МРНТИ 30.17.35
УДК 533.15:536.25

10.51889/2959-5894.2023.83.3.009

FORMATION OF CONVECTIVE FLOWS IN THREE-COMPONENT GAS MIXTURES AT DIFFERENT DIFFUSION CHANNEL RADIUS

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Abstract

There are number of effects that distinguish multi-component gas diffusion from binary diffusion. According to the analysis of existing works, convective instability occurs in some systems with significantly different diffusion coefficients and specific geometric and thermophysical properties. By analyzing the stability of the system, it is possible to determine the parameters that can cause transition from diffusive state to convective one. However, this method does not allow to consider the dynamics of the process to characterize the development of convective flows in three-component systems and to assess the influence of the diffusion channel size on the occurrence of concentration gravitational convection. The paper considers the application of mathematical model implementing the method of splitting by physical parameters to describe the occurrence of convective flows. The concentration fields of gas with the highest molecular mass at different moments of time are numerically calculated.

Keywords: gas mixtures; diffusion; convection; instability; numerical calculation; isothermal mixing.

Аңдатпа

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ДИФФУЗИЯЛЫҚ КАНАЛДЫҢ ӘРТҮРЛІ РАДИУСТАРЫНДА ҮШКОМПОНЕНТТІ ГАЗ ҚОСПАЛАРЫНДАҒЫ КОНВЕКТИВТІ АҒЫНДАРДЫҢ ПАЙДА БОЛУЫ

Көпкомпонентті газ диффузиясын бинарлы диффузиядан ажыратын бірқатар әсерлер бар. Қолданыстағы жұмыстар көрсеткендей, конвективті орнықсыздық диффузия коэффициенттері мен геометриялық және жылуфизикалық қасиеттері ерекшеленетін кейбір жүйелерде дамуы мүмкін. Орнықтылықты талдауды қолдана отырып, диффузиялық күйден конвективті күйге ауысуға болатын параметрлер ауқымын анықтауға болады. Алайда, бұл әдіс үшкомпонентті жүйелердегі конвективті ағындардың дамуын сипаттау және диффузиялық канал өлшемдерінің концентрация-гравитациялық конвекцияның пайда болуына әсерін бағалау үшін процестің динамикасын қарастыруға мүмкіндік бермейді. Жұмыста конвективті ағындардың пайда болуын сипаттау үшін физикалық параметрлер бойынша бөлу әдісін жүзеге асыратын математикалық модельді қолдану қарастырылған. Уақыттың әртүрлі мәндерінде ең үлкен молекулалық массаға ие газ концентрациясының өрістері сандық есептеледі.

Түйін сөздер: газ қоспалары; диффузия; конвекция; орнықсыздық; сандық есептеу; изотермдік араласу.

Аннотация

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ОБРАЗОВАНИЕ КОНВЕКТИВНЫХ ТЕЧЕНИЙ В ТРЕХКОМПОНЕНТНЫХ ГАЗОВЫХ СМЕСЯХ ПРИ РАЗЛИЧНЫХ РАДИУСАХ ДИФФУЗИОННОГО КАНАЛА

Существует ряд эффектов, которые отличают многокомпонентную газовую диффузию от бинарной. Согласно анализу существующих работ, конвективная неустойчивость может развиться в некоторых системах со значительно различающимися коэффициентами диффузии и специфическими геометрическими и теплофизическими свойствами. Можно определить диапазон параметров, при которых возможен переход из диффузионного состояния в конвективное, используя анализ на устойчивость. Тем не менее, этот метод не позволяет рассмотреть динамику процесса, чтобы охарактеризовать развитие конвективных потоков в трехкомпонентных системах и оценить влияние размера диффузионного канала на возникновение концентрационной гравитационной конвекции. В работе рассмотрено применение математической модели, реализующей метод расщепления по физическим параметрам для описания возникновения конвективных потоков. Численно рассчитаны поля концентрации газа с наибольшей молекулярной массой в разные моменты времени.

Ключевые слова: газовые смеси; диффузия; конвекция; нестабильность; численный расчет; изотермическое перемешивание.

1. Introduction

In recent times, there has been a growing emphasis on investigating the Rayleigh-Bénard convection problem, which stands as one of the few classical quandaries in the domain of heat and mass transfer [1]. The diverse array of flow patterns that manifest within diffusion channels characterized by specific geometries and gas medium properties is a subject of great intrigue, holding significance from both theoretical and practical standpoints. Through the exploration of gravitational convection within a homogeneously heated environment, it becomes feasible to ascertain parameters related to heat and mass transfer while also facilitating the modeling of optimal geometric attributes for the diffusion channel [2, 3].

The occurrence of instability in mechanical equilibrium and the consequent development of structured convective patterns are notably influenced by the orientations of the density and gravity gradient vectors. Remarkably, it has been observed that the inception and evolution of convective disturbances transpire under varying directions of these mentioned vectors [3].

Nonetheless, when dealing with multicomponent mixtures characterized by the interaction of multiple concentration fluxes, the extension of theoretical frameworks used to describe system behavior may introduce distortions in the anticipated outcomes pertaining to mass transfer. This arises from the omission of factors that are absent in binary systems but come into play in more complex scenarios. One of these crucial factors is the destabilizing influence of diffusion on the evolution of convective flows within mixtures [4].

Convective flows arising in this case form synergetic mixing mechanism leading to selective transfer of a component with given thermophysical properties.

Experimental investigations into diffusion-driven mass transfer within isothermal three-component gas mixtures have revealed that under specific circumstances, such as variations in the diffusion coefficients of the individual components, particular pressure and temperature conditions, initial mixture compositions, and geometric characteristics of the diffusion channel, conditions conducive to the onset of concentration-driven convection manifest themselves [5, 6].

An intriguing aspect of these experimental studies is that concentration-driven gravitational convection was observed in systems initially characterized by stable density stratification of the mixture. Subsequent investigations into the modulation of "diffusion-concentration gravitational convection" regimes have highlighted that the initiation and progression of convective patterns are notably contingent on various factors. These factors include the relative diffusion coefficients of the components, the prevailing pressure, the initial composition of the mixtures scrutinized [9], as well as the geometric attributes of the cavities and the properties of the medium. These influences have been analyzed in a manner akin to analogous issues related to heat-driven gravitational convection.

Observations of convection utilizing both the shadow method and the catarometric sensor method have unveiled a spectrum of convective patterns, typically categorized into the following groups [4]: 1 - chaotic currents characterized by a multitude of counterflows; 2 - dripping convection regime marked by easily discernible convective structures; 3 - fixed strata that define a state of laminar convection.

Elucidating the factors governing the emergence of these patterns is a pertinent endeavor. In this paper, numerical modeling is used to investigate conditions leading to droplet type convection. The results are compared with experimental data.

2. Research method

The collective movement of an isothermal ternary gas mixture on a macroscopic scale is elucidated through a comprehensive system of hydrodynamic equations. This system encompasses the Navier-Stokes equations, equations governing the conservation of particle numbers, and equations pertaining to the behavior of individual components within the mixture. Assuming independent diffusion for an isothermal gas mixture, when $\sum_{i=1}^{3}\vec{J_i}=0$, $\sum_{i=1}^{3}\vec{c_i}=1$, the system of equations governing its macroscopic motion can be expressed in the following form:

$$\rho[\frac{\partial\vec{u}}{\partial t}+(\vec{u}\nabla\vec{u})]=-\nabla p+\eta\nabla^2\vec{u}+(\frac{\eta}{3}+\xi)\nabla div\vec{u}+\rho\vec{g},$$

$$\frac{\partial n}{\partial t}+div(n\vec{v})=0,$$

$$\frac{\partial c_i}{\partial t} + \vec{v}\nabla c_i = -div\vec{J_i}, \tag{1}$$

$$\vec{J_1} = -(D^*_{11}\nabla c_1 + D^*_{12}\nabla c_2),$$

$$\vec{J_2} = -(D^*_{21}\nabla c_1 + D^*_{22}\nabla c_2),$$

where $\vec{u}$ – average mass velocity, $\vec{v}$ – average numerical speed, p – pressure, c_i – concentration of the i-th component, ρ – density, $\vec{g}$ – acceleration of gravity, $\vec{J}_i$ – the density of the diffusion flow of the i-th component, η и ξ – shear and volumetric viscosity coefficients, n – numerical density, D^*_{ij} – practical coefficients of triple diffusion which are defined as follows:

$$D^*_{11} = \frac{D_{13}[c_1 D_{32} + (c_2 + c_3)D_{12}]}{D}$$

$$D^*_{12} = -\frac{c_1 D_{23}[D_{12} - D_{13}]}{D}$$

$$D^*_{22} = \frac{D_{23}[c_2 D_{13} + (c_1 + c_3)D_{12}]}{D} \tag{1.1}$$

$$D^*_{21} = -\frac{c_2 D_{13}[D_{12} - D_{23}]}{D}$$

$$D = c_1 D_{23} + c_2 D_{13} + c_3 D_{12}$$

The equations presented earlier are accompanied by an equation that describes the state of the gas mixture

$$\rho = \rho(c_1, c_2, p), T = const,$$

which relates the thermodynamic parameters in (1).

The method of small perturbations was applied to resolve the system of equations presented in (1). The resultant system of equations, which describes gravitational concentration convection for perturbed quantities in dimensionless units, can be expressed as follows:

$$\mathrm{Pr}_{22}\frac{\partial c_1}{\partial t} - (\vec{u}\vec{\gamma}) = \tau_{11}\nabla^2 c_1 + \frac{A_2}{A_1}\tau_{12}\nabla^2 c_2,$$

$$\mathrm{Pr}_{22}\frac{\partial c_2}{\partial t} - (\vec{u}\vec{\gamma}) = \frac{A_1}{A_2}\tau_{21}\nabla^2 c_1 + \nabla^2 c_2, \tag{2}$$

$$\frac{\partial \vec{u}}{\partial t} = -\nabla p + \nabla^2\vec{u} + (Ra_1\tau_{11}c_1 + Ra_2 c_2)\vec{\gamma},$$

$$div\vec{v} = 0,$$

where

$\mathrm{Pr}_{ii} = \frac{\nu}{D^*_{ii}}$ – the diffusion Prandtl number;

$Ra_i = \frac{g\beta_i A_i r^4}{\nu D^*_{ii}}$ – partial Rayleigh number;

$\tau_{ij} = \frac{D_{ij}^*}{D_{22}^*}$ – parameters determining the relationship between practical diffusion coefficients,

v – kinematic viscosity, r – characteristic geometric size, β_i – **the coefficient of linear dependence of the density of the i-th component on the concentration**, A_i – dimensionless concentration gradient of the i-th component**,** $\vec{\gamma}$ – a unit vector directed vertically upwards.

The system of equations (2) was solved numerically using the splitting method by physical parameters [7-9]. The set of equations given in (2) is addressed by solving them within the context of flat vertical cylindrical channel. This is done while considering boundary conditions that involve the cessation of both velocity and the movement of substances at the boundaries of the channel.

$$r = \pm 1,\ x = \pm 1,\ u = 0,\ \frac{\partial c_i}{\partial r} = 0,\ \frac{\partial c_i}{\partial x} = 0 \tag{2.2}$$

where x is the horizontal axis.

The resolution of the equations in the system described by system of equations (2) for a vertical cylindrical channel with finite height led to the establishment of a boundary relationship, expressed in terms of Rayleigh numbers. This relationship determines the alteration of diffusion-convection modes, as depicted by equation (2.3):

$$\tau_{11}(1-\frac{A_2}{A_1}\tau_{12})Ra_1 + (\tau_{11} - \frac{A_1}{A_2}\tau_{21})Ra_2 = \gamma^4(\tau_{11} - \tau_{12}\tau_{21}) \tag{2.3}$$

where $\gamma = Ra^{1/4}$, i.e., $\gamma = (Ra_1\tau_{11}K_1 + Ra_2K_2)^{1/4}$, $K_1 = (1-(A_2/A_1)\tau_{12})/(\tau_{11} - \tau_{12}\tau_{21})$, $K_2 = (1-(A_1/A_2)\tau_{21})/(\tau_{11} - \tau_{12}\tau_{21})$; A_i is a partial concentration gradient of component i; and equation (2.4):

$$\int u\nabla^2 u dV + Ra_1\tau_{11}\int u_z c_1 dV + Ra_2 = 0 \tag{2.4}$$

where u_z is the vertical velocity component.

Equations (2.3) and (2.4) establish the boundary that characterizes monotonic instability in the (Ra_1, Ra_2) coordinates. This boundary serves to delineate the areas corresponding to stable diffusion and diffusion instability.

To facilitate the calculated data for assessing stability and instability regions, the Rayleigh partial numbers are provided in alignment with equations (2.3) and (2.4) as detailed in equation (2.5):

$$Ra_1 = \frac{gnr^4\Delta m_1}{\rho v D_{11}^*}\frac{\partial c_1}{\partial z}$$

$$Ra_1 = \frac{gnr^4\Delta m_2}{\rho v D_{22}^*}\frac{\partial c_2}{\partial z} \tag{2.5}$$

where g is an acceleration due to gravity, m_i is the mass of the molecule i, $\Delta m_1 = m_1 - m_3$, $\Delta m_2 = m_2 - m_3$, and n is the molecular concentration.

Two - dimensional cross-sectional area of a cylinder with an area $H \times d$ (H – the height of the cylinder, d – the diameter of the cylinder) in a Cartesian coordinate system is considered. The calculations were performed using a grid that was dimensionless and had dimensions of 128×128. The calculations were divided into four stages:

1. The initial phase of the computation relies on the utilization of the Adams-Bashforth and Crank-Nicolson method. Under this assumption, it is implied that the transfer of momentum occurs exclusively through the mechanisms of convection and diffusion.

$$\frac{u^{n}-u^{n}}{\Delta t}=-\frac{1}{2}(3H^{n}-H^{n-1})+\frac{1}{2}\Delta(u^{n}+u^{n})+(Ra_1\tau_{11}c_1+Ra_2c_2)\gamma, \quad (3)$$

where $H^{n}=-\nabla(u*u)^{n}$.

2. The second phase of the calculation entails modeling the interplay between pressure and velocity.

$$\Delta p=\frac{\nabla*u^{n}}{\Delta t} \quad (4)$$

3. In the third stage of the calculation, it is outlined how the velocity field evolves in response to a reduction in pressure.

$$\frac{u^{n+1}-u^{n}}{\Delta t}=-\nabla p \quad (5)$$

4. In the fourth stage of the calculation, the focus shifts to determining the concentration of the components within the mixture.

$$\begin{gathered}\frac{c_1^{n+1}-c_1^{n}}{\Delta t}=\frac{1}{2}(F^{n+1}-F^{n})+\frac{1}{\mathrm{Pr}_{11}}\Delta c_1^{n}+\frac{1}{\mathrm{Pr}_{12}}\Delta c_2^{n}\\ \frac{c_2^{n+1}-c_2^{n}}{\Delta t}=\frac{1}{2}(F^{n+1}-F^{n})+\frac{1}{\mathrm{Pr}_{21}}\Delta c_1^{n}+\frac{1}{\mathrm{Pr}_{22}}\Delta c_2^{n}\\ c_3^{n+1}=1-c_1^{n+1}-c_2^{n+1}\end{gathered} \quad (6)$$

where $F^{n}=-\nabla(u^{n+1}*c^{n})$ – convection members.

The problem at hand was resolved under the subsequent boundary conditions:

$$u(x_b,\tau)=0,\frac{\partial c_i}{\partial n}=0,i=1...3. \quad (7)$$

3. Numerical calculation results

The results of the numerical study are illustrated in Figure 1, which portrays the temporal evolution of the density-normalized concentration of the heaviest component, butanol, within the ternary system composed of 0.8 H_2O + 0.2 $C_4H_{10}O$ – N_2. This system was experimentally examined in [10]. By scrutinizing the graphs, various mixing modes can be discerned.

Initially, as depicted in Fig. 1a, diffusion dominates the process. However, at a time of 6.32 seconds from the initiation of the process, there is an evident departure from the expected monotonic distribution of isoconcentration lines (Fig. 1b). This deviation is not characteristic of the diffusion-based mixing process and implies the occurrence of mechanical equilibrium instability within the system at that particular moment.

Figure 1d shows the convective cell that emerged 12.63 seconds after the start of mixing. Continued mixing time leads to the creation of a droplet (Fig. 1e) and its subsequent detachment (Fig. 1f), indicating a drip mixing mode in the vapor-gas system. With different initial conditions, the process of structure formation starts again.

Given that one of the parameters influencing the onset of the convective process is the geometry of the diffusion channel, specifically its radius, calculations were carried out to ascertain the time points at which distinct types of flows are established for varying channel radii.

Table 1 presents the mixing times for different diffusion channel radii in a triple mixture of 0.8 H_2O + 0.2 $C_4H_{10}O$ – N_2. The mixing times include the time for diffusion (t_1), the occurrence of mechanical equilibrium instability (t_2), and the formation of structure (t_3).

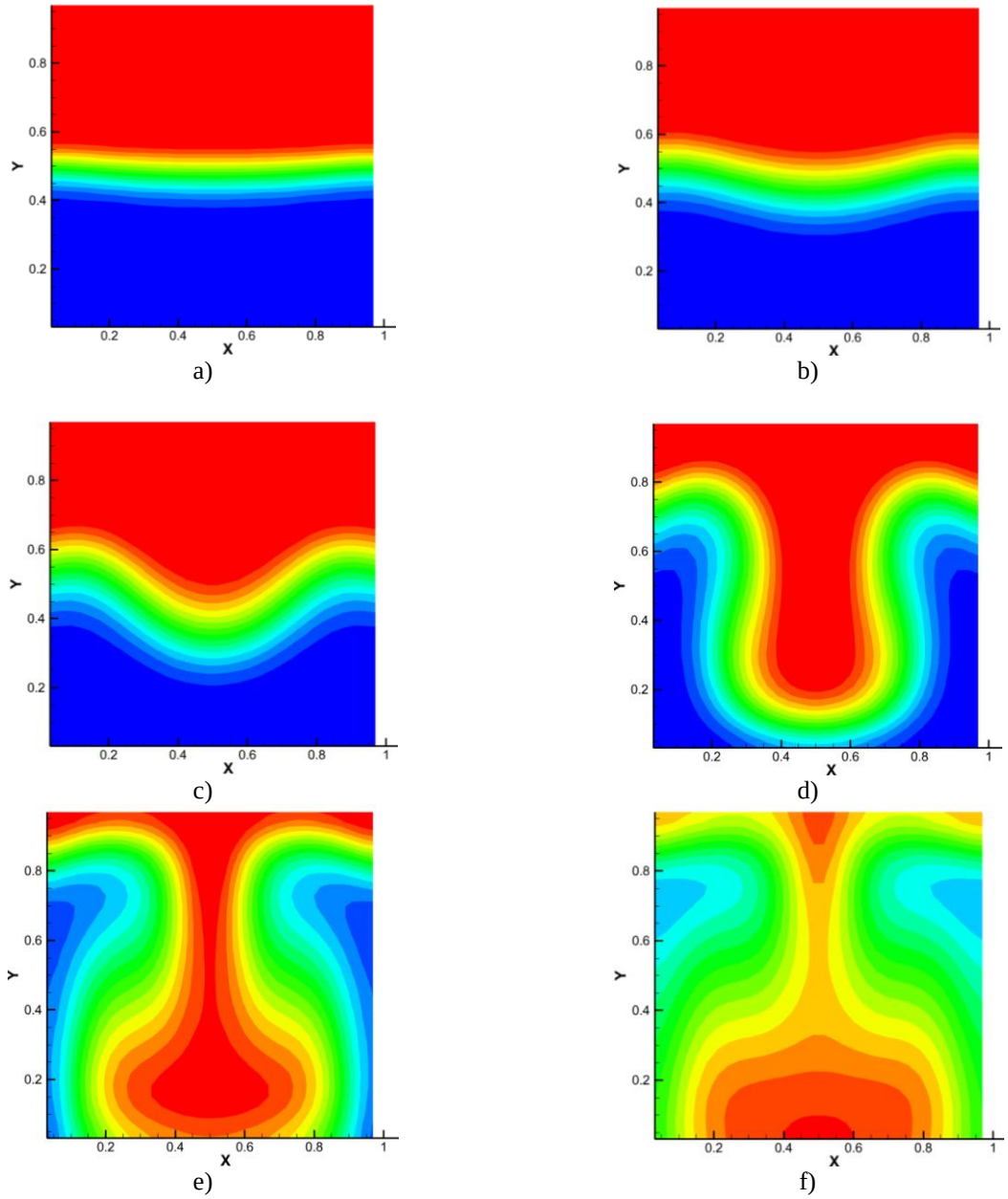

Figure 1. Butanol isoconcentration lines for the system 0.8 H_2O + 0.2 $C_4H_{10}O$ – N_2 at p = 0.1 MPa, T = 298.0 K and r = 8.5 mm:
a) t_1 = 4.24 s; b) t_2 = 6.32 s; c) t_3 = 8.42 s; d) t_4 = 12.63 s; e) t_5 = 14.7 s; f) t_6 = 18.96 s.

Table 1. Mixing time of butanol at different radii of the diffusion channel

0.8 H_2O + 0.2 $C_4H_{10}O$ – N_2 (p = 0.1 MPa, T = 298.0 K)				
t, s	*r = 6.5 mm*	*r = 7.5 mm*	*r = 8.5 mm*	*r = 10 mm*
1	*16.95*	*6.35*	*4.24*	*3.22*
2	*21*	*10.53*	*6.32*	*4.21*
3	-	*14.7*	*8.42*	*5.44*
4	-	*23.17*	*12.63*	*6.32*
5	-	*31.6*	*14.7*	*8.42*
6	-	*42.12*	*18.96*	*10.53*

The results presented in Table 1 demonstrate a direct correlation between the mixing times at various stages of convective flows and the radius of the diffusion channel. It's evident that critical radius values exist at which the transition from pure diffusion to diffusion-convective mixing takes place. Specifically, when the radius is

6.5 mm or smaller, mixing solely occurs due to diffusion. However, when the radius reaches 7.0 *mm* or larger, the formation of structured flow and observable convective drops becomes evident.

Significantly, there are substantial variations in the time required for different radius values to progress through various stages of mixing. For instance, the formation and separation of a convective droplet at a radius of 10 *mm* occurs approximately four times faster than at a radius of 7.5 *mm*.

The calculations indicate that there are specific channel sizes where the conditions for convection do not exist. At a certain diameter of the diffusion channel, a transition from "diffusion" to "convection" occurs, characterized by the emergence of a non-monotonic isoconcentration distribution of the component with the highest molecular weight, and this transition has a characteristic formation time. As the radius of the diffusion channel increases, the time at which the non-monotonic concentration distribution appears decreases, indicating an enhancement in the mixing process. Through the conducted studies, it has been established that enlarging the radius of the diffusion channel leads to a heightened drip regime within the examined mixture. Importantly, the results obtained from the numerical calculations align with the experimental data, affirming the consistency of the findings.

4. Conclusion

The numerical investigation conducted in this study delves into the emergence of mechanical equilibrium instability and subsequent structural development within the $H_2O + C_4H_{10}O - N_2$ steam-gas system. This investigation is founded on a splitting scheme that considers various physical parameters. The employed mathematical model facilitates the characterization of convective structural formation at different diffusion channel radii. The key observation is that convective instability arises primarily due to the notable curvature of the isoconcentration lines, a feature absent in pure diffusion processes. Notably, the results obtained from this numerical study align with the experimental data. Furthermore, the study reveals that an increase in the diffusion channel's radius intensifies the dripping regime within the studied mixture.

Thus, the proposed numerical algorithm adequately describes the "diffusion – convection" transition, allows us to determine the spectrum of thermophysical and geometric parameters affecting the occurrence of the convective regime.

5. Acknowledgments

The work was carried out with the financial support of the Science Committee of the Ministry of Science and Higher Education of the Republic of Kazakhstan (project AP09259248 «The emergence of concentration gravitational convection during multicomponent diffusion in stably – stratified media»).

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