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V.N. Kossov[1], S.A. Krasikov[1], M.K. Asembaeva[2]✉, E. Meirambekuly[2]

[1]*Abai Kazakh National Pedagogical University, Almaty, Kazakhstan;*
[2]*Al-Farabi Kazakh National University, Almaty, Kazakhstan*

Image of the Experimental 3D Concentration Field of the Separated Gas with Specified Thermophysical Properties under Conditions of Mechanical Equilibrium Instability in an Isothermal Ternary Mixture

Isothermal diffusion and convective mixing in a ternary He–Ar–N_2 mixture at varying pressures and initial compositions were examined experimentally by means of the two-flask method. The study was conducted under strictly controlled laboratory conditions to ensure precise, comparable, and reproducible results across all experiments. It was found that when the density decreases with height, the mechanical equilibrium of the mixture can be disturbed, causing gravitational flows and partial convection within the system. Anomalous transfer of the component with the greatest molecular mass was detected at particular pressures and starting compositions, pointing to the influence of supplementary convective processes beyond ordinary diffusion. When the experimental results were plotted in the phase space of the three variables — pressure, initial composition, and diffused component concentration — a pronounced wave-like iso-concentration surface emerged. This surface appeared in regions corresponding to well-developed convective flows. Its formation correlated with the highest intensity of partial component mixing, highlighting the combined influence of pressure, composition, and gravitational effects on the mixture's dynamic equilibrium. These findings provide insight into the interplay between diffusion and convection in multicomponent gas systems, revealing conditions under which gravitationally induced flows significantly alter component transport and overall mixture behavior.

Keywords: diffusion, instability, convection, pressure, initial composition, concentration

✉*Corresponding author*: Asembaeva, Mansiya, m.asembaeva@physics.kz

Introduction

Modern technologies for material production and the prediction of natural phenomena require a more accurate interpretation of heat and mass transfer processes in liquid and gaseous media. Complexities in the description of multicomponent systems are caused by the simultaneous influence of different heat and mass transfer mechanisms. The control of processes in such systems requires new knowledge about the specifics of combined mass transfer at different stages of mixing, especially in conditions where equilibrium is disturbed by gravitational convection [1, 2]. The generalizations presented in review studies [3, 4] on the investigation of non-isothermal mixing have shown that the emergence of different types of equilibrium instabilities is related to the complex interaction between temperature and concentration gradients. It was also noted that the transition of the studied system to a thermo-gravitational mixing regime can be realized within the framework of convective stability theory by determining the critical Rayleigh numbers that define the transition of the system from a convective state to a mixing regime. Researches of stability of equilibrium states of binary systems in non-isothermal conditions, carried out on the basis of methods presented in [1–4], have shown the existence of various specific features of thermal-concentration mixing, associated with the nature of boundary conditions, interface surfaces and the appearance of combined flows, and so on. These findings make it possible to systematically refine and apply the obtained information within the context of the studied problem. For example, the investigation of the mixing of binary mixtures of magnetic fluids in a non-uniform temperature field has shown that as a result of positive thermo-diffusion, density inversion regions are formed, which under the action of gravity destabilize the equilibrium of the system, causing the development of convective bursts [5, 6]. The peculiarity of this mixing regime is that the corresponding hydrodynamic currents manifest themselves in the so-called subcritical region, i.e., at Rayleigh numbers lower than the critical value that defines the transition between regimes, indicating a situation where, at the initial stage of mixing, the system exhibits a stable state of mechanical equilibrium.

A similar situation can occur in isothermal gas mixtures due to the diversity of diffusion mixing mechanisms. However, for binary mixtures, convective transfer arises within the traditional framework of Rayleigh–Taylor gravitational convection [7], which represents the mixing of a denser medium with a less dense one and corresponds to Rayleigh numbers exceeding the calculated critical value [8]. The opposite direction of the mixture's density gradient determines diffusive mixing [9], which is characterized by Rayleigh numbers significantly lower than the critical value that marks the onset of convection. Nevertheless, in ternary gas mixtures isothermal diffusion under certain conditions is capable of causing the appearance of analogs of convective bursts similar to those noted in [5, 6], with the subsequent development of concentration gravitational convection [9]. Despite the similarity of the effects observed in thermal and isoconcentration convection, the mechanisms responsible for the formation of inversion layers exhibit distinctive features. These differences are associated with the presence of multiple concentration gradients that give rise to specific diffusion regimes [10], which are absent in the thermal diffusion mixing described in [4–6]. One manifestation of such special diffusion regimes was reported in [11], where significant differences in the diffusion coefficients of the components lead to a nonlinear concentration distribution in a vertical channel, ultimately resulting in the inversion of the density gradient and the subsequent onset of isothermal gravitational convective flows. Several other features of diffusion mechanisms that cause concentration-driven gravitational convection are discussed in the review [12]. Among them, it is worth noting experimental results showing that the loss of system equilibrium stability leads to isothermal concentration gravitational convection accompanied by a synergistic increase in the rate of component mixing [9, 13]. In this case, the mixing intensity exhibits a nonlinear dependence on pressure, initial composition, and several other thermophysical parameters.

Thus, failure to take diffusion into account in multicomponent mass transfer leads to distortion in the description of concentration fields and their subsequent evolution. The need to take into account the appearance of gravitational convection in gas mixtures due to the violation of the mechanical equilibrium of the system will allow for a more accurate description of heat and mass transfer in multicomponent mixtures, which is not always done in diffusion experimental and computational-theoretical studies. This paper presents experimental results on the study of diffusion and the recording of convective currents caused by the disturbance of the equilibrium of a ternary mixture of helium, argon, and nitrogen in a wide range of concentrations and pressures and at a constant temperature. Based on the obtained experimental data, a quantitative assessment was made of the degree of mixing of the partial component fluxes in both the diffusive and combined regimes.

Materials and methods

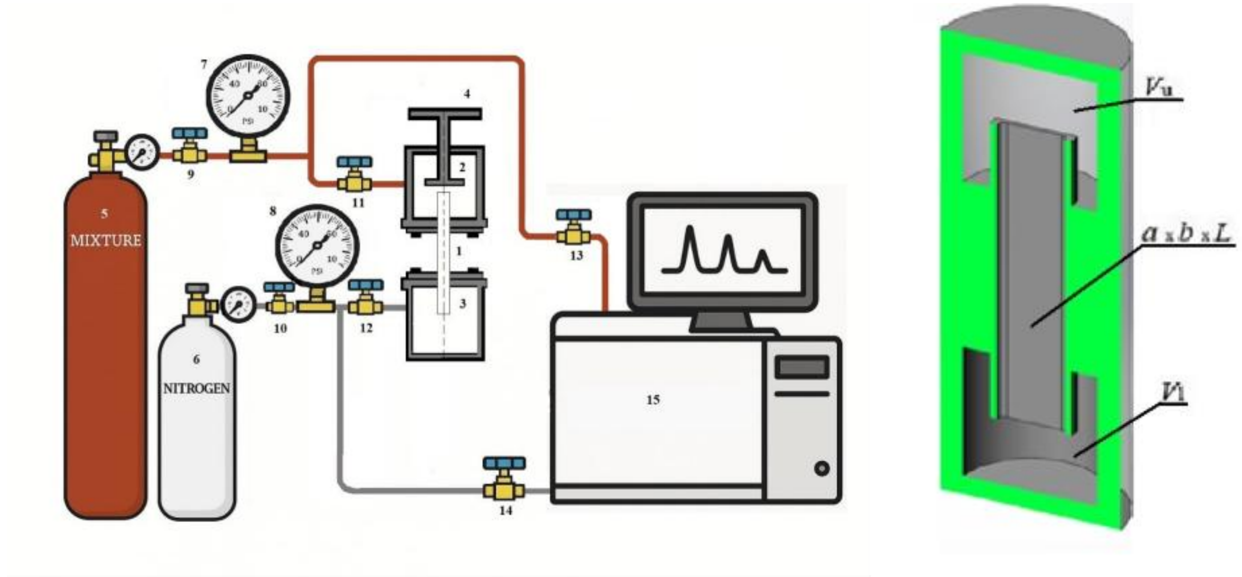
Experiment

Experimental device and experimental procedure

Experimental studies were performed on a device that implements the two-flask method [14], upgraded to measure the partial concentrations of components not only in diffusion modes, but also in the field of convective transfer using flat and cylindrical channels with different geometric parameters [15]. The schematic representation of the measuring complex is reproduced in Figure 1.

Linear parameters and dimensions of the measuring device (diffusion cell) correspond to the following values: volume of the upper bulb $V_u = (185.0 \pm 0.5)\cdot 10^{-6}$ m^3; volume of the lower bulb $V_l = (186.0 \pm 0.5)\cdot 10^{-6}$ m^3; dimensions of the rectangular vertical channel $a\times b\times L = (30.00 \pm 0.05 \times 6.00 \pm 0.05 \times 165 \pm 0.05)\cdot 10^{-3}$ m (Figure 1*b*). In all measurements, the temperature was 298 K. Mixing time 300 s.

The features of isothermal mixing were studied by analyzing the concentration and baric dependences of the ternary system He (1) + Ar (2) – N_2 (3) at constant temperature. Table 1 presents the mutual diffusion coefficients as well as some thermophysical characteristics of the miscible components at normal pressure and temperature T = 298.0 K. In further discussion, we will assume that the numbers in parentheses after the chemical element determine its numbering in the system under study. The values given before the chemical elements of the mixture determine the initial composition in mole fractions. It should also be noted that in the pressure range of 0.1–2.0 MPa and temperature T = 298.0 K, the studied helium — argon — nitrogen mixture can be considered ideal for any initial compositions.



a) Experimental setup scheme *b)* Diffusion cell

Figure 1. Implementation of the two-column method

Table 1

Some thermophysical properties of the mixture components and coefficients of mutual diffusion of gases

Properties of gases [16]			
Gas	Molecular weight, 10^{-3} kg/mole	Density, kg/m^3	Viscosity, 10^{-5}Pa·s
Helium He	4.0026	0.16	1.997
Nitrogen N_2	28.0134	1.13828	1.775
Argon Ar	39.94	1.6	2.271

Interdiffusion coefficients of gases, $D_{ij}\cdot 10^{-4}$ m^2/s [8, 16]		
$D_{\text{He-Ar}}$	$D_{\text{He-N}_2}$	$D_{\text{Ar-N}_2}$
0.745	0.621	0.230

At the initial stage of the experiment, aimed at studying both diffusive and convective separation of gas mixtures, the procedure of vacuumization of the internal volumes of the unit, including the main elements of the diffusion cell: the upper flask (2), the lower flask (3) and the connecting channel (1) is carried out. In the process of vacuumization the valve (4) remains closed. Further, the upper flask (2) is filled with a gas mixture of helium and argon from the cylinder (5) through open valves (9) and (11) with closed valve (13). The pressure of the mixture is brought to the experimental value and controlled by the manometer (7), after which the valves (9) and (11) are closed. The lower flask (3) is filled with nitrogen from the cylinder (6) through the open system of valves (10) and (12) with the valve (14) closed. The gas is supplied until the required pressure is reached, monitored by the manometer (8). When the filling of the lower flask is complete, valves (10) and (12) are closed. When the procedure of pressure equalization in flasks (2) and (3) is completed, valve (4) is opened and the process of multi-component mixing begins.

Experimental studies were carried out at pressures ranging from 0.15 to 2.55 MPa and temperature 298.0 K. The accuracy of temperature control was 0.1 K, pressure — 0.02 MPa. Determination of component concentrations was performed by gas chromatography with relative error not more than 1–3 %. Direct measurements were carried out for argon and nitrogen, and helium concentration was calculated from the condition of particle number conservation

$$\sum_{i=1}^{n} c_i = 1$$

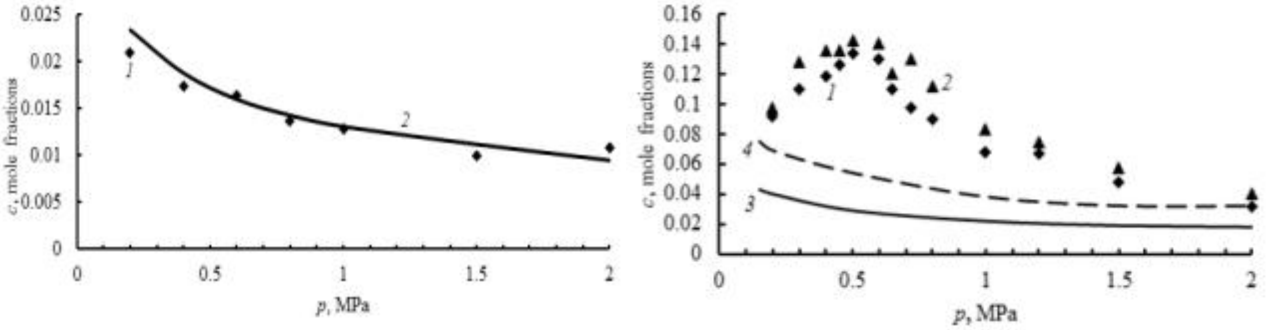
where c_i is the concentration of the i-th component. To increase reliability, the results were averaged over several series of measurements under identical conditions. In all experiments, the upper flask of the diffusion cell contained a mixture of helium and argon, whereas the lower flask contained nitrogen.

The method of obtaining the concentration and baric dependence for isolated argon from experimental data

The intensity of diffusive and convective mixing in ternary gas mixtures was quantitatively evaluated by comparing experimental concentration data with values calculated from kinetic models assuming diffusion [8]. If the experimental and calculated data coincided within the experimental error, then this type of displacement was defined as diffusive. In the case of a discrepancy between them of tens or more percent, we can talk about the manifestation of convective transfer. By applying this approach at different pressures and initial compositions of the mixture and observing the constancy of all other experimental conditions in two-dimensional coordinates, it is possible to obtain baric or isoconcentration dependencies that characterize the corresponding type of mixing. By combining the characteristic dependencies and fixing the pressure, the initial mixture composition, and the concentration of the component governing the mixing behavior, an isoconcentration surface can be obtained in the coordinates of pressure, initial composition, and concentration of the transferred component.

Results and discussion

The main key parameter for understanding the dynamics of mixing and the kinetic transition from the diffusion regime to the convective one is to record the discrepancy between experimental and calculated concentrations at a certain value of the variable parameter (pressure, initial mixture composition). Figure 3 illustrates typical dependences of the concentrations of components diffused into the lower (helium and argon) and upper (nitrogen) flasks of the diffusion cell, respectively. Figure 2*a* shows that in the 0.68 He(1) + 0.32 Ar(2) – N$_2$(3) system, in the pressure range of 0.1–1.5 MPa, the coincidence of experimental and calculated using the Stefan-Maxwell equations [8] concentrations of components is recorded. This type of mixing can be characterized as diffusion. With an increase in the fraction of argon, the component with the highest molecular weight in the mixture, a dependence uncharacteristic of diffusion is observed: the mixing intensity increases with increasing pressure (Figure 2*b*).



a) *b)*

Figure 2. Concentrations of components at different pressures in a vertical flat channel at T = 298.0 K: (*a*) — System 0.66 He + 0.34 Ar – N$_2$. Experimental points 1 — ♦ correspond to argon. Solid line 2 is calculated assuming diffusion for argon; (*b*) — System 0.43 He + 0.57 Ar – N$_2$. Experimental points ♦, ▲ correspond to: 1 — argon, 2 — nitrogen. Solid line 3 and dotted line 4 correspond to data calculated assuming diffusion for argon and nitrogen

For a number of compositions, pressure ranges are noted in which argon has a transport advantage over other components. This deviation from the classical diffusion concepts, assuming a weakening of mass transfer with increasing pressure, indicates the occurrence of convection due to the violation of mechanical equilibrium of the ternary system. The range of thermophysical parameters that determines the change in the "diffusion-convection" modes can be defined within the framework of stability theory [2], extended to the case of isothermal ternary gas mixtures [9]. However, it cannot be extended to describe combined mass transfer in the range of parameters significantly exceeding the boundary values for which nonlinear dependencies of the intensity of partial mixing of components on pressure are marked in Figure 3*b*. In this case, the condition of neutrality of convective perturbations [2] is violated, and the formalism of stability theory leads to significant quantitative discrepancies between experimental and calculated data.

Figure 3 shows the baric dependences of ternary mixtures with different argon contents in the mixture. Further experimental results are given for argon, since convective effects are most pronounced for argon. This allows us to avoid excessive detailing in the graphs and provide a more visual representation of the observed phenomena. The points correspond to experimental data for different argon contents in the initial composition of the mixture under study. Solid lines are approximation curves. According to the data obtained and the results published in [9, 13], an increase in the mixing intensity is observed as the content of the component with the highest molecular weight in the mixture increases. Visualization of this type of mixing carried out in [17] showed the presence of complex convective flows with structural formations. It should also be noted that for some convective regimes, the maximum mixing intensity is fixed.

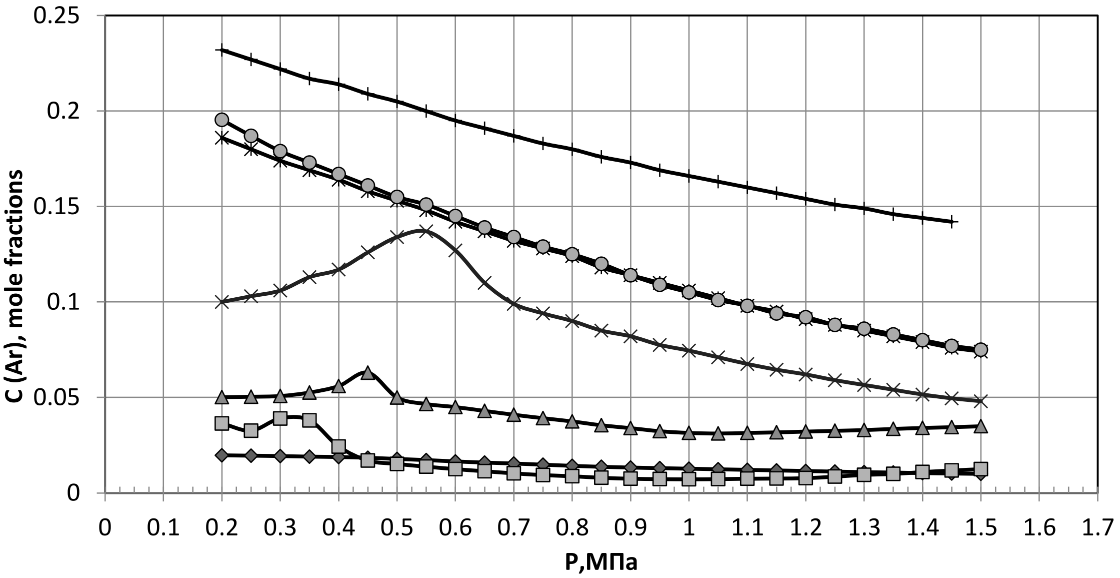


Figure 3. Baric dependencies of argon concentrations when mixing in the He + Ar – N_2 system with different initial compositions at T=298.0 K. The mixing time is $\tau = 300$ s. The points determine the argon content at different pressures in the initial composition of the mixture expressed in mole fractions and correspond to: ◆ — 0.340; ■ — 0.420; ▲ — 0.516; ✕ — 0.570; * — 0.610; ● — 0.649; + — 0.697. Solid lines are approximations of experimental data

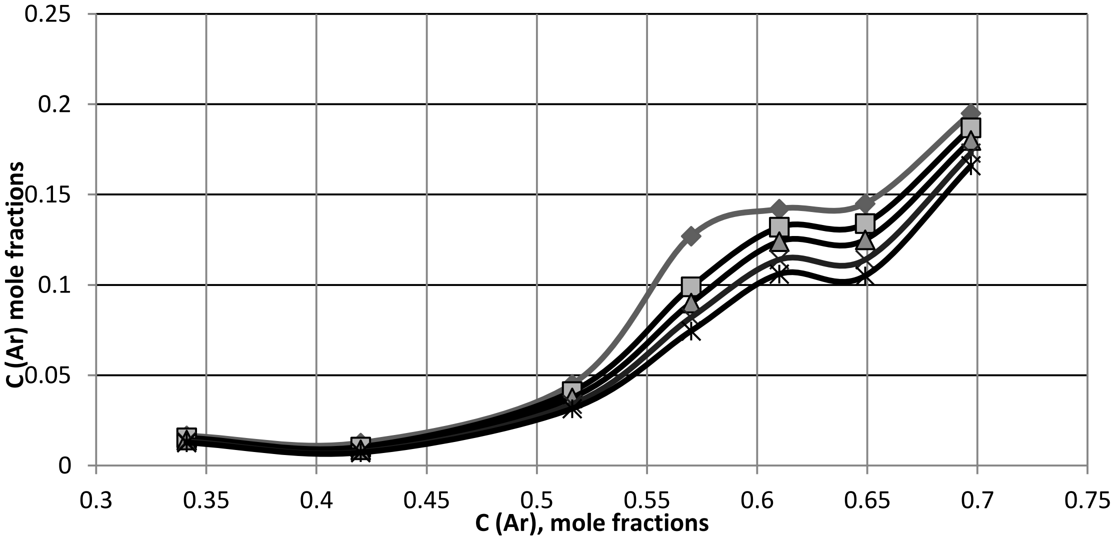


Figure 4. Argon concentrations when mixing in the He + Ar – N_2 system at a given pressure and various initial compositions of argon in a binary mixture with helium at T = 298.0 K. The mixing time is $\tau = 300$ s. Points at different compositions are determined by a given pressure value in MPa and correspond to: ◆ — 0.60; ■ — 0.70; ▲ — 0.80; ✕ — 0.90; * — 1.00. Solid lines approximate the experimental data

Figure 4 shows the isoconcentration lines, a distinct nonlinear dependence is observed: with increasing content of the component with the highest molecular weight, the intensity of convective mixing increases significantly. Also, as for the previous case, there are characteristic areas of the initial composition (0.55–0.65 mole fractions of argon) at which a pronounced nonlinearity of the mixing intensity is recorded. In figures 3 and 4, a number of characteristic modes of mass transfer can be distinguished:

1 — at concentrations of 0.34 mole fractions of argon and below, we observe a purely diffusive mass transfer;

2 — at concentrations from 0.34 to 0.61 mole fractions of argon, determining the condition of mixture density decrease with height, convection and diffusion processes are observed in the system, generating separation of the mixture into heavy (in terms of density) and light components. At certain pressures and compositions, the preferential transfer of argon is observed, which is atypical for diffusion processes;

3 — at concentrations from 0.61 to 0.649 mole fraction of argon in the system under study, the condition of approximate parity of densities in the upper and lower parts of the channel is realized (approximate equality of the mixture density gradient to zero). Convective mixing modes continue in the system, but the signs of nonlinearity of partial mixing are not registered;

4 — at concentrations from 0.649 to 0.697 mole fractions of argon, the density gradient of the mixture changes sign, convective and diffusion processes separate the components of the mixture according to the mechanisms determined by Rayleigh–Taylor convection [7].

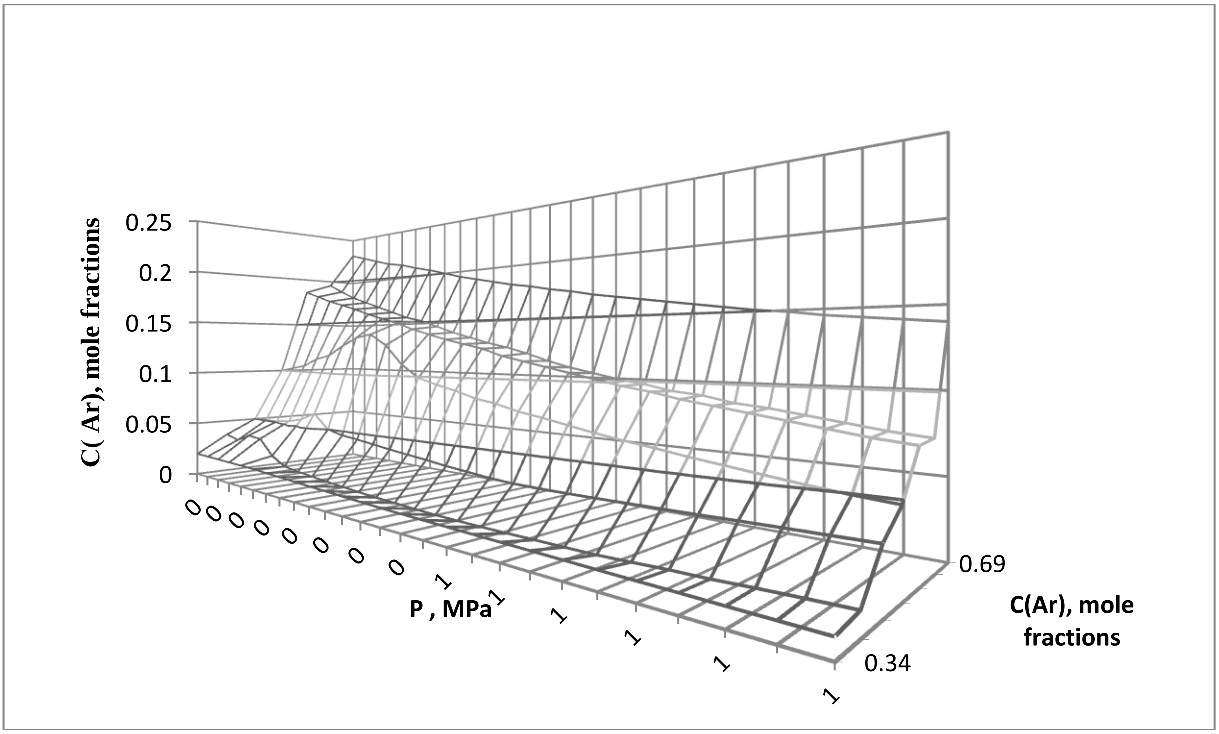


Figure 5. Three-dimensional field of argon concentrations in diffusion and convection modes in the He + Ar – N_2 system at T = 298.0 K and various initial compositions and pressures. Mixing time $\tau = 300$ s

Combining the experimental data presented in Figures 3 and 4, in the coordinates “pressure – initial composition of the mixture – diffused concentration of the component” and supplementing them with approximation lines, it is possible to obtain an experimental surface characterizing the concentration field in both diffusion and convective modes. Figure 5 shows a three-dimensional argon concentration field in the diffusion regime (up to 0.34 mole fractions of argon in the initial composition of the mixture) and then a kinetic transition to a state of gravitational concentration convection with a pronounced nonlinear increase in the intensity of mass transfer in the pressure range (0.25–0.7) MPa and the initial composition (0.42–0.57) mole fractions. The experimental values presented allow us to speak about the occurrence of spatial concentration waves, which occur at certain ratios between mixture composition and pressure leads to preferential transfer of the most dense component (see Table 1), i.e. argon. Such resonant manifestations significantly turbulate convective flows created as a result of instability of the mechanical equilibrium, as can be judged by comparing the concentrations of argon during mixing at the diffusion–convection boundary and in areas where pronounced nonlinearities are present. Moreover, as can be seen from Figure 5, the resulting concentration wave can be controlled not only by pressure and temperature, but also by the influence of these two parameters simultaneously.

Conclusion

The conducted studies show that in multicomponent mixtures, the difference in the diffusion coefficients leads to a violation of the mechanical equilibrium of the mixture and the occurrence of gravitational concentration convection. Experimental study of the helium + argon – nitrogen system under the condition of decreasing density of the mixture with altitude has shown that in certain pressure ranges and initial composition of the mixture there is a nonlinear increase in the mixing intensity for the component with the highest molecular weight, which is not characteristic of classical diffusion. Representation of this effect in the phase space of the three measured quantities —“pressure – initial mixture composition – diffused component concentration” — showed a non-monotonic, isoconcentration wave-like surface that appears in the region of developed convective flows. Its emergence corresponds to the highest intensity of partial component mixing. Control of its displacement in the phase coordinates can be achieved either through individual variations in pressure and initial mixture composition or by simultaneous adjustment of these parameters.

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В.Н. Косов, С.А. Красиков, М.К. Асембаева, Е. Мейрамбекұлы

Изотермиялық үштік қоспадағы механикалық тепе-теңдік тұрақсыздығы жағдайында көрсетілген жылуфизикалық қасиеттері бар бөлінген газдың тәжірибелік 3D концентрация өрісінің кескіні

Изотермиялық диффузия және He–Ar–N_2 үштік газ қоспасындағы конвективтік араласу әртүрлі қысымдар мен бастапқы құрамдар кезінде тік жазық каналмен жалғанған екі колба әдісі арқылы эксперименттік түрде зерттелді. Зерттеулер 0,15-тен 2,55 МПа-ға дейінгі қысымдар диапазонында және 298,0 К температурада жүргізілді. Қоспа компоненттерінің концентрациялары газдық хроматография әдісімен анықталды. Белгілі бір шарттарда тәжірибе бірнеше рет қайталанып, өлшенген шамаларды орташа алу арқылы компоненттердің концентрацияларының нақты мәндері анықталды. Қоспаның тығыздығы биіктік бойынша азаятын жағдайларда компоненттердің диффузия коэффициенттерінің әртүрлі болуы жүйеде тығыздықтың стратификациясына әкелетіні көрсетілді. Бұл өз кезегінде механикалық тепе-теңдіктің тұрақтылығының бұзылуына себеп болып, әртүрлі қарқындылықтағы гравитациялық ағындардың пайда болуын туындатады. Эксперименттік түрде белгілі бір қысымдар мен құрамдар кезінде үштік жүйелерде қоспадағы молекулалық массасы ең үлкен компоненттің басым тасымалдануы байқалатыны анықталды. Мұндай араласу диффузияға тән емес. Тәжірибелік нәтижелерді «қысым — қоспаның бастапқы құрамы — диффузияланатын компоненттің концентрациясы» атты үш өлшенетін шама фазалық кеңістігінде ұсыну барысында толқын тәрізді изоконцентрациялық бет анықталды. Бұл бет дамыған конвективтік ағындарға сәйкес келетін қысым мен құрам аймақтарында пайда болады. Оның қалыптасуы молекулалық массасы ең үлкен компоненттің араласу қарқындылығының жоғары болуымен, сондай-ақ қысым мен құрамның қоспаның динамикалық тепе-теңдігіне әсерімен байланысты.

Кілт сөздер: диффузия, тұрақсыздық, конвекция, қысым, бастапқы құрамы, концентрация

В.Н. Косов, С.А. Красиков, М.К. Асембаева, Е. Мейрамбекулы

Изображение экспериментального 3D поля концентрации выделенного газа с заданными теплофизическими свойствами в условиях неустойчивости механического равновесия в изотермической тройной смеси

Изотермическая диффузия и конвективное перемешивание в тройной газовой смеси He–Ar–N_2 при различных давлениях и начальных составах были экспериментально исследованы методом двух колб, соединенных вертикальным плоским каналом. Исследования проводились в диапазоне давлений от 0,15 до 2,55 МПа и при температуре 298,0 К. Концентрации компонентов определялись методом газовой хроматографии. При заданных условиях опыт повторялся несколько раз, и путем усреднения измеряемых величин определялось конкретное значение концентрации компонентов. Показано, что при условиях, когда плотность смеси уменьшается с высотой, различие коэффициентов диффузии компонентов приводит к стратификации плотности в системе с последующим нарушением устойчивости механического равновесия, которое вызывает появление гравитационных потоков различной интенсивности. Экспериментально обнаружено, что при определенных давлениях и составах в тройных системах возникает приоритетный перенос компонента с наибольшим молекулярным весом в смеси. Такое смешения не типично для диффузии. При представлении опытных результатов в фазовом пространстве трёх измеряемых величин — «давление – начальный состав смеси – концентрация диффундирующего компонента» — была обнаружена волнообразная изоконцентрационная поверхность. Эта поверхность появляется в областях давления и состава, соответствующих развитым конвективным течениям. Её формирование связано с наибольшей интенсивностью перемешивания компонента с наибольшим молекулярным весом и влиянием давления и состава на динамическое равновесие смеси.

Ключевые слова: диффузия, нестабильность, конвекция, давление, исходный состав, концентрация

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Information about the authors

Kossov, Vladimir — Doctor of Physical and Mathematical Sciences, Professor, Academician of the National Academy of Sciences of the Republic of Kazakhstan, Academician of the National Academy of Sciences of the High School of RK, Head of the Physics Department, Physics Department, Abai Kazakh National Pedagogical University, Almaty, Kazakhstan; e-mail: kosov_vlad_nik@list.ru; ORCID ID: https://orcid.org/0000-0002-8001-1644

Krasikov, Sergei — Candidate of Technical Sciences, Leading Research Scientist, Physics Department, Abai Kazakh National Pedagogical University, Almaty, Kazakhstan; e-mail: sa.krassikov@mail.ru; ORCID ID: https://orcid.org/0000-0001-7202-7251

Asembaeva, Mansiya (*corresponding author*) — Candidate of Physical and Mathematical Sciences, Associate Professor, Department of Thermal Physics and Technical Physics, Al-Farabi Kazakh National University, Almaty, Kazakhstan; e-mail: m.asembaeva@physics.kz; ORCID ID: https://orcid.org/0000-0003-2022-3888

Meirambekuly, Esbol — PhD student, Department of Thermal Physics and Technical Physics, Al-Farabi Kazakh National University, Almaty, Kazakhstan; e-mail: basatar_94@mail.ru