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ANALYSIS OF CONVECTIVE FLOWS DURING ISOTHERMAL MIXING OF THREE-COMPONENT GAS MIXTURES BY COMPUTER SIMULATION

Abstract. In multicomponent gas mixtures, there is a wide variety of mixing regimes. A key challenge is determining the boundary between diffusion and convection regimes, as well as identifying the parameters that define the convective mode, which plays a crucial role in mass transfer processes. This study presents a computational model for examining isothermal diffusion transport in three-component gas mixtures. The analysis focuses on the relationship between transport coefficients over time using a three-dimensional numerical simulation in a vertical cylindrical channel with finite dimensions. By separating the physical parameters of the studied system, the isothermal diffusion mixing process in three-component gas mixtures within cylindrical vertical channels is investigated. The results indicate that in systems where the diffusion coefficients of the components are comparable, mechanical equilibrium is unstable. In cases where there is a significant difference in diffusion coefficients, convective motion may emerge. Initially, these convective flows develop at low speeds, but they eventually evolve into structured flow patterns. The computational results align well with experimental data, supporting the proposed approach for determining the boundary between diffusion and concentration-driven convection regimes in gas mixtures.
Keywords: diffusion, convection, gases, computational modeling.

Introduction. Multicomponent gas mixtures exhibit a wide variety of mixing mechanisms, ranging from purely molecular diffusion to complex convective flows. The nature and intensity of mass transfer in such systems depend on the dominant transport processes, which can be molecular, convective, or a combination of both [1,2]. In many practical scenarios, the role of molecular diffusion is often considered secondary to convective effects. However, diffusion alone can sometimes lead to instability, disrupting the mechanical equilibrium of the system. This instability may result in the spontaneous formation of convective motion, significantly en-

hancing the overall efficiency of mass transfer [3]. Understanding these processes is essential for accurately predicting mixing behavior in various applications, such as chemical engineering, environmental sciences, and industrial gas separation technologies.

One of the critical challenges in studying mass transfer in multicomponent gas mixtures is identifying the conditions under which diffusion-driven mixing transitions into convective motion. Unlike traditional thermal convection, where fluid motion is driven by temperature gradients, concentration-driven convection can arise purely from differences in diffusion rates between gas components [4,5]. This phenomenon has been observed not only in classical Rayleigh-type stratification problems but also in cases where stable density stratification theoretically prevents large-scale motion. Experimental and numerical studies have shown that, in certain conditions, diffusion alone can generate convective flows, leading to the development of structured flow patterns that significantly alter the expected mixing dynamics.

Previous research on multicomponent gas mixing has demonstrated that convective motion can emerge due to differences in diffusion coefficients between components [6-8]. When the diffusion rates of different gases vary significantly, the resulting imbalance can trigger flow patterns resembling a sedimentation effect, where heavier and lighter components separate dynamically [9-12]. This effect introduces additional complexity in predicting and modeling mass transfer in such systems, making it essential to develop computational approaches that accurately capture the interplay between diffusion and convection [13,14].

To address these challenges, this study presents a computational model designed to investigate the diffusion-driven transport of gases under isothermal conditions. The focus is on three-component gas mixtures with varying transport properties, analyzed through a two-dimensional numerical simulation of a vertical cylindrical channel with finite dimensions [15]. The numerical results are systematically compared with experimental data to validate the approach and improve our understanding of the mechanisms governing the transition from diffusion to convective mixing [16].

Materials and methods. By exploring the interplay between diffusion and convection in multicomponent gas mixtures, this research aims to provide valuable insights into the fundamental principles of mass transfer in such systems. The findings may have significant implications for optimizing industrial processes, refining theoretical models of gas transport,

and developing more efficient strategies for controlling mixing dynamics in practical applications. The objective of this study is to conduct a numerical simulation of the transition from a purely diffusive state to a convective state in a multicomponent system, where mechanical equilibrium instability is observed. This simulation will be performed using the ANSYS Fluent software package [17]. Additionally, the study aims to compare the numerical results with experimental data. Through this analysis, we seek to gain deeper insights into the mechanisms governing the shift between diffusion and convection and to develop recommendations for a more precise characterization of mixing processes in multicomponent systems.

Problem statement and setup algorithm. Figure 1 illustrates the schematic representation of the diffusion cell (DC) utilized in the two-flask apparatus, where concentration measurements were conducted to study both diffusive and convective mixing processes [11]. The experimental method enabled the capture of shadow images of structural formations, as demonstrated in [17], allowing for a rapid identification of the mixing type and facilitating comparisons with numerical simulation results. During the numerical modeling process, it was assumed that, initially, the upper chamber of the apparatus contained a gas mixture with a chemical composition different from that of the gas in the lower chamber. Additionally, it was considered that, at the initial moment, the density of the three-component mixture decreased with height.

The model's analyzed region comprises three primary components: the upper cylinder, the lower cylinder, and the diffusion channel connecting them. This structure serves as the foundation for examining the mechanisms governing mass transfer and the transition between diffusive and convective mixing regimes. The cylinder volumes were assumed to be the same $V_{up} = V_{lower} = 227x10^{-6}m^3$ and the dimensions of the diffusion channel are as follows: $d=6.1\ x10^{-3}m$ – diameter, $L = 165x10^{-3}m$ – height.

In this study, a hybrid mesh model was employed, where triangular elements were used to discretize the upper and lower cylinders, and square elements were applied to discretize the diffusion channel. This combination ensures an efficient adaptation to the different geometrical characteristics of the system components.

The grid element size was set to 1.5 mm to achieve a detailed representation of the geometry and ensure the convergence of the numerical solution. The total number of grid elements reached 404 395, which demonstrates the model's high resolution and its ability to accurately capture the key physical processes occurring within the system.

The use of both triangular and square elements strikes a balance between modeling precision and computational efficiency. Triangular elements in the cylindrical regions offer flexibility for adapting to curved surfaces, while square elements in the channel simplify calculations in areas with simpler geometry. This approach aligns with modern practices in computational mesh design, where the choice of element type and size is guided by the complexity of the geometry and the required modeling accuracy.

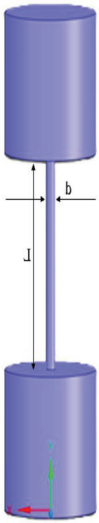

Figure 1 – 3D model of the diffusion cell and the simulation area under study

The pressure-based coupled algorithm is designed to simultaneously solve the momentum equations and the pressure-based continuity equation as a unified system. This differs from the segregated approach, where these equations are handled separately in consecutive steps. In the coupled method, momentum and pressure corrections are computed together in a single operation, while the remaining governing equations are still solved independently, as in the segregated approach [17].

Each iteration of the coupled algorithm follows a structured sequence of steps:

1. Updating Fluid Properties – The physical properties of the fluid, such as density, viscosity, and specific heat, are recalculated based on the current solution state. Additionally, turbulent viscosity (or diffusivity) is updated to maintain accurate flow dynamics.

2. Solving Momentum Equations – The momentum equations are solved sequentially using the most recent values of pressure and mass flux at cell faces.

3. Solving the Pressure Correction Equation – The pressure correction equation is formulated and solved based on the newly computed velocity field and mass flux values.

4. Applying Corrections – Face mass fluxes, pressure values, and velocity fields are corrected using the pressure correction obtained in the previous step.

5. Solving Additional Scalar Equations – If additional scalar equations are present (such as those for turbulence, energy, species transport, or radiation intensity), they are solved using the most recent solution variables.

6. Updating Source Terms – The source terms resulting from interactions between different phases are updated (for example, the source term representing the influence of discrete particles on the carrier phase).

7. Convergence Check – The solution is evaluated for convergence, ensuring numerical stability and consistency.

ANSYS Fluent offers multiple schemes for interpolating pressure values at the faces of computational cells. By default, the Second Order scheme is used for general applications. However, for simulations involving multiphase flow models such as the Mixture Model or VOF (Volume of Fluid) Model, the PRESTO! (Pressure Staggering Option) scheme is preferred to enhance accuracy.

The Second Order scheme reconstructs face pressure using a central differencing approach, ensuring improved accuracy in pressure distribution. The pressure values at cell faces are computed as:

$$P_f = \frac{1}{2}(P_{c_0} + P_{c_1}) + \frac{1}{2}(\nabla P_{c_0}\ \vec{r}_{c_0} + \nabla P_{c_1}\ \vec{r}_{c_1}) \qquad (1)$$

The SIMPLE algorithm establishes a connection between velocity and pressure corrections to ensure mass conservation, allowing for the accurate calculation of the pressure field.

If the momentum equation is solved using an initial guessed pressure field, the resulting face flux may be inaccurate and require corrections to achieve mass conservation and accurate flow predictions.

$$J_f^* = J_f^* + d_f(p_{c_0}^* - p_{c_1}^*) \tag{2}$$

To address this issue, a correction term is added to the face flux. This adjustment ensures that the corrected face flux satisfies the continuity equation, maintaining mass conservation in the flow simulation.

$$J_f = J_f^* + J_f' \tag{3}$$

The SIMPLE algorithm assumes that the corrected face flux can be expressed as a combination of the initial flux and a correction term:

$$J_f' = d_f(p_{c_0}' - p_{c_1}') \tag{4}$$

The SIMPLE algorithm inserts the flux correction equations (3)-(4) into the discrete continuity equation. This process leads to a discrete equation that solves for the pressure correction in each cell, ensuring that mass conservation is satisfied throughout the domain [17].

$$a_p p' = \sum_{nb} a_{nb} p'_{nb} + b \tag{5}$$

where the source term is the net flow rate into the cell:

$$b = \sum_{f}^{N_{faces}} J_f^* A_f \tag{6}$$

The pressure-correction equation (4) can be efficiently solved using the algebraic multigrid (AMG) method outlined in the Algebraic Multigrid (AMG) section. After obtaining the solution, corrections are applied to both the cell pressure and face flux to improve the accuracy of the flow field and ensure mass conservation.

$$p = p^* + \alpha_p p'$$

$$J_f^* = J_f^* + d_f (p'_{c0} - p'_{c1}) \quad (7)$$

The parameter α_p represents the under-relaxation factor for pressure, which helps stabilize the solution process. With this correction, the face flux ensures that the discrete continuity equation is fully satisfied at each iteration, improving convergence and solution accuracy.

When working with ANSYS Fluent, it is essential to configure all parameters to ensure both computational efficiency and a high level of accuracy. The key settings and adjustments made in this study are outlined below.

Model Setup and Physical Parameters

Physics:

• Energy equation: Enabled

• Turbulence model: Standard k-ω

• Material properties: Helium (He), Argon (Ar), and Nitrogen (N_2) added to the material database

• Species model: Species Transport

• Boundary conditions: Defined operating pressure, temperature, and density; walls specified as steel

Solution Settings

• Residuals: Set to 10^{-6} for all variables to ensure solution accuracy.

• Numerical Methods: SIMPLE (Semi-Implicit Method for Pressure-Linked Equations) scheme was chosen for pressure-velocity coupling.

• Initialization: Applied patching for pressure, temperature, and mole fractions to establish initial conditions.

• Report Definitions: Created plots of mole fractions as a function of time to track species evolution during mixing.

Post-Processing and Results Analysis

• Contour Plots: Generated mixing contours for the heavier element to visualize concentration distribution.

By carefully configuring these parameters, the simulation effectively models the mixing of multicomponent gas mixtures with high accuracy, considering the given geometry and thermophysical conditions.

The study focuses on mixing under conditions where the density gradient is positive (Figure 1). Specifically, a denser binary gas mixture is placed in the upper chamber of the diffusion cell, while a lighter gas is con-

tained in the lower flask. This configuration replicates experimental conditions observed in real laboratory setups.

All physical and chemical properties of the gases were assumed to be constant and sourced from the ANSYS 2024 R2 chemical library [17].

Since the system operates as a closed apparatus, the boundary conditions were set to impermeability, preventing any mass transfer across system boundaries. This ensures that no gas components enter or leave the system. Additionally, thermal boundary conditions were applied to maintain isothermal mixing conditions, with the walls defined as solid stainless steel surfaces. The corresponding material properties were retrieved from the ANSYS 2024 R2 library [17].

A pressure-based solver was used for all simulations. The pressure-velocity relationship was handled using the SIMPLE scheme, as previously mentioned. Computational Fluid Dynamics (CFD) equations were solved using spatial discretization methods, which have demonstrated high effectiveness in prior studies [11] (Table 1).

Table 1 – Solution methods

Quantity	Discretization
Gradient	Least Square Cell Based
Pressure	Second order
Momentum	Second Order Upwind
Turbulent Kinetic Energy	Second Order Upwind
Specific Dissipation Rate	Second Order Upwind
Pseudo Time Method	Off
Transient Formulation	Second Order Implicit

Computer modeling results. Table 2 presents a comparison between experimental and numerically simulated component concentrations under different transport assumptions. The table includes:

• *Experimental concentration values*, obtained under controlled laboratory conditions.

• *Theoretical concentration values*, calculated assuming purely diffusive transport based on the Stefan-Maxwell equations.

• *Numerical concentration values*, obtained using ANSYS Fluent, which accounts for both diffusive and convective transport mechanisms.

The results demonstrate a high degree of agreement between the experimental data and the numerical simulation results, indicating that ANSYS

Fluent accurately captures the impact of convective processes on the system. Conversely, the theoretical calculations using the diffusion-only model fail to reflect these convective effects, highlighting the limitations of the purely diffusive approach in describing real mixing dynamics.

Table 2 presents both experimental and numerically calculated component concentrations, assuming purely diffusive transport, as well as partial concentrations obtained using ANSYS Fluent for the case of combined transport. The data show that the numerical simulation results align well with the experimental findings, confirming the software's capability to account for the impact of convective flows within the system. In contrast, theoretical calculations based solely on the Stefan-Maxwell diffusion model do not capture these convective effects.

Table 2 - **Amount of diffusing gas from one flask to another as a function of experimental pressure**

0.8846 H_2 + 0.1154 CH_4 – He (*t=3 hours*, *P=4.07 MPa*, *T=295 K*)			
Method	H_2	CH_4	He
Stefan-Maxwell	0.0299	0.0012	0.0312
Experiment [11]	0.1190	0.0310	0.1500
Ansys	0.1297	0.0273	0.6497

The median deviation between the numerical simulation and experimental data across all gases is approximately 12%. However, some individual discrepancies exceed too much, particularly for the lightest component in the mixture. This may be due to non-idealities in the numerical calculations, as simulations inherently approximate physical processes. Another possible reason is the difference in molecular properties (such as molecular weight, viscosity, and diffusivity) between the actual gases and their assumed values in the computational model. Despite these inconsistencies, the overall agreement between experimental and numerical results confirms the capability of ANSYS Fluent for quantitative estimation of partial fluxes in conditions where mechanical equilibrium is disrupted. In contrast, calculations based solely on the diffusion model (using the Stefan-Maxwell approach) show errors exceeding hundreds of percent, further emphasizing the presence of convective currents in the system.

In an experimental study [11], an attempt was made to evaluate the evolution of the occurrence of convective flows when the mechanical

equilibrium of the mixture is unstable, the intensity of partial mixing of the components decreases, and the subsequent diffusion occurs. Figure 2 shows the characteristic stages of multicomponent mixing and numerical values of component concentrations.

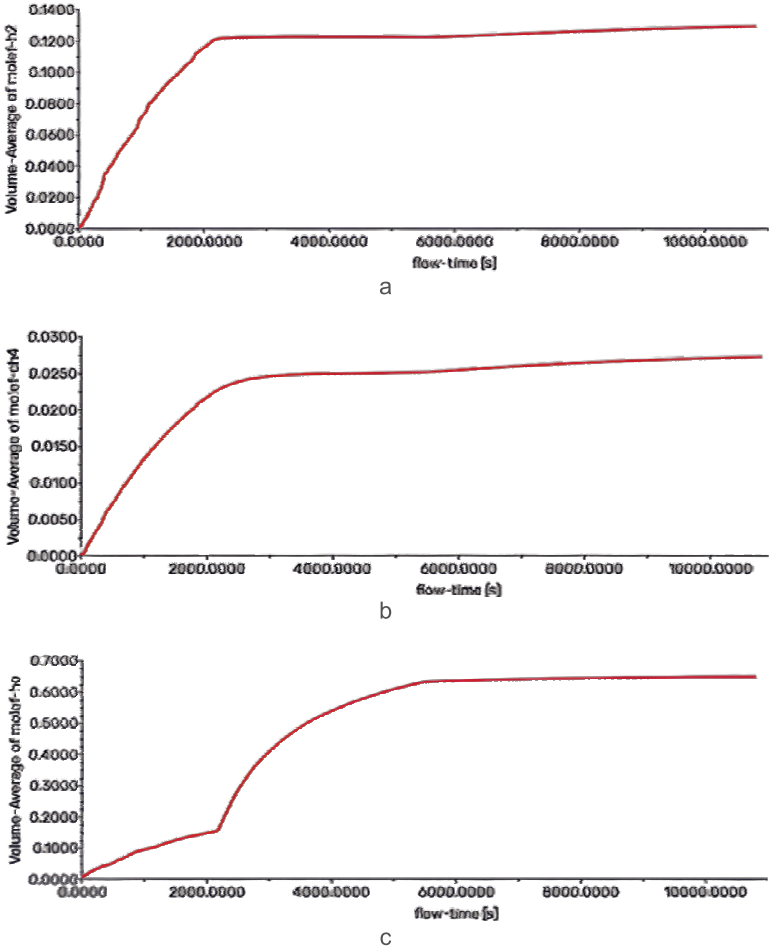

Figure 2 – Calculated values of component concentrations at different mixing times in the system 0.8846 H_2 + 0.1154 CH_4 – *He* by Ansys Fluent: a - change in the hydrogen concentration in the lower flask; b - change in the methane concentration in the lower flask;
c - change in the helium concentration in the upper flask.

For gases hydrogen and methane, one pattern is visible – an intensive transfer of the amount of gas occurs in the first 2000 seconds, then an almost constant straight line is observed, which indicates a diffusive type of mixing. This is also confirmed by experimental data. But for the helium that was originally in the lower flask, a different picture is observed. The initial stage of transfer corresponds to diffusion mixing – the first 2000 s, but then a sharp increase in the amount of gas from 2000 s to 5000 s. At the end, you can see, again, a monotonous change in concentration. There is a double transition here, first from the diffusion of convection, then back from convection to diffusion.

Conclusion. This study explored convective mixing in an isothermal helium-argon-nitrogen system through both experiments and numerical simulations using ANSYS Fluent. The results showed that the software could accurately describe the combined mass transfer processes, closely matching experimental data and proving its reliability for modeling complex multicomponent mixing. The research also provided insights into the transitions between different types of convective mixing, identifying key stages such as diffusion, the formation of convective currents, and their further development. The findings highlight the intricate nature of mass transfer and demonstrate how ANSYS Fluent effectively captures structural flow patterns, including rising and sinking convective currents. Moreover, the study confirmed that, with the right initial and boundary conditions, ANSYS Fluent can account for crucial factors like density gradients, initial composition, pressure, temperature, and the geometry of the diffusion channel. This accuracy in reproducing experimental results reinforces its potential as a valuable tool for studying such processes.

Overall, the approaches and models developed in this work can be applied to optimize various technological processes involving both convective and diffusive mass transfer, making them useful for future research and industrial applications.

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ҮШ КОМПОНЕНТТІ ГАЗ ҚОСПАЛАРЫН ИЗОТЕРМИЯЛЫҚ АРАЛАСТЫРУ КЕЗІНДЕГІ КОНВЕКТИВТІ АҒЫНДАРДЫ КОМПЬЮТЕРЛІК МОДЕЛЬДЕУ ӘДІСІМЕН ТАЛДАУ

Түйіндеме. Көп компонентті газ қоспаларында араласу режимдерінің алуан түрлілігі бар. Жұмыстың негізгі мақсаты диффузиялық және конвективті режимдер арасындағы шекараны анықтау, сондай-ақ массатасымалдау процестерінде шешуші рөл атқаратын конвективті режимді анықтайтын параметрлерді анықтау. Бұл зерттеу үшкомпонентті газ қоспаларында изотермдік диффузиялық тасымалдауды зерттеуге арналған есептеу моделін ұсынады. Талдау соңғы өлшемдегі тік цилиндрлік арнада үшөлшемді сандық модельдеуді қолдана отырып, уақыт бойынша тасымалдау коэффициенттерінің арақатынасына бағытталған. Зерттелетін жүйенің физикалық параметрлерін бөлу арқылы цилиндрлік тік арналардағы үшкомпонентті газ қоспаларының изотермдік диффузиялық араласу процесі зерттелді. Нәтижелер компоненттердің диффузиялық коэффициенттері салыстырмалы болатын жүйелерде механикалық тепе-теңдік тұрақсыз екенін көрсетеді. Диффузия коэффициенттерінде айтарлықтай айырмашылық болған жағдайда конвективті қоз-

ғалыс пайда болуы мүмкін. Бастапқыда бұл конвективті ағындар төмен жылдамдықпен дамиды, бірақ уақыт өте келе олар құрылымдық ағындарға айналады. Есептеу нәтижелері газ қоспаларындағы концентрацияға байланысты диффузия мен конвекция режимдері арасындағы шекараны анықтауға ұсынылған тәсілді қолдана отырып, эксперименттік деректермен жақсы үйлеседі.
Түйінді сөздер: диффузия, конвекция, газдар, компьютерлік модельдеу.

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АНАЛИЗ КОНВЕКТИВНЫХ ПОТОКОВ ПРИ ИЗОТЕРМИЧЕСКОМ ПЕРЕМЕШИВАНИИ ТРЕХКОМПОНЕНТНЫХ ГАЗОВЫХ СМЕСЕЙ МЕТОДОМ КОМПЬЮТЕРНОГО МОДЕЛИРОВАНИЯ

Аннотация. В многокомпонентных газовых смесях существует большое разнообразие режимов перемешивания. Ключевой задачей является определение границы между диффузионным и конвективным режимами, а также определение параметров, определяющих конвективный режим, который играет решающую роль в процессах массопереноса. В данном исследовании представлена вычислительная модель для изучения изотермического диффузионного переноса в трехкомпонентных газовых смесях. Анализ фокусируется на соотношении коэффициентов переноса с течением времени с использованием трехмерного численного моделирования в вертикальном цилиндрическом канале конечных размеров. Путем разделения физических параметров исследуемой системы исследован процесс изотермического диффузионного перемешивания трехкомпонентных газовых смесей в цилиндрических вертикальных каналах. Результаты показывают, что в системах, где коэффициенты диффузии компонентов сопоставимы, механическое равновесие неустойчиво. В тех случаях, когда существует значительная разница в коэффициентах диффузии, может возникнуть конвективное движение. Первоначально эти конвективные потоки развиваются с низкими скоростями, но со временем они превращаются в структурированные структуры. Результаты расчетов хорошо согласуются с экспериментальными данными, подтверждая предложенный подход к определению границы между режимами диффузии и конвекции, обусловленной концентрацией, в газовых смесях.
Ключевые слова: диффузия, конвекция, газы, компьютерное моделирование.

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