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Numerical research of characteristic mixing times of isothermal three-component steam-gas systems

Multicomponent diffusion in gases is characterized by a number of effects that are not observed in binary diffusion. Analysis of existing works shows that convective instability may occur in some systems with significantly different diffusion coefficients with certain geometric and thermophysical characteristics. Stability analysis allows determining the spectrum of parameters at which a transition from a diffusive state to a convective is possible. However, this approach does not allow the researchers to investigate the dynamics of the process. Therefore, this work aims to describe emergence and evolution of convective flows in three-component systems and assess the influence of the initial composition on the occurrence of concentration gravitational convection. The main part of the work presents a mathematical model describing the occurrence of convective flows based on the splitting scheme according to physical parameters. Numerical data on the concentration fields of the gas with the highest molecular weight at various time points is obtained. It is established that curvature of the isoconcentration lines of the diffusing components can be associated with instability of the mechanical equilibrium of the system. Degree of curvature is determined by the initial concentration of components of the mixture. The obtained data can be used to determine the main characteristics of mass transfer used in calculations related to combined heat and mass transfer in a wide range of thermophysical parameters.

Keywords: gas mixtures, diffusion, convection, instability, initial composition, numerical calculation, isothermal mixing, formic acid vapors.

Introduction

Rayleigh-Benard convection is one of the few classical problems of heat and mass transfer to which there is increased attention at the present time [1]. Interest in this problem is associated with the study of the variety of flows arising in channels from a given geometry and the properties of the studied liquid or gas medium. Generalization of studies on the occurrence of gravitational convection in an inhomogeneous temperature field made it possible to develop recommendations for determining the main parameters of heat and mass transfer associated with properties of the substance under research. These recommendations allow for specific conditions to predict mixing modes. At the same time, the occurrence of instability of mechanical equilibrium and the subsequent development of convective formations depend on the direction of the vectors of density gradients and gravity [2, 3]. Occurrence and further evolution of convective perturbances are fixed, in the case when directions of the corresponding gradients do not coincide. Opposite situation assumes a stable stratification of medium and absence of convective motion.

However, for multicomponent mixtures in which several concentration flows interfere, the spread of approaches [2, 3] to describe the behavior of system can lead to distortion of the expected results associated

with mass transfer, since the features, that are absent in binary systems are unconsidered. These include, first of all, the fact of the destabilizing effect of diffusion on the development of convective flows in mixtures [4, 5]. Convective flows that arise in this case form a synergistic mixing mechanism, leading to the selective transfer of a component with specified thermophysical properties.

Experimental studies [6, 7] have shown that even in the extreme case of isothermal multicomponent mixing, convective separation effects associated with differences in the diffusion coefficients of components, pressure, and composition occur. Distinctive feature of the studies conducted in [6,7] was the condition for the implementation of multicomponent mixing, which assumed that density of the medium in the upper part of the diffusion channel was less than density of the gas localized in the lower part of the channel. Physical meaning, emergence of the convection instability and subsequent development of hydrodynamic perturbances are described in detail in [2, 3]. However, using the formalism of the Rayleigh thermal problem to describe isothermal multicomponent mixing under conditions of initial stable stratification of the mixture observed in [4–7] requires consideration of the influence of several partial concentration gradients. Importance of such an adjustment was shown in [8, 9], in which it was found that in ternary systems with different molecular weights M_i and different diffusion coefficients D_{ij} [10] there are areas of damping and increasing convective perturbances. Extreme perturbances can be the reason of structured flows in multicomponent diffusion. However, the approach developed in [8, 9] defines only the boundary of the "diffusion–convection" regime change in the area of specified thermophysical and geometric parameters. Process of the emergence of structured convective flows and their subsequent evolution has not been considered. In this regard, it is relevant to study the dynamics of the emergence of structural formations at the boundary of the "diffusion – convection" regime change during isothermal multicomponent mixing. This work presents the results of numerical studies on the mixing of triple gas mixtures in vertical diffusion channels with different initial compositions. The comparative analysis presented with obtained and experimental data.

Problem statement and numerical method

The diffusion transfer of a triple gas mixture in a flat (or cylindrical) channel is studied. The problem statement is shown in Figure 1.

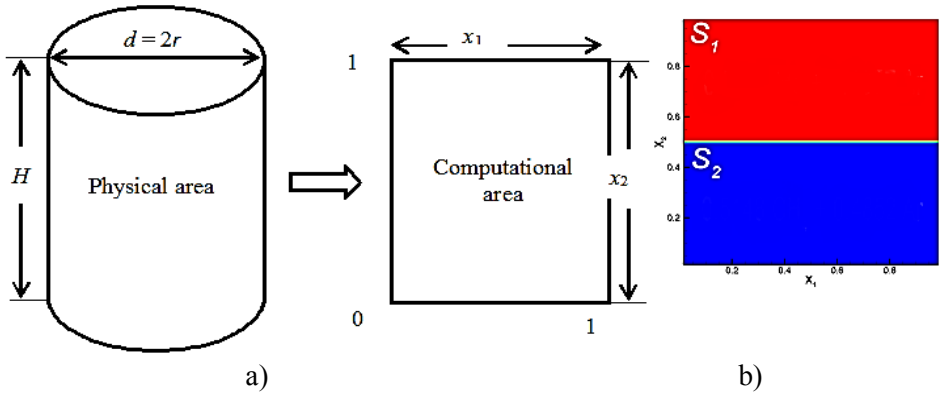

Figure 1. Model of multicomponent mass transfer:
a) mixing areas; b) placement of three-component mixtures in the diffusion cell.

The upper part of the channel S_1 contains a mixture of gases with the M_1 and M_3 molecular weights, mixture diffuses into a gas with a molecular weight M_2, which is located at the bottom of the channel S_2. At the same time, the following assumptions occur:

1. For the molecular weights of the components M_i, the condition is accepted: $M_3 > M_2 > M_1$.
2. The condition of independent diffusion assumes:

$$\sum_{i=1}^{3} j_i = 0 \text{ and } \sum_{i=1}^{3} c_i = 1$$

Multicomponent mixing is studied near the boundary of the "diffusion–convection" regime change, total transfer of components can be described by a system of hydrodynamic equations for perturbed quantities written in the Boussinesq approximation [11, 12].

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

$$\frac{\partial c_1}{\partial t}+\mathbf{v}\nabla\langle c_1\rangle=D_{11}^{*}\nabla^2 c_1+D_{12}^{*}\nabla^2 c_2, \qquad (1)$$

$$\frac{\partial c_2}{\partial t}+\mathbf{v}\nabla\langle c_2\rangle=D_{21}^{*}\nabla^2 c_1+D_{22}^{*}\nabla^2 c_2,$$

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

where $\langle c_i\rangle$ – constant average concentration value, taken as the starting point, $\vec{\gamma}$ – the unit vector, ν – coefficient of kinematic viscosity, ρ_0 – average density of the mixture. The dependence of the density mixture on the concentration is determined as follows:

$$\beta_i=\frac{1}{\rho_0}\left(\frac{\partial\rho}{\partial c_i}\right)_{p,T,c_j}, \quad \rho=\rho_0(1-\beta_1 c_1-\beta_2 c_2),$$

Diffusion complexes D_{ij}^{*} related to the coefficients of mutual diffusion D_{ij} by the relations:

$$D_{11}^{*}=\frac{D_{13}[c_1 D_{32}+(c_2+c_3)D_{12}]}{D},\ D_{12}^{*}=\frac{c_1 D_{23}+(D_{12}+D_{13})}{D},$$

$$D_{22}^{*}=\frac{D_{23}[c_2 D_{13}+(c_1+c_2)D_{12}]}{D},\ D_{21}^{*}=\frac{c_2 D_{13}+(D_{12}+D_{23})}{D},$$

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

Perturbations of the average mass and average numerical velocities are of the same order, therefore, in further relations, we will replace the perturbed quantities **v** with **u** [8]. Let us choose the characteristic measurement scales: H – linear size of the cavity, H^2/ν – time, D_{22}^{*}/H – velocity, $A_i H$ – concentration, $\rho_0\nu D_{22}^{*}/H^2$ – pressure. System of equations (1) in dimensionless quantities has the form:

$$\frac{\partial c_1}{\partial t}+\frac{1}{\mathrm{Pr}_{22}}\mathbf{u}\nabla c_1=\frac{1}{\mathrm{Pr}_{11}}\Delta c_1+\frac{1}{\mathrm{Pr}_{22}}\tau_{12}\Delta c_2,$$

$$\frac{\partial c_2}{\partial t}+\frac{1}{\mathrm{Pr}_{22}}\mathbf{v}\nabla c_2=\frac{A_1}{A_2}\frac{1}{\mathrm{Pr}_{22}}\tau_{21}\Delta c_1+\frac{1}{\mathrm{Pr}_{22}}\Delta c_2, \qquad (2)$$

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

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

where $\mathrm{Pr}_{ii}=\nu/D_{ii}^{*}$ – the diffusion Prandtl number, $\mathrm{Ra}_i=g\beta_i A_i H^4/D_{22}^{*}\nu$ – partial Rayleigh number, where A_i – dimensionless initial gradient, $\tau_{ij}=D_{ij}^{*}/D_{22}^{*}$ – parameter, which determines the relationship between the practical diffusion coefficients.

Next, we will use the algorithm written in [12]. The system of equations (2) is solved numerically by the splitting scheme according to physical parameters [13]. The two-dimensional cross-sectional area of the

cylinder area $H \times d$ in the Cartesian coordinate system is considered (Fig. 1a). Calculations were carried out in a dimensionless grid with dimensions of 128 × 128.

The first stage of the calculation assumes, that transfer of the quantity of motion is carried out only by convection and diffusion. The intermediate velocity field is found according to the Adams-Bashforth and Crank-Nicholson schemes [14].

$$\frac{\hat{\mathbf{u}}^n - \mathbf{u}^n}{\Delta t} = -\frac{1}{2}\left(3H^n - H^{n-1}\right) + \frac{1}{2}\Delta\left(\hat{\mathbf{u}}^n + \mathbf{u}^n\right) + \left(\mathrm{Ra}_1\tau_{11}c_1 + \mathrm{Ra}_2 c_2\right)\boldsymbol{\gamma}, \tag{3}$$

where $H^n = -\nabla\left(\mathbf{u}\cdot\mathbf{u}\right)^n$ – convective members, $\hat{\mathbf{u}}$ – intermediate velocity field, Δt – time step, n – is the iteration number at time t^n.

The second stage of the calculation determines the relationship of pressure with the velocity value:

$$\Delta p = \frac{\nabla\cdot\hat{\mathbf{u}}^n}{\Delta t}. \tag{4}$$

The third stage assumes recalculation of the velocity field exceptionally due to the pressure drop:

$$\frac{\mathbf{u}^{n+1} - \hat{\mathbf{u}}^n}{\Delta t} = -\nabla p. \tag{5}$$

At the fourth stage, the concentration of the mixture components is calculated:

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

where $F^n = -\nabla\left(\mathbf{u}^{\mathbf{n+1}}\cdot\mathbf{c}^{\mathbf{n}}\right)$ – convective terms.

The boundary conditions are set as follows:

$$\mathbf{u}(\boldsymbol{x}_{\boldsymbol{B}}, \tau) = 0, \ \frac{\partial c_i}{\partial \boldsymbol{n}} = 0, \quad i = 1-3, \tag{7}$$

where $n = \left(n_1, n_2\right)$ – external normal to the boundary of the computational area.

The initial conditions are written as follows:

$$\begin{gathered}
\mathbf{u}(\boldsymbol{x}, \tau) = 0, \\
c_1\left(\boldsymbol{x}, \tau = 0\right)\big|_{\boldsymbol{x}\in S_1} = X_1, \ \ c_1\left(\boldsymbol{x}, \tau = 0\right)\big|_{\boldsymbol{x}\in S_2} = 0, \\
c_2\left(\boldsymbol{x}, \tau = 0\right)\big|_{\boldsymbol{x}\in S_1} = X_2, \ c_2\left(\boldsymbol{x}, \tau = 0\right)\big|_{\boldsymbol{x}\in S_2} = 0, \\
c_3\left(\boldsymbol{x}, \tau = 0\right)\big|_{\boldsymbol{x}\in S_1} = 0, \ c_3\left(\boldsymbol{x}, \tau = 0\right)\big|_{\boldsymbol{x}\in S_2} = X_3.
\end{gathered} \tag{8}$$

where X_i – concentrations of components in the upper S_1 and lower S_2 areas.

Numerical calculation results

The numerical results (Fig. 2) illustrate the dependence of concentration on time for the heaviest component in terms of density in a triple system of formic acid vapors and water diffusing into argon at *p=0.1 MPa*, *T=298.0 K* experimentally studied in [15]. Several mixing modes are clearly visible. At the initial stage (Fig. 2a), diffusion takes place. After 0.93 s, a violation of monotony in the distribution of isoconcentration lines (Fig. 2b) is recorded. This behavior is not typical for diffusion mixing. It can be assumed that starting from this time, an instability of mechanical equilibrium occurs in the system under study.

In this case, instability is the reason of convection formation. Fig. 2c presents a convective cell formed 1.4 seconds after the start of mixing. At the final stage (Fig. 2d), the convective formation begins to move in the gravity field relative to the diffusion interface. Then, under different initial conditions (Fig. 2c), the process of structural formation begins again, i.e., in a steam-gas system, a drip mixing mode may occur.

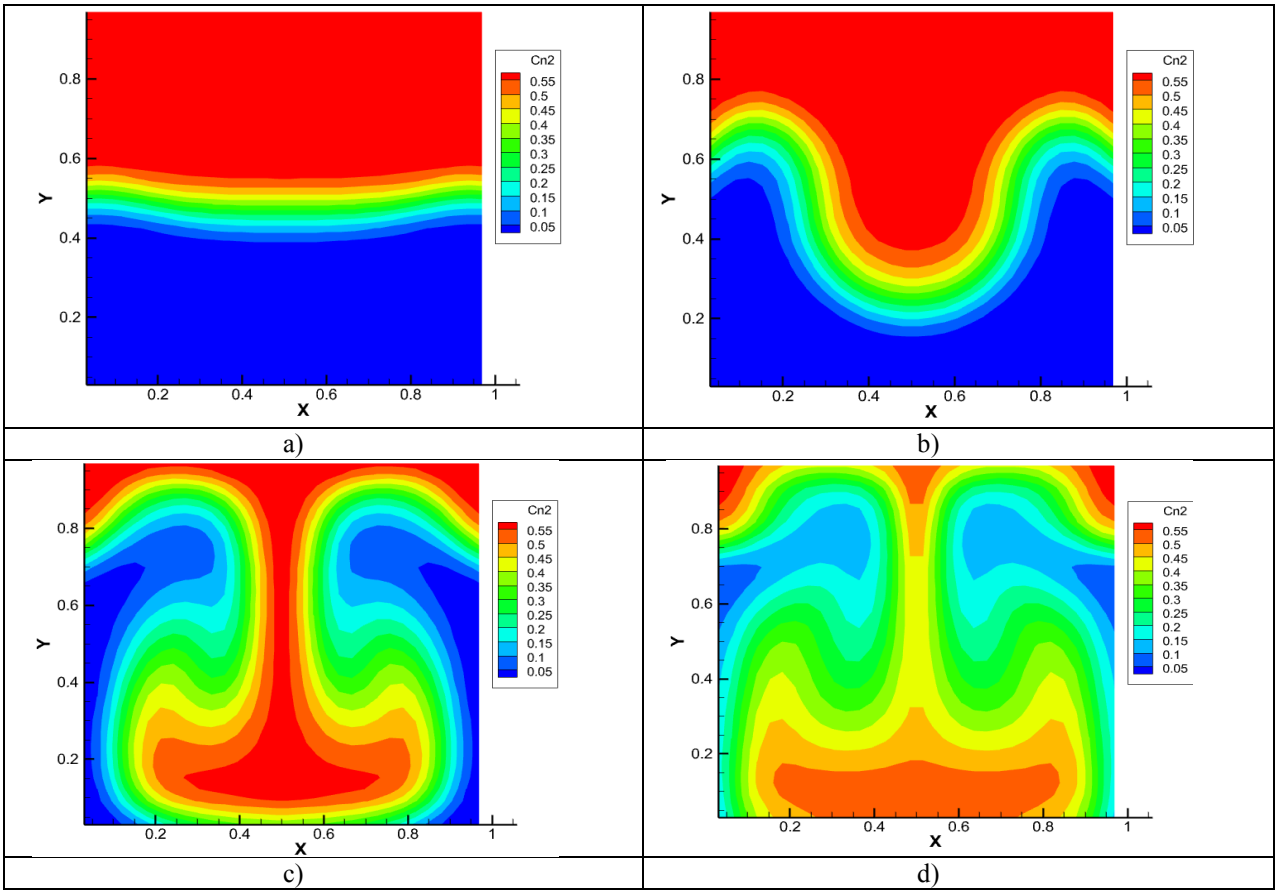

Figure 2. Isoconcentration lines of formic acid vapors for the system $0.6H_2O(1)+0.4CH_2O_2(3)-Ar(2)$ at $p=0.1MPa$, $T=298.0K$, $L=0.257m$, $r=11.2\cdot10^{-3}m$. a) t = 0.47 s b) t = 0.93 s; c) t = 1.4 s; d) t = 1.86 s.

In Table 1, for different compositions of formic acid vapors in a triple mixture $H_2O+CH_2O_2-Ar$ the characteristic mixing times describing the diffusion (t_1), the occurrence of instability of mechanical equilibrium (t_2), formation of a structural formation (t_3) and the initial displacement in the channel due to gravity (t_4) are provided.

Table 1

Characteristic mixing times for different vapor compositions of formic acid at P = 0.1 MPa, T = 298.0 K.

Molar composition CH_2O_2, mol.fraction	t_1, s	t_2, s	t_3, s	t_4, s
0.89	0.47	0.93	1.4	1.86
0.60	0.47	0.93	1.4	1.86
0.40	0.44	0.88	1.32	1.76
0.30	diffusion			

Table 1 data indicates weak dependence characteristic times of occurrence of the convection formation on the initial composition of the mixture.

Conclusions

Numerical study of the occurrence of instability of mechanical equilibrium and subsequent structure formation in the $H_2O+CH_2O_2-Ar$ parabasic system with different CH_2O_2 composition in the mixture can be carried out based on a splitting scheme according to physical parameters. Mathematical model allows describing the process of a convective structure formation for different values of the composition of a triple mixture. Definition of convective instability may be associated with a significant curvature of the isoconcentration distributions, which are absent during diffusion. Degree of curvature of concentration distributions depends on the content of the component with the highest molecular weight in the system. With certain compositions, curvature disappears and diffusion is realized in the system. It was found that when vapor content of formic acid is less than 0.3 mole fractions in the mixture corresponds to the diffusion mode of mixing.

Acknowledgments

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А.К. Жусанбаева, В. Мукамеденкызы, В.Н. Косов, А.А. Акжолова

Үшкомпонентті бугаз жүйелерінің изотермиялық араласуының сипаттамалық уақыттарын сандық зерттеу

Газдардағы көп компонентті диффузия бинарлық диффузияда байқалмайтын бірқатар әсерлермен сипатталады. Қолданыстағы жұмыстарды талдау белгілі бір геометриялық және жылуфизикалық сипаттамалары бар диффузия коэффициенттері айтарлықтай ерекшеленетін кейбір жүйелерде конвективті тұрақсыздық орын алуы мүмкін екенін көрсетеді. Тұрақтылықты талдау диффузиялық күйден конвективке ауысуға болатын параметрлер спектрін анықтауға мүмкіндік береді. Алайда, бұл тәсіл процестің динамикасын зерттеуге мүмкіндік бермейді. Сондықтан жұмыстың мақсаты үшкомпонентті жүйелердегі конвективті ағындардың пайда болуы мен эволюциясын сипаттау және концентрациялық гравитациялық конвекцияның пайда болуына бастапқы құрамның әсерін бағалау болып табылады. Жұмыстың негізгі бөлімінде физикалық параметрлер бойынша бөліну схемасы негізінде конвективті ағындардың пайда болуын сипаттайтын математикалық модель ұсынылған. Әр түрлі уақыт аралықтарында ең жоғары молекулалық салмағы бар газ концентрациясының өрістері туралы сандық мәліметтер алынды. Диффузияланушы компоненттердің изоконцентрлік сызықтарының қисықтығы жүйенің механикалық тепе-теңдігінің тұрақсыздығымен байланысты болуы мүмкін екендігі көрсетілген, ал қисықтық дәрежесі қоспа компоненттерінің бастапқы концентрациясына байланысты. Алынған мәліметтер жылуфизикалық параметрлердің кең диапазонындағы аралас жылумасса тасымалымен байланысты есептеулерде қолданылатын масса алмасудың негізгі сипаттамаларын анықтау үшін пайдаланылуы мүмкін.

Кілт сөздер: газ қоспалары, диффузия, конвекция, орнықсыздық, бастапқы құрам, сандық есептеу, изотермиялық араласу, құмырсқа қышқылының буы.

А.К. Жусанбаева, В. Мукамеденкызы, В.Н. Косов, А.А. Акжолова

Численное исследование характерных времен смешения изотермических трехкомпонентных парогазовых систем

Многокомпонентная диффузия в газах характеризуется рядом эффектов, которые не наблюдаются при бинарной диффузии. Анализ существующих работ показывает, что в некоторых системах с существенно отличающимися коэффициентами диффузии при определенных геометрических и теплофизических характеристиках может иметь место конвективная неустойчивость. Анализ на устойчивость позволяет определить спектр параметров, при которых возможен переход из диффузионного состояния в конвективное. Однако этот подход не позволяет исследовать динамику процесса. Поэтому целью работы является описание возникновения и эволюции конвективных течений в трехкомпонентных системах и оценка влияния исходного состава на возникновение концентрационной гравитационной конвекции. В основной части работы представлена математическая модель, описывающая возникновение конвективных течений на основе схемы расщепления по физическим параметрам. Получены численные данные о полях концентрации газа с наибольшим молекулярным весом в различные моменты времени. Показано, что искривление изоконцентрационных линий диффундирующих компонентов может быть связано с неустойчивостью механического равновесия системы, а степень искривления обусловлена начальной концентрацией компонентов смеси.

Ключевые слова: газовые смеси, диффузия, конвекция, неустойчивость, исходный состав, численный расчет, изотермическое смешение, пары муравьиной кислоты.

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