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Instability of mechanical equilibrium and some features of concentration convection in isothermal ternary gaseous systems

A. Zhussanbayeva , V.N. Kossov* , O.V. Fedorenko and M. Zhaneli

Abai Kazakh National Pedagogical University, Almaty, Kazakhstan,
*e-mail: kosov_vlad_nik@list.ru

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Empirical research of the formation of convection streams in a ternary gaseous compound He + R12 – Ar at various pressures and concentrations of the miscible components has been implemented. Experimental data obtained have demonstrated that the intensiveness of convection streams increases with increasing pressure. It is noted that in the system under consideration, the initiation of convection streams is possible when the content of Freon-12 in the mixture is more than 0.1 mole fraction. A numerical study of the unstableness of mechanical balance in the mixture He + R12 – Ar with significantly different diffusion coefficients of gas mixture components has been carried out by means of the splitting scheme in terms of physical parameters. The presented mathematical model makes it possible to describe the process of formation of a convective structure for various compositions of mixture. The onset of instability is accompanied by the formation of curved concentration profiles. Within the numerical framework, the onset times of various mixing regimes are determined.

Keywords: diffusion, convection, gas mixtures.
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1 Introduction

Convective motions in a single-component medium where density has monotonous temperature dependence are researched carefully and reported in [1-4]. Mentioned works present approaches, which allow for channels of various geometric shapes to investigate free gravitational convection, which makes it possible to predict mixing modes for specific conditions. For single-phase systems, the occurrence of thermoconcentration gravitational flows is the heterogeneity of the density of medium caused by temperature variations. The directions of the force of gravity and the density gradient of the medium have a significant effect on the formation of convective instability. In this case, the medium has a stable stratification if the gravity and density gradient vectors are codirectional. This situation does not contribute to the emergence of convective flows.

In binary non-isothermal mixtures, it is necessary to take into account diffusion, which leads to additional mechanisms for the occurrence and development of convective flows [5, 6]. One of the features of the research, which conducted in [5, 6] was the assumption, that heat and dissolved matter as components affecting the general medium density distribution. It follows from this that for multicomponent mixtures, convective separation effects will also appear, the reason of occurrence of which is distinction between the interdiffusion constants of the mixture constituents. Finding out of the influence of the constituents' diffusibility is possible when studying isothermal mixing. Experimental and numerical data of multicomponent gas mixtures [7-14] have shown that the unstableness of mechanical balance of the mixture may be realized under the condition of a decrease in density with height. This condition is not representative when studying the thermal convection problems described in [1-5] for single-component systems. For such situations, the component with the highest molar mass was transferred more intensively, which was noted by the authors of the work [15]. Thus, when solving problems of complex mass transfer, it is necessary to pay special attention to problems aimed at studying and clarifying the conditions under which it is possible to separate a component with certain properties during multicomponent mixing.

Purpose of the article is an empirical research of the unstableness of mechanical balance and the

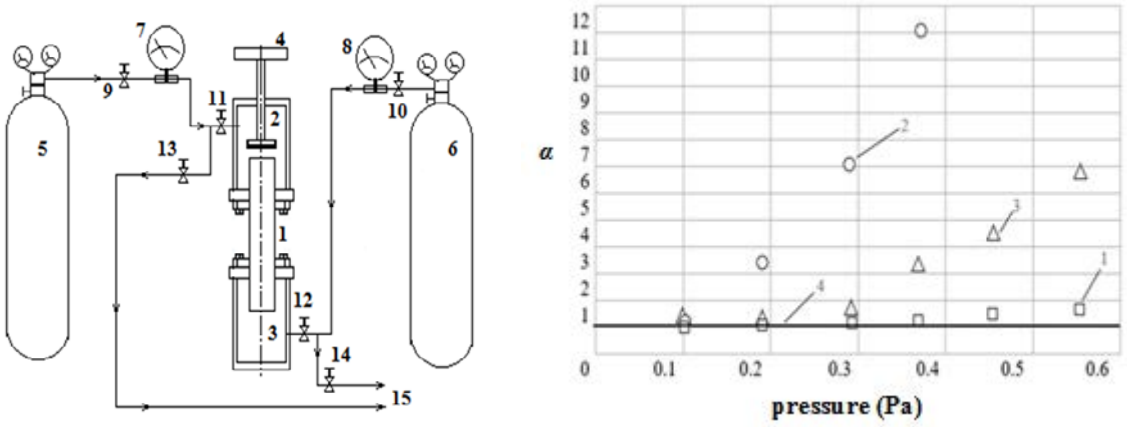
change of modes of "diffusion – concentration gravitational convection" in an isothermal three-component helium – Freon – argon gas mixture at various pressures and compositions for a case when the mixture density deflates with altitude change. The regime change "diffusion – concentration gravitational convection" border is defined. A comparison of the computation data with the empirical results is implemented.

2 Experiment

A ternary mixture of gases He (1) + R12 (2) – Ar (3) was subject to study at different contents of Freon-12 in the mixture. The order of the components in the mixture under study is indicated by the quantity in parentheses. The composition of the initial mixtures was selected in such a way that the investigated mixture was hydrostatically stable when the immixture process started.

The divided transport of constituents was carried out in an experimental device that implements the two-flask method. The diffusion cell consists of two flasks (an upper one with volume V_u and a lower one with volume V_l) connected by a vertical cylindrical channel (see Fig. 1a). The geometric characteristics of a diffusion cell with a cylindrical channel have the following values: V_l = (214.5 ± 0.5)×10^{-6} m^3, V_u = (226.8 ± 0.5)×10^{-6} m^3, L = (165.00 ± 0.05)×10^{-3} m. The studies were fulfilled at a constant temperature equal to 298.0 K and pressures within (0.15 – 0.60) MPa. The pressure and the temperature measurement accuracy was 0.02 MPa and 0.1 K respectively. Measurement of the concentration of components was implemented by gas chromatography. The error in measuring the concentration did not exceed 1–3%.

Operation method on the experimental bench was described in detail for similar devices in [16], so we will dwell on the main procedures that allow us to obtain experimental data for the mixture under study. The upper flask 2 through the system of valves 9, 11 and the pressure gauge 7 is filled with the initial mixture from the cylinder 5 to a predetermined pressure. A similar procedure for filling gas the lower flask 3 of the apparatus from the cylinder 6 with is carried out through the manometer 8 and the system of valves 10, 12. After fixing the experiment pressure, the valves 9, 11 and 10, 12 are blocked. The mixing of gases through the connecting channel of a given geometry 1 occurs when the valve 4 is opened with simultaneous registration of time. When the experiment is finished, the valve 4 is closed and the mixing time is fixed. Then the studied mixtures are fed to the outlets 15, and then transported to the chromatograph for analysis.



a) (b)

Figure 1 – Experimental studies of the unstableness of mechanical balance in triple gas mixtures: (a) Experimental setup [17]; (b) Pressure dependence of the factor α for the system 0.7 He (1) + 0.3 R12 (2) – Ar (3), T = 298.0 K: 1 – helium, 2 – Freon-12, 3 – argon, 4 – calculation assuming diffusion

During experiments, it was supposed to study the mixing of a two-component system arranged in the top flask 2 with a pure constituent located in the bottom flask 3 (see Fig. 1a). To exclude convective mechanisms caused by the unstable density stratification of the system [1-5], the lighter (in terms of density) binary mixture was located above the heavy gas. Thus, the condition of the negative direction of the density gradient was initially formed, under which the implementation of the diffusion type of mixing without convective disturbances is possible. The placement of the mixture components relative to the diffusion channel associated with the opposite direction of the change in density per unit length was not considered. The duration of the experiment at predetermined temperature and pressure was 60 minutes. Such mixing times provided sufficient values for the diffused concentrations of the components, which made it possible to carry out chromatographic analysis.

The experimental concentration values c_{exp} were normalized to the theoretical values c_{theor} calculated in the diffusion approximation using the Stefan-Maxwell equations [18]. Figure 1b indicates a typical pressure relationship of the nondimensional factor $\alpha = c_{exp} / c_{theor}$ for the analyzed system. An analysis of the data presented in this figure shows that in the pressure range up to 0.15 MPa, diffusion occurs in the system. It is confirmed by the fact that all constituents of the considered system have the value of the nondimensional factor α_i equal to one. For each component of the mixture, the parameter α_i starts to raise when an experimental pressure equals to 0.15 MPa. A particularly significant increase was noted for Freon-12 and, to a lesser extent, for the most diffusively mobile He. Thus, the transition from diffusion to convective mixing is possible when the pressure in the experiment is equal to or exceeds 0.15 MPa. The growth of the rate of convective flows formation is associated with a subsequent raise in pressure. The type of mixing in which the intensity of mixing decreases with increasing pressure is not representative when studying diffusion process. Distributions similar in form were also observed for other primary combinations of the systems. If the Freon-12 content in the initial mixture is less than 0.1 mole fraction, diffusion occurs in the system. The α_i parameters for each component will be approximately equal to one.

3 Numerical analysis

Let us consider the process of mixing in a cylindrical channel, in which the main diffusing components are diluted by the third component. Figure 2 explains the problem statement. A mixture of light gas 1 (with molar mass M_1) and heavy gas 2 (with molar mass M_2) contains in the upper part of channel S_1. This mixture diffuses into a gas with an intermediate molar mass M_3, which arranges in the section S_2 (where S_2 is the bottom region of channel).

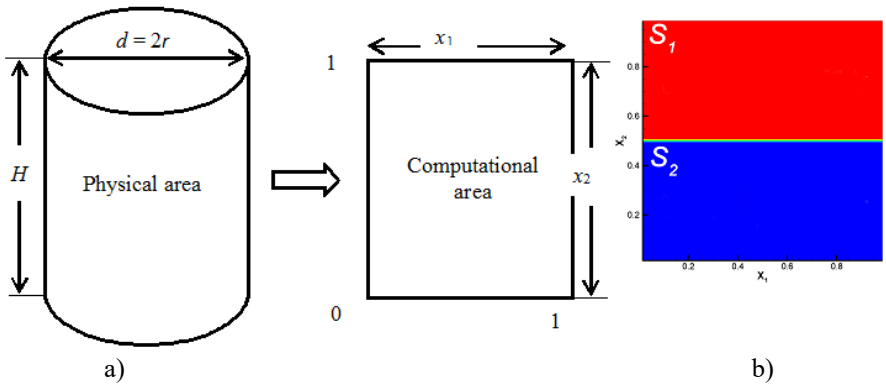


Figure 2 – Model of multicomponent diffusion: (a) Natural and numerical regions of immixture; (b) Placement of three-component system at the starting terms

The use of the Boussinesq approximation for a system of fluid dynamics equation written for disturbed quantities makes it possible to determine the conditions for three-component gas mixtures under which the transition from diffusion to convection is possible. The mentioned system of

equations is composed of the motion equation in the form of the Navier-Stokes equation, the equations of convective diffusion and the conservation equation of particles number in the mixture. When writing the considered system of equations, it is necessary to take into account the conditions of independent diffusion, which for a three-component system under the condition of constant temperature can be represented as follows $\sum_{i=1}^{3}\vec{j}_i = 0$ and $\sum_{i=1}^{3} c_i = 1$. Then the considered system of equations has the form [19]:

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

$$\frac{\partial c_1}{\partial t}+\vec{v}\nabla c_1=\mathrm{div}\left[D_{11}^{*}\nabla c_1+D_{12}^{*}\nabla c_2\right], \\ \frac{\partial c_2}{\partial t}+\vec{v}\nabla c_2=\mathrm{div}\left[D_{21}^{*}\nabla c_1+D_{22}^{*}\nabla c_2\right], \quad (1)$$

$$\frac{\partial n}{\partial t}=-\mathrm{div}\left(n\vec{v}\right),$$

where c_i is the concentration of the i-th component; D_{ij}^{*} are the practical diffusion coefficients; $\vec{g}$ is the gravity acceleration vector; n is the number density; p is the pressure; t is the time; $\vec{u}$ is the mass-averaged velocity; $\vec{v}$ is the mean-number velocity of the ternary mixture; ρ is the density; η and ξ are the shear and bulk viscosity coefficients.

The equation of medium state should be added to the system of equations (1):

$$\rho=\rho_0(1-\beta_1 c_1^{/}-\beta_2 c_2^{/}), \quad (2)$$

where $\beta_i=\frac{1}{\rho_0}\left(\frac{\partial\rho}{\partial c_i}\right)_{p,T,c_j}$; $c_i^{/}$ is the concentration perturbation of the i-th constituent.

The method of small perturbations is used when solving the system of equations (1). Using this method, the system under consideration is written for the perturbed quantities as:

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

$$\frac{\partial c_1}{\partial t}+\vec{v}\nabla\langle c_1\rangle=D_{11}^{*}\nabla^2 c_1+D_{12}^{*}\nabla^2 c_2, \\ \frac{\partial c_2}{\partial t}+\vec{v}\nabla\langle c_2\rangle=D_{21}^{*}\nabla^2 c_1+D_{22}^{*}\nabla^2 c_2, \quad (3) \\ \mathrm{div}\,\vec{v}=0,$$

where β_i is the factor specifying the i-th component concentration dependence of the gas mixture density, $\boldsymbol{\gamma}$ is the unitary vector, ν is the kinematic viscosity, ρ_0 is the average mixture density. $< c_i >$ is the invariable average of the concentration considering as the reference point.

The weight-average and number-average velocities have the disturbances of the same order of magnitude. Thus, it is possible to replace $\vec{v}$ by $\vec{u}$ when the diffusion channel length H is much more than the semidiameters r of the diffusion canal.

The combined equation (3) can be presented in the nondimensional form by means of the next measures: H for the specific linear size of the cave, H^2/ν for the time, D^{*}_{22}/H for the velocity, A_iH for the i-th component concentration, and $\rho_0\nu D^{*}_{22}/H^2$ for the pressure. Then equations (3) may be written as:

$$\frac{\partial\vec{u}}{\partial t}+\frac{1}{\mathrm{Pr}_{22}}\nabla\left(\vec{u}\cdot\vec{u}\right)= \\ =-\nabla p+\Delta\vec{u}+\left(\mathrm{Ra}_1\tau_{11}c_1+\mathrm{Ra}_2 c_2\right)\vec{\gamma},$$

$$\frac{\partial c_1}{\partial t}+\frac{1}{\mathrm{Pr}_{22}}\vec{u}\nabla c_1=\frac{1}{\mathrm{Pr}_{11}}\Delta c_1+\frac{1}{\mathrm{Pr}_{22}}\tau_{12}\Delta c_2, \\ \frac{\partial c_2}{\partial t}+\frac{1}{\mathrm{Pr}_{22}}\vec{v}\nabla c_2=\frac{A_1}{A_2}\frac{1}{\mathrm{Pr}_{22}}\tau_{21}\Delta c_1+\frac{1}{\mathrm{Pr}_{22}}\Delta c_2, \quad (4)$$

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

where $\mathrm{Ra}_i=g\beta_i A_i H^4/D_{22}^{*}\nu$ is the partial Rayleigh number, where A_i is the nondimensional primary gradient of the i-th component concentration, $\mathrm{Pr}_{ii}=\nu/D_{ii}^{*}$ is the diffusion Prandtl number, $\tau_{ij}=D_{ij}^{*}/D_{22}^{*}$ are the factors defining the dependence of practical diffusion coefficients.

The numerical solution of the system of equations (4) is based on the application of a splitting methods by physical quantities [20]. The problem is modeled in the rectangular coordinate system, in which a bidimensional region of the cylinder section with dimensions $H \times d$ is allocated. The considered cylindrical region with a height H and a diameter d is shown in Figure 2a. The calculations used a nondimensional grid 128 x 128, which made it possible to fix the concentration profiles corresponding to the onset and subsequent growth of convective flows.

We assume that initially convection and diffusion lead to the momentum transfer. To find the intermediate velocity field, the three-point double-sweep method is used. In this case, for the convective and diffusion members, which have the second order of accuracy in space, the explicit Adams-Bashforth scheme and the implicit Crank-Nicolson scheme are used [21, 22]. The discretization in space is implemented by the second-order accuracy. The pressure field is determined from the obtained intermediate velocity field by means of the Fourier method. The final recalculation of the velocity field is made on the assumption that the pressure gradient is the only cause of the transfer. Applying the Crank-Nicolson scheme and taking into account the found velocity field, the concentrations of the mixture components are determined using the three-point sweep method.

4 Numerical results

Figure 3 shows the results of a numerical calculation for the mixture 0.70 He (1) + 0.30 R12 (2) – Ar (3) at a pressure of 0.5 MPa and a temperature of 298 K, illustrating the change in the concentration of the heavy component of the mixture with time. The mixture placed at the top of the diffusion channel has a density less than the density of argon located at the bottom. The isoconcentration lines presented in Figure 3 show the following sequence of mixing modes. Figure 3a shows that at the initial time the diffusion process is implemented in the system. The components' concentration profile change in a monotonous manner. Further observation of the mixing dynamics shows that the components' concentration profile are curved (see Fig. 3b). This moment of time determines the unstableness of mechanical balance with the subsequent growth of convective disturbances. Over time, the isoconcentration lines continue to bend, and there are structural formations due to the fact that in the lower part of the computational domain the component concentration with a large molar mass increases. Subsequently, the convective structure detaches (see Fig. 3d) and moves in the diffusion channel in the gravity field. The performed computational investigation subject to pressure influence indicated that at the final stages of mixing, compound structured streams arise due to a significant bend of the concentration profiles with pressure growth. Moreover, the characteristic times of the onset of the corresponding mixing regimes decrease. Thus, the intensity of convective mixing increases with increasing pressure, which is also confirmed by the experimental data shown in Figure 1b.

5 Conclusions

An experimental study was carried out to define the conditions that determine the transition from a stable diffusion process to diffusion mixing with the formation of convective flows. For the experimental study of this transition, the experimentally measured concentrations were compared with the concentration values calculated using the Stefan-Maxwell equations. Such a comparison is presented as a pressure dependence of the nondimensional factor α.

Empirical results obtained for the mixture 0.70 He (1) + 0.30 R12 (2) – Ar (3) show that, starting from a pressure of 0.15 MPa, there is change of the diffusion mode to convective one due to the violation of the mechanical balance of the mixture. Examination of the change of nondimensional factor α subject to pressure indicated that the component with the large molar mass has the significant transference in the pressure range from 0.15 to 0.60 MPa. This behavior of Freon-12 concentration on pressure indicates an intensification of the convective flows rate.

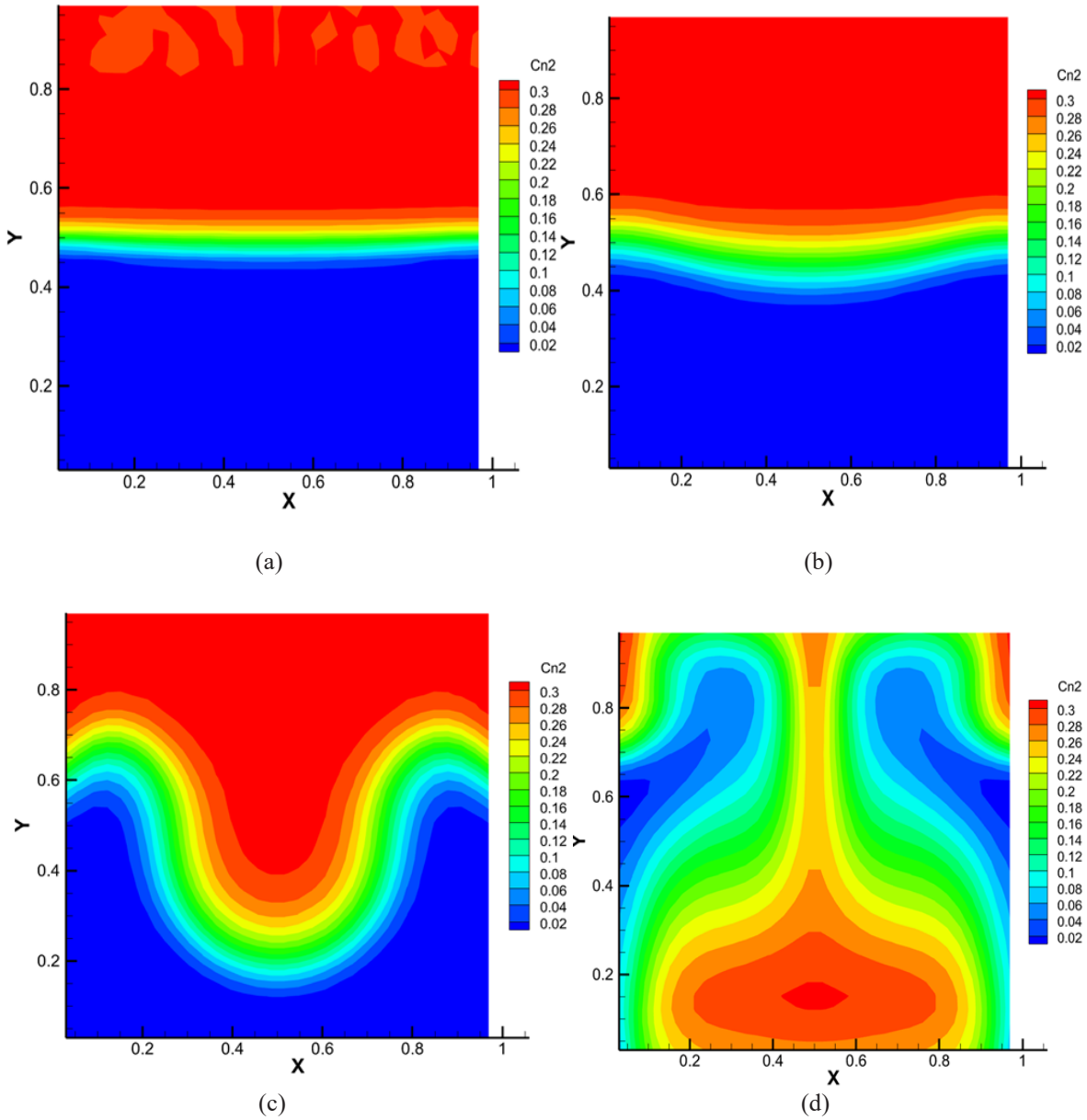


Figure 3 – Concentration profiles for the system 0.70 He (1) + 0.30 R12 (2) – Ar (3) at p = 0.5 MPa, T = 298.0 K, L = 0.165 m, $r = 3.05\times10^{-3}$ m: (a) t = 3.6 s; (b) t = 9.7 s; (c) t = 21.2 s; (d) t = 43.2 s

Within the framework of a numerical study, the dynamics of the formation of convective structures in the system 0.70 He (1) + 0.30 R12 (2) – Ar (3) at a pressure of 0.5 MPa was studied, at which, as shown by the experimental data presented in the work, a convective type of mixing is observed in the system. The curvature of the concentration profiles causes the initiation of unstability. The intense distortion of the components concentration profiles is associated with the pressure growth in the experiment.

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