1}_{i+\frac{1}{2},j}^{n+\frac{1}{2}} + \Lambda_2 \overline{C_1}_{i+\frac{1}{2},j}^{n} \right]. \quad (10)$$

where

$$\Lambda_1 \overline{C_1}_{i+\frac{1}{2},j} = -\bar{u}_1 \frac{\partial \overline{C_1}}{\partial x_1}\Bigg|_{i+\frac{1}{2},j} + \frac{1}{\mathrm{Pr}_{11}} \frac{\partial}{\partial x_1} \left(\frac{\partial \overline{C_1}}{\partial x_1} \right)\Bigg|_{i+\frac{1}{2},j},$$

$$\Lambda_2 \overline{C_1}_{i+\frac{1}{2},j} = -u_2 \frac{\partial \overline{C_1}}{\partial x_2}\Bigg|_{i+\frac{1}{2},j} + \frac{1}{\mathrm{Pr}_{11}} \frac{\partial}{\partial x_2} \left(\frac{\partial \overline{C_1}}{\partial x_2} \right)\Bigg|_{i+\frac{1}{2},j}$$

This equation is solved by the sweep method and for result we get $\overline{C_1}_{i+\frac{1}{2},j}^{n+\frac{1}{2}}$.

$$a_i \overline{C_1}_{i+1,j}^{n+\frac{1}{2}} - b_i \overline{C_1}_{i,j}^{n+\frac{1}{2}} + c_i \overline{C_1}_{i-1,j}^{n+\frac{1}{2}} = -d_i . \quad (11)$$

where

$$a_i = \frac{1}{2} \left[-\bar{u}_{1i,j}^{n} \frac{1}{2\Delta x_1} + \frac{1}{\mathrm{Pr}_{11}} \frac{1}{\Delta x_1^2} \right],$$

$$b_i = \frac{1}{\tau} + \frac{1}{\mathrm{Pr}_{11}} \frac{1}{\Delta x_1^2},$$

$$c_i = \frac{1}{2} \left[\bar{u}_{1i,j}^{n} \frac{1}{2\Delta x_1} + \frac{1}{\mathrm{Pr}_{11}} \frac{1}{\Delta x_1^2} \right],$$

$$d_i = \frac{\overline{C}^{\,n}_{1\,i,j}}{\tau} + \frac{1}{2}\Lambda_2 \overline{C}^{\,n}_{1\,i+\frac{1}{2},j}$$

In the second stage concentration is looked for in the direction of coordinates x_2:

$$\frac{\overline{C}^{\,n+1}_{1\,i+\frac{1}{2},j} - \overline{C}^{\,n+\frac{1}{2}}_{1\,i+\frac{1}{2},j}}{\tau} = \frac{1}{2}\left[\Lambda_1 \overline{C}^{\,n+\frac{1}{2}}_{1\,i+\frac{1}{2},j} + \Lambda_2 \overline{C}^{\,n+1}_{1\,i+\frac{1}{2},j}\right]. \qquad (12)$$

This equation is solved by the sweep method and for result we get $\overline{C}^{\,n+1}_{1\,i+\frac{1}{2},j}$.

$$a_j \overline{C}^{\,n+1}_{1\,i,j+1} - b_j \overline{C}^{\,n+1}_{1\,i,j} + c_j \overline{C}^{\,n+1}_{1\,i,j-1} = -d_j\,. \qquad (13)$$

where

$$a_j = \frac{1}{2}\left[-\overline{u}^{\,n}_{2i,j}\frac{1}{2\Delta x_2} + \frac{1}{\mathrm{Pr}_{11}}\frac{1}{\Delta x_2^2}\right],$$

$$b_j = \frac{1}{\tau} + \frac{1}{\mathrm{Pr}_{11}}\frac{1}{\Delta x_2^2},$$

$$c_j = \frac{1}{2}\left[\overline{u}^{\,n}_{2i,j}\frac{1}{2\Delta x_2} + \frac{1}{\mathrm{Pr}_{11}}\frac{1}{\Delta x_2^2}\right],$$

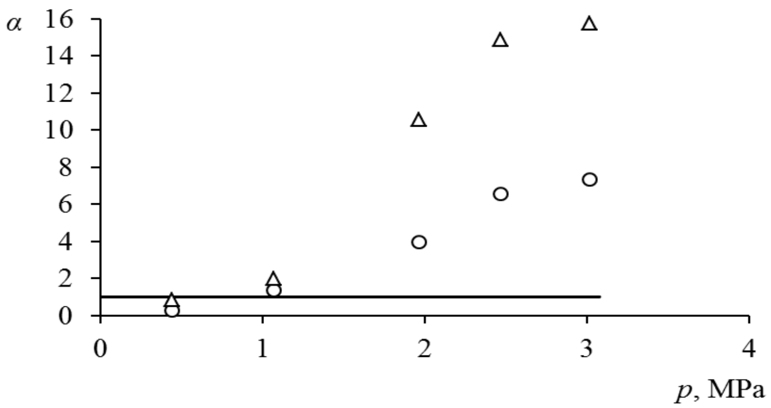
$$d_j = \frac{\overline{C}^{\,n+\frac{1}{2}}_{1\,i,j}}{\tau} + \frac{1}{2}\Lambda_1 \overline{C}^{\,n+\frac{1}{2}}_{1\,i+\frac{1}{2},j} + \frac{1}{\mathrm{Pr}_{12}}\Delta\overline{C}_2\,..$$

Further, in the similar fractional steps method at the grid point $(i+\tfrac{1}{2}, j)$ the the concentration were obtained.

4. Experimental results

The pressure is an important characteristic of mass transfer in multicomponent systems. The increase in pressure can lead to a breach of sustainable diffusion process and the onset of convection. The authors of "Instabilities in ternary diffusion" Miller L., Spurling T.H., Mason E.A. [8] are one of the firsts, who drew the attention to the effect of pressure to instability, and it is studied in more detail in [9-11].

The experimental system 0.4688 He + 0.5312 Ar - N_2 is chosen to study the effect of pressure. Fig. 1 presents data for the system to the stable and unstable transfer at T = 298.0 K and different values of pressure [10].



Solid line - calculation under the assumption of stable diffusion; ○ - helium; Δ - nitrogen.
Figure 1: The dependence of the parameter α on the pressure

As it is seen from the data shown in Fig. 1, at pressure fields up to 1.5 MPa the steady diffusive transport is observed, since the dimensionless criterion α = 1. The dimensionless criterion α for component *i* is determined from the ratio of the measured concentration to its theoretical value calculated under the assumption diffusion Stefan-Maxwell equations. With increasing pressure the unstable process is observed, characterized by a sharp increase in the parameter α, which indicates the occurrence of convection.

5. Numerical results

The numerical model allows to describe the occurrence of diffusion instability depending on the pressure. For this problem the pressure is selected in the range $P \approx 0.2 \div 3.0\, MPa$. The calculations used the mesh size 128x128x128. The time step is taken as $\Delta\tau = 0.001$.

In the calculations the diffusion instability is registered at *P = 1.5 MPa*. In figure 2 the "diffusion - convection gravity concentration" transition illustrate at the moment t = 2. As can be seen from the figure 3, the oscillation amplitude of the convection is increased, indicating a violation of the diffusive instability. The vertical velocity and concentration distributions at the mid-height (y=0.5) are presented in Figure 4 and show the occurrence of diffusion instability.

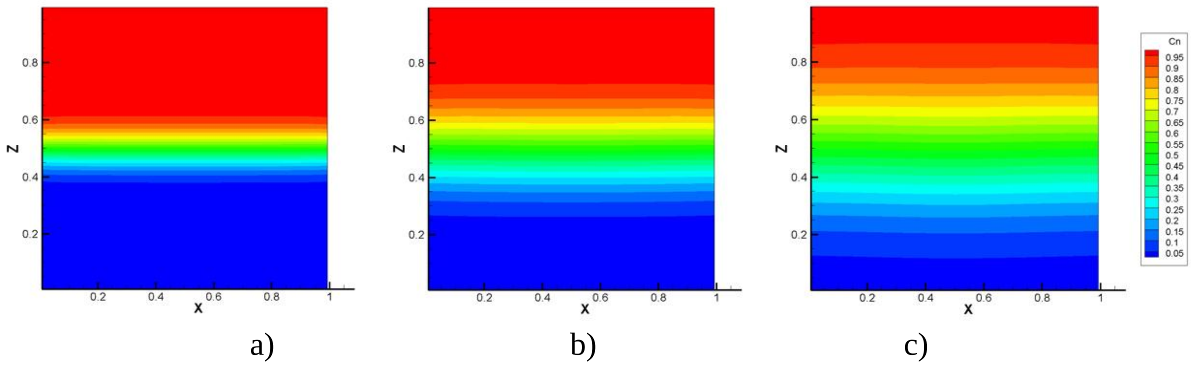
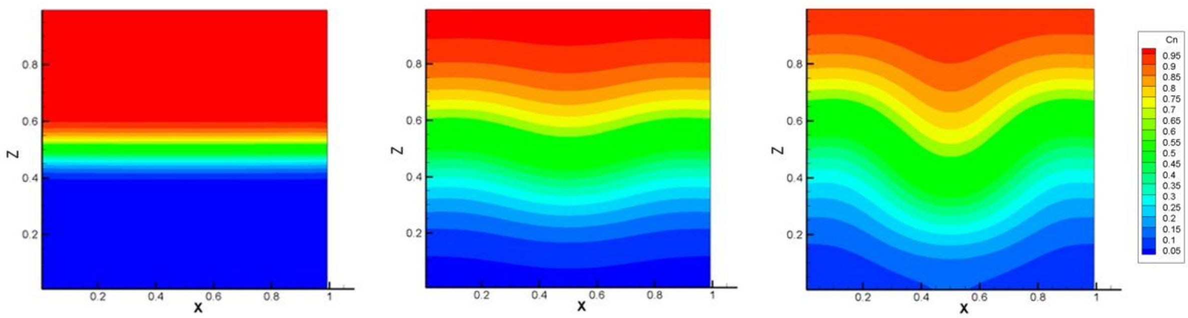


Figure 2: Change of concentration of 0.5312 Ar + 0.4688 $He - N_2$ gas system at P = 1.5 MPa: $a)\ t = 0.25;\ b)\ t = 1;\ c)\ t = 2.$



a) b) c)

Figure 3: Change of concentration of 0.5312 Ar + 0.4688 $He-N_2$ gas system at P = 2.0 MPa: $a)\ t=0.25;\ b)\ t=1;\ c)\ t=2.$

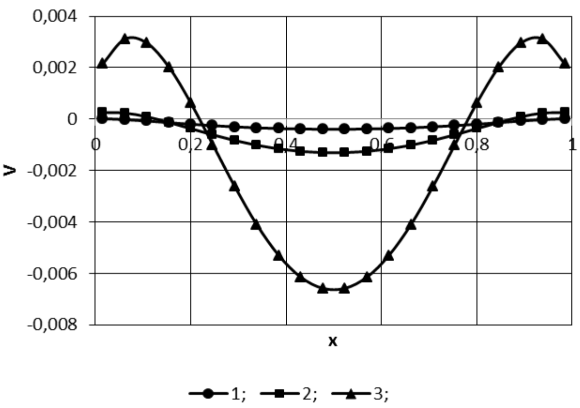


Figure 4: The velocity profile $V(x)$ in a section $y=0.5$ at $t=2$: 1) $P=0.5\ MPa$; 2) $P=1.5\ MPa$; 3) $P=2.0\ MPa$.

7. Conclusion

This studies show, that the pressure is one of the factors contributing to the emergence of unstable diffusion process in the gas system. The critical Rayleigh number is determined basing on the numerical solution of the gas motion in the cylindrical region and solving the unsteady Navier-Stokes equation and the continuity equation with the concentration equation, which determines the transition border from the steady diffusion to the region of the concentrating gravitational convection, and shows the change of velocity profiles and component concentrations. At the critical values of pressure the velocity profiles have not flat front, which indicates the presence of upstream and downstream. Investigation of the influence pressure on the occurrence of convective diffusion has shown, that increase in pressure leads to an increase in the intensity of convective flows, i.e., to process intensification.

Comparative analysis of numerical simulation based on the method of splitting into physical parameters with the experimental data for the system 0.4688 He + 0.5312 Ar – N_2 showed qualitative and quantitative agreement. Therefore, the approach described in this investigation can be used to determine the critical parameters under which a transition from stable to unstable diffusion process is observed, characterized by the occurrence of convective flows.

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