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**Comparative study of evolution of structured flows at boundary of the regime change
“diffusion — concentration convection” in isothermal multicomponent mixing in gases
by techniques of visual and numerical analysis**

During isothermal multicomponent diffusion process, the number of effects appear that are not observed visually when mixed in binary mixtures. These include occurrence of convective instability with subsequent formation of structured flows. The feature of this type of mixing is that convection is realized under conditions of decrease in density of mixture with height. Flow visualization method allows to fix information about distribution of medium parameters by dynamics of structures in convective flows. Application of computer processing methods, as well as techniques of identifying images of thermophysical fields, allows to obtain quantitative information about convective flows. For an isothermal ternary gas mixture heliumargon-nitrogen, shadow images of structural formations formed in convective flows due to the instability of mechanical equilibrium are represented in this work. To carry out digital analysis of experimental shadow images, a simplified virtual model of the lower chamber of the diffusion cell was created. Based on digital analysis of visual images, quantitative characteristics related to estimation of the size of convective formations, period of their formation, and linear velocity of convection cells when moving through diffusion channel are presented. It has been established that the growing convective disturbances arising in the system cause a change in the characteristic scale of convective cells. The analysis of shadow images also showed that a vortex is formed in convective flows, which consists mainly of a component with the highest molecular weight. Comparison of visual images of experimental fields with simulation flows is implemented, on the basis of which composition of mixture components in convective structures is estimated. It is shown that the obtained value of the concentration of the heavy component in the vortex filament can be taken as the minimum.

Keywords: gas mixtures, diffusion, instability, convection, visualization, shadow image, digital technologies, numerical modeling.

Introduction

Convective instability in gases and liquids is occurred in many technological schemes, therefore its description appeared wide interest in experimental-practical and computational-theoretical plans. Natural gravitational convection caused by instability of mechanical equilibrium of the system is complex type of motion of continuous medium with different spatial and temporal scales [1, 2]. Most modern studies of systems in which Rayleigh-Benard convection and its analogues occur themselves are based on a system of equations of continuum dynamics, mass transfer and heat simplified within the Boussinesq approximation. This approach proved to be very productive, as it allows to identify spectrum of parameters that determine transition from stable state to an unstable one [3, 4]. Due to number of simplifications, approach presented in [1-4] does not

fully consider dynamics of continuous medium in channel of given shape, as well as nonlinear spatial effects. Obviously, that with invention of powerful modern computers appears possibility to conduct computational experiments based on the numerical solution of equations used in mathematical model of physical phenomenon or process under study. However, question of correctness of comparative analysis of numerical results with reference experimental indicators raises. Moreover, such indicators can be not only direct and indirect experimental data, but also images of convective flow fields, which are the main source of information about distribution of medium parameters, configurations and dynamics of structures in the flow, current lines, turbulent vortices, etc. Panoramic visualization of convective flow field of medium is not only an important way to obtain information about structured flows in experimental studies of gas, liquid and plasma, but also provides benchmarking for testing software packages and algorithms in computational thermophysics [5].

Among wide variety of experimental visualization of convective flows in gaseous media, optical methods for studying transparent media based on the phenomenon of light deflection when it passes through inhomogeneities of density of transparent medium have become widespread [6]. As it is known, optical refractive index of medium n is equal to ratio of speed of light in medium to the speed of light in vacuum and is related to the local density of medium ρ by the Lorentz-Lorentz formula, which for gases has the form [5, 6]:

$$\frac{n-1}{\rho} = k, \quad (1)$$

where k is a constant value that has its characteristic value for particular gas.

If gas flow is inhomogeneous, in this case optical refractive index of medium in the studied flow area depends on coordinates (x, y, z) . During flow area with variable density is illuminated, beam propagating parallel to the z axis and passing through inhomogeneity deviates from original direction of propagation by an angle α

$$\alpha = \int_0^L \frac{\partial}{\partial x} \ln n(x, y, z) dz. \quad (2)$$

Density changes are summed up in direction of the light beam in medium under study and thus integral value of density change is recorded. In shadow visualization of the gas flow field, change in illumination is proportional to the degree of change in density gradient of gas. In presence of strong density gradients in flow, additional deflections of beam occur on surface of convective formation, which is dark area from the side of incoming flow in the form of light field of varying intensity.

Until recently, images obtained in framework of approximation (1), (2) were mainly of qualitative nature. However, with advent of digital age, computer processing methods, image recognition analysis tools for thermophysical fields, numerical methods modeling the motion of continuous medium allow to obtain quantitative information about flows [7-9]. Therefore, investigate of dynamics of structured flows resulting from convective instability of isothermal triple gas mixtures by comparing visual and numerical methods seems relevant, since it allows to obtain new quantitative information about partial flows of components.

The objectives of the proposed study are:

1. Obtaining visual shadow images of convective formations caused by the instability of mechanical equilibrium in an isothermal ternary mixture of helium – argon – nitrogen at high pressures.
2. Estimation of the characteristic dimensions of convective structures, which mainly consisted of a component with the highest molecular weight, and the speed of their movement in a medium with a lower density.
3. Using the FlowSimulation software included in the SolidWorks package [9], to consider the possibility of numerical simulation of individual stages of the movement of a transient structured convective flow, within which it is possible to calculate the threshold values of the concentration of the heavy component in the convective structure in the lower flask of the diffusion cell (DC).
4. Comparison of the results of numerical simulation with quantitative characteristics obtained from the analysis of shadow visual images.

Optical experimental visualization of convective flows during isothermal mixing of triple gas mixtures

Experimental studies of multicomponent mixing in gas mixtures with significant difference in diffusion coefficients have shown that, under certain conditions, convective instability may occur in them, followed by

appearance of structured flows [10, 11]. Further experimental [12-14] and computational-theoretical [15-17] studies have shown variety of thermophysical concentration flows at the boundary of “diffusion–convection” regime change. Feature of registered flows is that occur in gas systems under condition of decrease in the density of mixture with height, which is not typical for diffusion. In this regard, visual registration of dynamic features of convective flows occur particular importance, as it allows obtaining new experimental information.

Traditionally, multicomponent mixing in gas mixtures at elevated pressures is studied on devices implementing the two-column method [10, 12]. Methodology of experiment is detailed in the works [10, 12, 13], so necessary to pay attention only to the main stages of experimental study. The upper and lower flasks of diffusion cell (Fig. 1a) were filled with studied gas mixtures up to pressure of the experiment. Next channel connecting flasks was opened and at the same time, start time of the mixing process was fixed. At the completion of the experiment, channel closed, after registered time of mixing completion, and then conducted mixtures analysis from every flask by chromatography method. Experimental concentrations compared with numerical by Stefan-Maxwell equations values [18] by assuming diffusion process.

Diffusion cell (DC) was modified and experimental procedure was adjusted to obtaining visual information about dynamic processes in gas flows. In the modified diffusion cell in the lower and upper cameras windows made of quartz glass with diameter of 60 mm and a thickness of 20 mm were viewing (Fig. 1b). Windows allowed viewing almost inside part of the upper or lower cameras. Flasks were connected by diffusion channel with optical windows, which allowed visualization of convective structures inside channel (Fig. 1c). Parts of connecting channel exits by center of upper and lows of the flasks of diffusion cells (DC). This construction allowed possibility to observe mixing process not only medium of channel, and also at the end of the channel. Geometric parameters of DC are follows: volumes of the lower and upper flasks are 226.8 and 214.5 sm^3 ; cross-sectional area of the channel is $6.1 \times 6.1 \text{ mm}^2$, and its length is 165.4 mm.

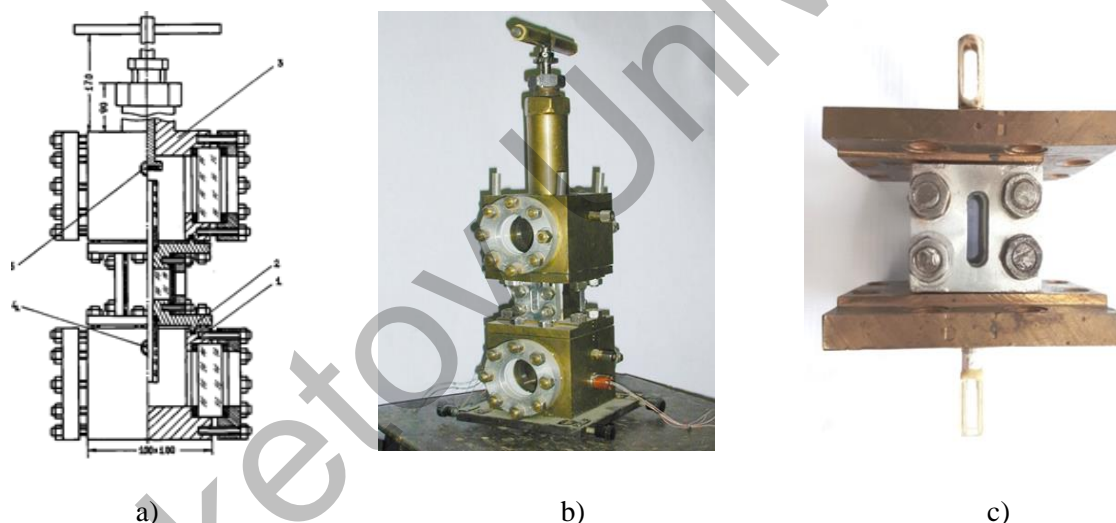


Figure 1. Diffusion cell of the two-column method: a) Plan DC. 1, 3 — lower and upper flasks; 2 — a block with diffusion channel; 4, 5 — fitting [14]; b) DC with viewing windows; c) Diffusion channel

To visualize convective flows, principle of the Schlieren system [6] was used, which consists in the fact that part of the light deflected when passing through inhomogeneity of gas density is delayed by the edge of knife installed in the focal plane of beam that passed through the area under study. As a result, change in illumination of the corresponding image areas is recorded. Changing illumination at point with density inhomogeneity is determined by magnitude of the beam deflection angle, focal length of the second lens and size of light source. When visualizing gas flow field by the Schlieren method, change in illumination is proportional to the gradient of gas density in area under study in the direction perpendicular to the knife edge. As a result, vortex structures and rarefaction areas are better visualized [5]. Optical scheme for obtaining shadow images is shown in Fig. 2 [14] and does not require additional explanations. Image was fixed to video camera by rotating the image 90° through prism not indicated in optical scheme (Fig. 2) and located after lens 11.

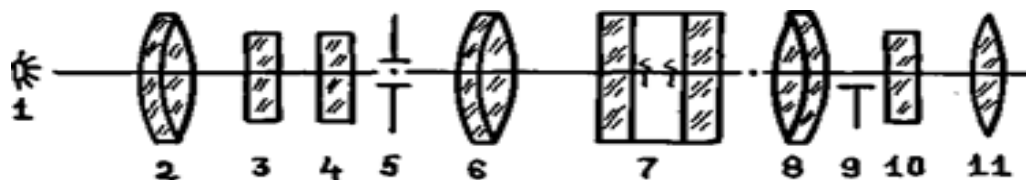


Figure 2. Optical scheme: 1 – light source; 2 – condenser lens; 3 – lightfilter; 4 – main protective glass lens; 5 – slit; 6 – main lens; 7 – camera of diffusion cell; 8 – receiving lens; 9 – knife; 10 – safety glass receiving lens; 11 – lens [14]

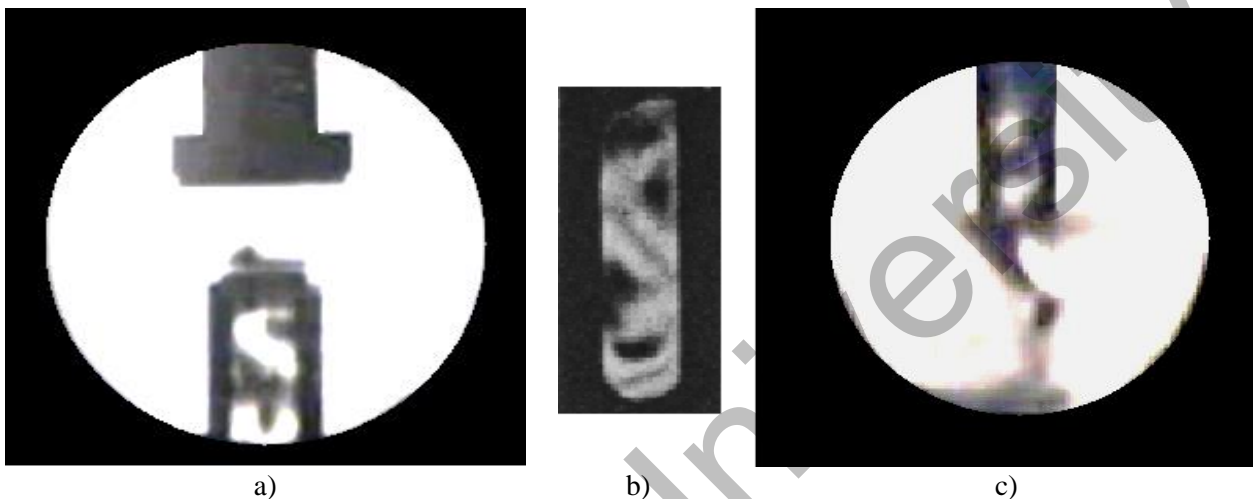


Figure 3. Motion of vortex cord in the diffusion cell. System 0.5143 He + 0.4857 Ar — N₂, $p = 2.54$ MPa, $T = 298.0$ K: a) — section of diffusion channel in the upper camera; b) — middle of the diffusion channel; c) — section of diffusion channel in the lower camera

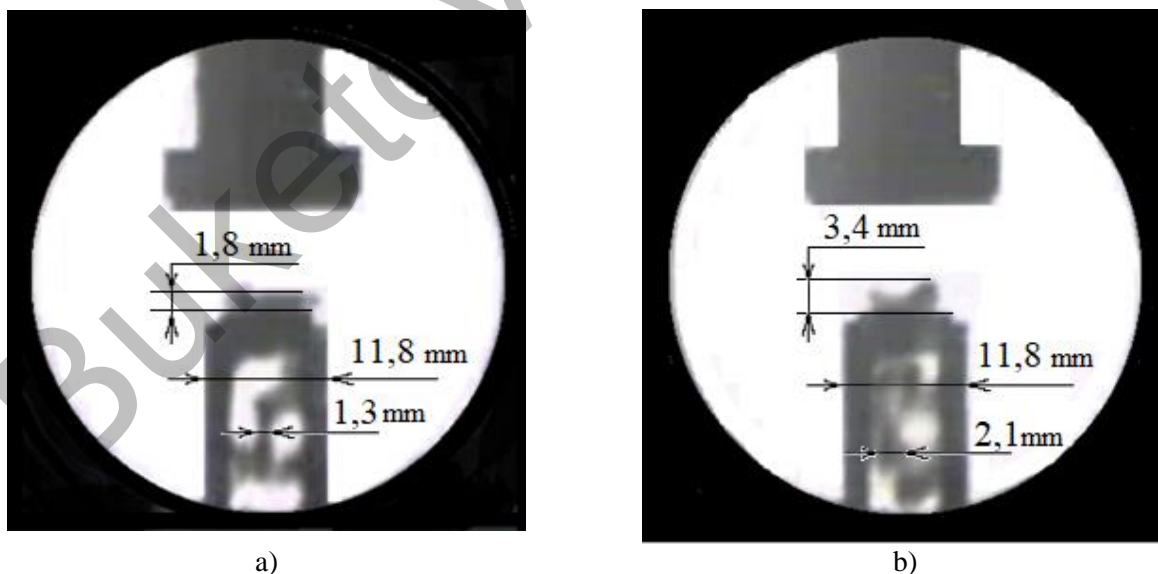


Figure 4. Shadow images of gas mixture separation zone in the upper chamber of diffusion cell for system 0.5143 He + 0.4857 Ar — N₂ at $p = 2.54$ MPa, $T = 298.0$ K: a) Initial stage of the process; b) Mode of periodic formation of vortex structures

Shadow visualization of various mixing modes was implemented when studying the evolution of convective flows in an isothermal triple gas mixture of helium-argon-nitrogen, which arose at boundary of the

regime change “diffusion — concentration gravitational convection”. Thermophysical parameters of transition and geometric characteristics of channel providing the kinetic transition from one state to another were calculated within the framework of theoretical analysis for convective stability of multicomponent gas mixture [15].

Figure 3 shows shadow images of vortex convective cords in different coordinates of the diffusion channel. Analysis of presented shadow patterns indicates that convective vortex is clearly recorded, mainly consisting of component with the highest molecular weight. A frame-by-frame analysis of visual images from the beginning of the diffusion process and the subsequent formation of convective flows makes it possible to estimate the time required to reach the front of the vortex filament from the cutoff of the diffusion channel (Fig. 3a) to the border of the lower flask window (Fig. 3c) in 1 s. This, according to rough estimates, is equivalent to a speed of $0.025 \text{ m}\cdot\text{s}^{-1}$.

Figure 4 shows shadow images of gas mixture separation zone at the initial stage of steady-state process (Fig. 4a) and after $\sim 0.5 \text{ s}$. (Fig. 4b). At the initial stage, descending convective structured vortex flows are clearly recorded. The change in the characteristic scale of convective cells from 1.3 mm (Fig. 4a) to 2.1 mm (Fig. 4b) is associated with growing convective disturbances in the system. A visual study of the evolution of convective flows at the initial stage of mixing showed that the characteristic size of the vortex filament in the diffusion channel is in the range of $1.3\text{--}3.5 \text{ mm}$.

Verification of experimental shadow images of convective flows with computational simulations of shadow images

The introduction of digital technologies in the methods of analysis of thermophysical flows and the expansion of the possibilities of numerical modeling makes it possible to mutually supplement the data of experimental visualization of flows, to validate models and algorithms for numerical calculations [19]. Verification of the calculated and experimental images of the fields of concentrations, density, temperature, and other thermophysical characteristics of the combined complex gas-dynamic flow shows satisfactory agreement in detailing the results of the experiment and subsequent interpretation of the characteristic features of the observed effects [19-20].

Based on the approaches outlined in [19-20], there is an assumption that some features associated with the evolution of convective flows caused by the instability of the mechanical equilibrium of the mixture can be detailed using algorithms for the numerical solution of continuum equations included in the packages of applied calculation programs. In this work, such a calculation was carried out using the FlowSimulation program included in the SolidWorks package created for a simplified virtual model of the lower chamber of a diffusion cell [9, 21].

Within the framework of the FlowSimulation calculation program, heat and mass transfer in vertical rectangular channels is modeled using the Navier–Stokes equations, as well as relations describing the conservation of mass, momentum and energy in a given medium [9]. When modeling convective flows, effect of turbulence averaged over small time scale on the flow parameters is used, and large-scale time changes in components of gas dynamic flow parameters averaged over small time scale are considered by introducing the corresponding time derivatives [9]. As a result, equations have additional terms — Reynolds stresses, and to close this system of equations, the equations of transfer of kinetic energy of turbulence and its dissipation are used in the framework of the $k\text{-}\varepsilon$ model of turbulence [22].

Comparison of the calculated simulation of convective formation with the experimentally observed shadow image is shown in Figure 5. When modeling movement process of front of structural formation in the lower flask, the concentration of the heaviest component of the mixture (argon) in the formed vortex structures with the following component concentrations at the outlet of the lower end of the diffusion channel was estimated: $c_{\text{Ar}} = 0.92$ and $c_{\text{He}} = 0.08$ mole fractions (Fig. 5a).

At lower values of argon, the implementation of the simulation flow was not observed. It can be considered that the obtained concentration value ($c_{\text{Ar}} = 0.92$ mole fractions) can be taken as a threshold value. In this case, it is obvious that the real value of the concentration of the heavy component in the vortex filament can be even higher. The velocity of movement of the imitation convective structure in the lower chamber of the diffusion cell is $0.027 \text{ m}\cdot\text{s}^{-1}$, which is in satisfactory agreement with the experimental values given in the previous section. In this regard, it can be assumed that for structured flows caused by the instability of the mechanical equilibrium of the mixture, the scales of inhomogeneities, sizes and dynamics of formations can be obtained in a simulation manner and compile quantitative information about the flows. It can be considered

that the comparison of numerical and experimental images of the flow visualization leads to quantitative estimates that refine the models for describing the thermo-concentration distributions for a given thermophysical field.

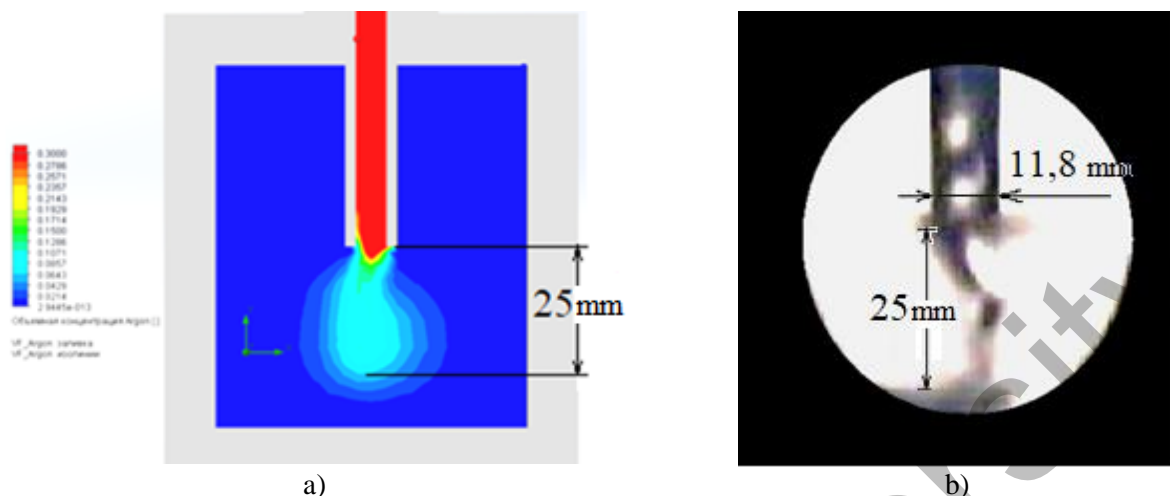


Figure 5. Comparison of the movement of convective flow front in the initial stage of mixing caused by the mechanical equilibrium instability of the ternary system $0.5143 \text{ He} + 0.4857 \text{ Ar} - \text{N}_2$ at $p = 2.54 \text{ MPa}$, $T = 298.0 \text{ K}$ in the lower chamber of diffusion cell: a) Computational simulation of shadow images at $c_{\text{Ar}} = 0.92$, $c_{\text{He}} = 0.08$, $t = 1.0 \text{ s}$; b) Shadow image of convective formation in the lower chamber of diffusion cell

Thus, analysis of images shown in Figures 3-5 allows to specify the mixing mechanism caused by instability of mechanical equilibrium in isothermal triple gas mixtures, provided that density of mixture decreases with the height of channel, which was formulated in [10, 11, 14, 15]. As it was shown in [23] under isothermal conditions due to the high mobility of helium molecules, its active penetration into nitrogen forms an increased argon content in the upper part of the channel, thereby forming an inversion layer in density, causing convective movement of the component with the highest molecular weight due to gravity. Active motion of the argon leads to convective displacement of nitrogen to upper chamber. Shadow images fix the dynamics of the separation process of argon and helium during their interaction with nitrogen in different coordinates of the diffusion channel (Figs. 3-4). Entry of convective flows, predominantly consisting of nitrogen into the upper camera, implements mixing mechanism similar an inversion layer formation. At the same time, the high diffusion mobility of helium ensures its priority penetration into the ascending nitrogen flows, thereby reducing its content in the local convective area and making shadow visualization invisible. Argon enrichment of convective formation with subsequent occurrence of hydrodynamic flow of component with the highest molecular weight leads to movement in vertical channel connecting flasks of the DC. In this case, counter (ascending and descending) convective flows are formed in the channel, i.e. a diametrically antisymmetric movement is observed (the channel is divided by a vertical plane passing through the axis into two parts, in one of which the gas rises, and in the other it falls). Counter flows differ in composition. The downstream contains more argon than the upstream, which is predominantly nitrogen. When moving due to transverse diffusion, the flows will exchange molecules of the light component (helium). The descending flow will be depleted in helium, which leads to an increase in the intensity of the convective flow. Process will continue until argon concentration exceeds certain critical value in the local areas near the cutoff in the upper part of diffusion channel. The resulting circulation of the gas mixture explains the continuity and sufficient duration of the process of convective separation in the case of diffusion instability, which was recorded in experiments [10, 11, 14].

Conclusions

Researches are conducted on the study of visual shadow images of structural formations that arose in convective flows due to the instability of the mechanical equilibrium of the isothermal triple gas mixture helium-argon-nitrogen showed:

1. Application of digital technologies for processing visual shadow images allows to obtain quantitative characteristics for estimating the size of convective formations, period of their formation, and linear velocity of convection cells when moving through diffusion channel.

2. Comparison of visual images of experimental fields with simulated flows obtained numerically provides an opportunity to evaluate quantitative characteristics associated with the composition of components in convective cells, their subsequent dynamics in a medium with different density.

3. The introduction of digital technologies in the methods of registration and analysis of visual images of convective flows makes it possible to verify numerical models of diffusion instability processes. Comparison of digital processing of the results of a physical experiment with the results of numerical simulation makes it possible to clarify the mechanism for the occurrence of convective instability during isothermal mixing in ternary gas mixtures, and to detail the types of vortex flows.

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Көрінетін және сандық талдау құралдарымен газдардағы изотермиялық көпкомпонентті араластыру кезінде «диффузия — концентрациялық конвекция» режимдерінің өзгеру шекарасындағы құрылымдалған ағыстардың эволюциясын салыстырмалы зерттеу

Газдардағы изотермиялық көпкомпонентті диффузия кезінде бинарлы қоспалар араласқан кезде байқалмайтын бірқатар әсерлер орын алады. Бұған конвективті тұрақсыздықтың пайда болуы, содан кейін құрылымдық ағыстардың пайда болуы жатады. Араластырудың бұл түрінің ерекшелігі — конвекция қоспаның тығыздығының биіктігімен төмендеуі жағдайында жүзеге асырылады. Ағысты визуализациялау әдісі қоршаған орта параметрлерінің таралуы, конвективті ағындардағы құрылымдардың динамикасы туралы ақпаратты алуға мүмкіндік береді. Компьютерлік өңдеу әдістерін, сондай-ақ жылуфизикалық өрістердің кескінін анықтауға арналған құралдарды пайдалану конвективті ағындар туралы сандық ақпаратты алуға ықпал етеді. Мақалада «гелий-аргон-азот» изотермиялық үштік газ қоспасы үшін механикалық тепе-теңдіктің тұрақсыздығынан конвективтік ағындарда түзілетін құрылымдық түзілістердің көлеңкелі кескіндері берілген. Эксперименттік көлеңкелі кескіндердің сандық талдауын жүргізу үшін диффузиялық ұяшықтың төменгі камерасының жеңілдетілген виртуалды моделі жасалды. Көрнекі кескіндерді сандық талдау негізінде конвективті түзілімдердің мөлшерін, олардың пайда болу кезеңін, диффузиялық канал арқылы қозғалу кезінде конвекция ұяшықтарының сызықтық жылдамдығын бағалауға байланысты сандық сипаттамалар берілген. Конвективті ұйытқулардың жүйеде өсуі конвективті ұяшықтың сипатты масштабының өзгеруіне әкелетіні анықталды. Сонымен қатар, көлеңкелі кескіндерді талдау конвективті ағыстарда ең үлкен молекулалық салмағы бар компоненттен тұратын құйын пайда болатындығын көрсетті. Эксперименттік өрістердің көрінетін кескіндерін имитациялық ағыстармен салыстыру жүргізілді, оның негізінде конвективті құрылымдардағы қоспа компоненттерінің құрамы бағаланды. Құйынды сымдағы ауыр компоненттің концентрациясының алынған мәні минималды ретінде қабылдануы мүмкін екендігі көрсетілген.

Кілт сөздер: газ қоспалары, диффузия, тұрақсыздық, конвекция, визуализация, көлеңкелі кескін, сандық технологиялар, сандық үлгілеу.

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Сравнительное исследование эволюции структурированных течений на границе смены режимов «диффузия–концентрационная конвекция» при изотермическом многокомпонентном смешении в газах средствами визуального и численного анализа

При изотермической многокомпонентной диффузии в газах проявляется ряд эффектов, которые не наблюдаются при смешении в бинарных смесях. К таковым можно отнести возникновение конвективной неустойчивости с последующим образованием структурированных течений.

Особенность такого типа смешения заключается в том, что конвекция реализуется при условиях уменьшения плотности смеси с высотой. Метод визуализации потоков позволяет фиксировать информацию о распределении параметров среды, динамике структур в конвективных потоках. Использование методов компьютерной обработки, а также средств идентификации изображений теплофизических полей способствует получению количественной информации о конвективных потоках. В статье для изотермической тройной газовой смеси «гелий–аргон–азот» представлены теневые изображения структурных формирований, образовавшихся в конвективных потоках, обусловленных неустойчивостью механического равновесия. Для осуществления цифрового анализа экспериментальных теневых изображений была создана упрощенная виртуальная модель нижней камеры диффузионной ячейки. На основе цифрового анализа визуальных изображений приведены количественные характеристики, связанные с оценкой размеров конвективных формирований, периода их образования, линейной скорости ячеек конвекции при движении по диффузионному каналу. Установлено, что возникающие в системе нарастающие конвективные возмущения обуславливают изменение характерного масштаба конвективных ячеек. Анализ теневых изображений также показал, что в конвективных потоках формируется вихрь, состоящий преимущественно из компонента с наибольшим молекулярным весом. Проведено сравнение визуальных изображений экспериментальных полей с имитационными течениями, на основе которого оценен состав компонентов смеси в конвективных структурах. Показано, что полученное значение концентрации тяжелого компонента в вихревом шнуре может быть принято как минимальное.

Ключевые слова: газовые смеси, диффузия, неустойчивость, конвекция, визуализация, теневое изображение, цифровые технологии, численное моделирование.

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