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Features of diffusion and convective mixing in mixtures containing hydrocarbons

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Abstract. Diffusion mixing in mixtures of C_3H_8 + CO_2 - N_2O, Ne + C_3H_8 - N_2O + C_3H_8, N_2O + He - C_3H_8 + CH_4 at different pressures has been studied experimentally. It has been found that at a certain pressure in the mixture there are convective flows distorting the expected diffusion transfer. The transition parameters from diffusion to convective mixing type can be determined in terms of stability theory. The numerical results are compared with experimental data.

1. Introduction

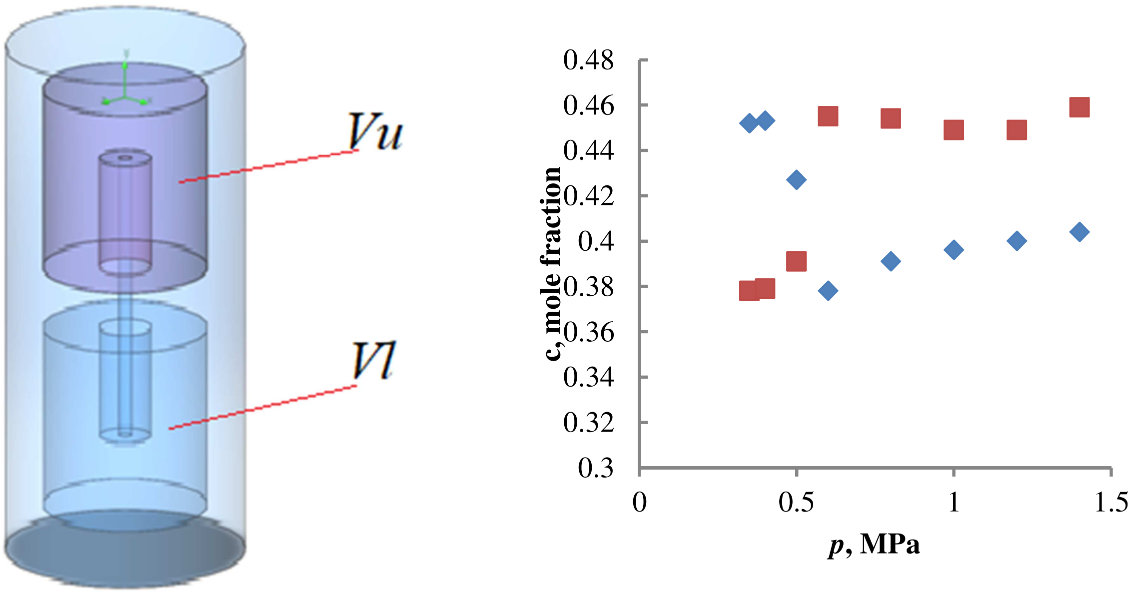
In the study of thermal diffusion effect in three-component gas mixtures, Miller and Mason discover the oscillatory nature of the temperature change in anticipation of its monotonic increase [1, 2]. Visualization of multicomponent mixing in this case registered convective structured flows not typical for diffusion [2]. Experiments in [3] show that during mixing in triple mixtures in a certain range of parameters (pressure, temperature, geometric characteristics of the diffusion channel) there is an oscillatory mode of change in the concentration of componentsin process of time. The partial flows of the components measured for this case were significantly superior to those calculated under the diffusion assumption. Further experimental study of multicomponent transport showed the presence of a synergistic increase in the intensity of partial mixing of components caused by the superposition of convective flows on the proper diffusion [4, 5]. In multicomponent diffusion the causes of occurrence of concentration-driven gravitational convection are associated with the instability of the mechanical equilibrium of the mixture and are described in [6, 7]. These and subsequent studies [8-10] develop approaches to determine the boundary parameters related to the change of diffusion and convective mixing modes.

The peculiar properties studied in [3-10] can also appear in gas mixtures containing technically important hydrocarbon components. Investigation of the peculiar properties of diffusion and convective mixing of such systems is important, as it allows obtaining information about the features of the transfer of components included in natural gas mixtures. In this work isothermal diffusion in triple gas mixtures containing propane at different pressures is investigated experimentally and computationally. As part of the stability analysis, the possibility of transition of the system from the diffusion to the convective state is considered.

2. Experimental results

Multicomponent mixtures of 0.220 C_3H_8 + 0.780 CO_2 – N_2O, 0.570 He + 0.430 C_3H_8 - 0.597 N_2O + 0.403 C_3H_8, 0.425C3H8 + 0.575 He – 0.426 N_2O + 0.574 CH_4 were selected for the investigation. Here and further we agree that the numbers before the chemical element correspond to the initial concentration of the components in the mixture in molar fractions. Partial transfer of components was studied on an experimental device implementing the method of two flasks connected by a vertical channel [11, 12]. The experiments were conducted in the pressure range from 0.4 to 1.8 MPa at a temperature of 298.0 K. The scheme of the experimental stand and the operational procedure were described in detail in [11-13]. The schematic representation of the diffusion cell is given in figure 1, which clarifies the features of the experiment. The geometrical characteristics of the apparatus are as follows: $V_u = V_l = (55.5 \pm 0.5)\cdot 10^{-6}$ m^3 – volumes of the upper and lower flasks, and $r = (2.00 \pm 0.05)\cdot 10^{-3}$ m and $L = (64.00 \pm 0.05)\cdot 10^{-3}$ m – radius and length of the connecting capillary. The duration of the experiment was 2-4 hours, which allowed obtaining information about the characteristic features of mass transfer.

The methodology of the experiment involved the following procedures. The upper V_u and lower V_l flasks of the apparatus were filled with the investigated gas mixtures up to the experimental pressure. Then, with the help of a special device, the channel connecting the flasks was opened and the start time of the mixing process was fixed. At the end of the experiment, the channel was blocked and the end time of mixing was recorded. The analysis of gas mixtures from each flask was conducted with a chromatograph with an error in the concentration measurement of up to ± 0.5%. In all experiments, the density of the mixture located in the upper flask was less (or not superior) to the density of the gas mixture located in the lower flask.



(a) (b)

Figure1. Experimental results on the triple mixtures mixing containing hydrocarbon components at different pressures: (a) diffusion cell; (b) the system 0.570 He + 0.430 C_3H_8 – 0.597 N_2O + 0.403 C_3H_8, T = 298.0 K. Concentrations of diluent gas C_3H_8 at different pressures, ■ – lower flask, ♦ – upper flask.

In the figure 1(b) the data for propane in the upper and lower flasks of the diffusion cell are given. Until the pressure p = 0.4 MPa, the concentration of evenly distributed propane at the initial time in the flasks of the apparatus, practically remains constant. This corresponds to the diffusion process of mixing gases. Further increase in pressure leads to the increase of the concentration of propane in the lower flask of the apparatus. In the upper flask, the concentration of the diluent gas decreases accordingly, which indicates the occurrence of convection. The mixing intensity in this case has complex nature, which is not typical for diffusion. A similar pressure dependence is observed for propane and nitrogen dioxide in the lower and upper flasks of the diffusion cell, respectively.

The dependence of propane mixing intensity on pressure, represented in figure 2a, also shows a complex nonlinear character. The peculiarity of mixing components in a mixture of 0.220 C_3H_8 + 0.780 CO_2 – N_2O is determined by the fact that the densities of the main diffusing components are almost the same. Therefore, the non-monotonic dependence of propane concentration on pressure is explained by the occurrence of concentration gravitational convection in the system caused by the instability of the mechanical equilibrium of the system.

3. Analysis on the stability

Verification of the experimental data with the calculated data is conducted in the framework of the analysis for the stability of the mechanical equilibrium of the isothermal triple gas mixture given in [14]. The motion of the isothermal triple gas mixture in the "diffusion – convection" modes boundary change is described by the total system of equations of fluid dynamics, including Navier-Stokes equations, conservation of particles' number and components, which for perturbed dimensionless quantities has the form:

$$\begin{gathered}
P_{22}\frac{\partial c_1}{\partial t}-(\mathbf{u}\mathbf{e}_z)=\tau_{11}\nabla^2 c_1+\frac{A_2}{A_1}\tau_{12}\nabla^2 c_2\\
P_{22}\frac{\partial c_2}{\partial t}-(\mathbf{u}\mathbf{e}_z)=\frac{A_1}{A_2}\tau_{21}\nabla^2 c_1+\nabla^2 c_2\\
\frac{\partial \mathbf{u}}{\partial t}=-\nabla p+\nabla^2\mathbf{u}+(R_1\tau_{11}c_1+R_2c_2)\mathbf{e}_z\\
\mathrm{div}\mathbf{u}=0
\end{gathered}\tag{1}$$

where u is the average mass velocity; e_z is the unit vector in the direction of the z axis; $P_{ii}=\nu/D_{ii}^*$ is the Prandtl diffusion number; $R_i=g\beta_i A_i d^4/\nu D_{ii}^*$ is the Rayleigh partial number; $\tau_{ij}=D_{ij}^*/D_{22}^*$ are the parameters determining the relationship between practical diffusion coefficients; $\beta_i=-\frac{1}{\rho_0}\left(\frac{\partial\rho}{\partial c_i}\right)_{p,T}$; $A_i\mathbf{e}_z=-\nabla c_{i0}$ (index 0 refers to the mean values), ci is the concentration of the i - th component. Other designations are generally accepted.

The stability problem (1) is reduced to determining the sequence of critical relations for Rayleigh partial numbers Ra and critical motions [15]. For odd perturbations, solution (1) has the following form:

$$u=\left(\frac{\mathrm{sh}\gamma x}{\mathrm{sh}\gamma}-\frac{\sin\gamma x}{\sin\gamma}\right)\exp[-\lambda t]$$

$$c_i=-\frac{K_i}{\gamma^4}\left(\frac{\mathrm{sh}\gamma x}{\mathrm{sh}\gamma}+\frac{\sin\gamma x}{\sin\gamma}\right)\exp[-\lambda t]$$

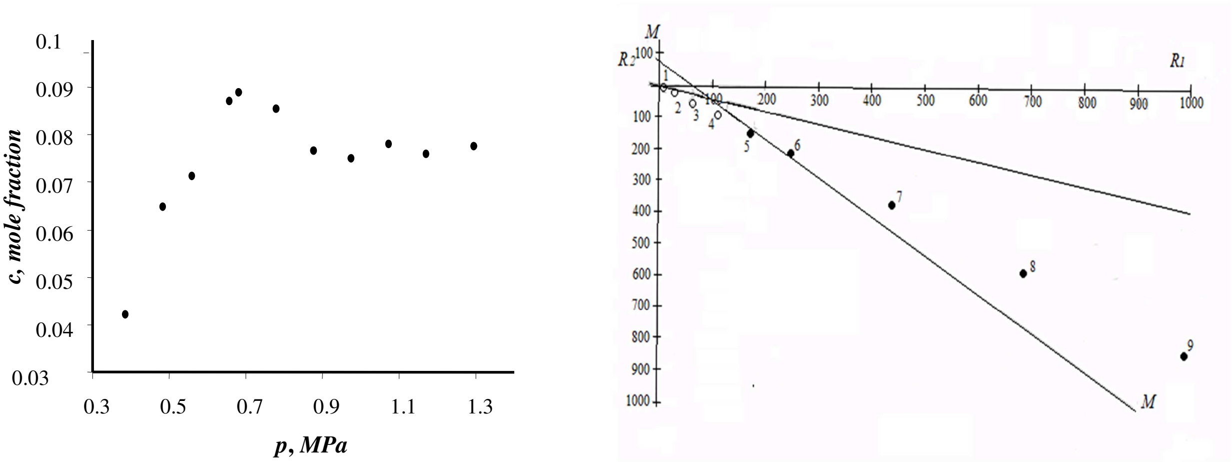
$$K_1 = \frac{\left(1 - \frac{A_2}{A_1}\tau_{12}\right)}{\tau_{11} - \tau_{12}\tau_{21}},\; K_2 = \frac{\left(\tau_{11} - \frac{A_2}{A_1}\tau_{21}\right)}{\tau_{11} - \tau_{12}\tau_{21}}, \tag{2}$$

where $i = 1{,}2$, $\gamma = (K_1\mathrm{Ra}_1\tau_{11} + K_2\mathrm{Ra}_2)^{1/4}$.

Following [16], we define the loss of stability of the gas mixture by the fact that the decrements λ of some perturbations at a certain critical number Ra*, change sign, and the perturbations themselves, fading at Ra < Ra*, begin to increase at Ra > Ra*. The zero reversal of the decrement λ determines the condition under which the perturbation is neutral, i.e. it neither increases nor decreases. This condition characterizes the stability boundary of the mechanical equilibrium with respect to the perturbation. Substituting (2) into the system of equations (1), excluding the amplitudes of concentrations and velocities in terms of Rayleigh numbers, we obtain a boundary relation defining the change of "diffusion-convection" modes in the form:

$$\tau_{11}\left(1 - \frac{A_2}{A_1}\tau_{12}\right)\mathrm{Ra}_1 + \left(\tau_{11} - \frac{A_1}{A_2}\tau_{21}\right)\mathrm{Ra}_2 = \gamma^4\left(\tau_{11} - \tau_{12}\tau_{21}\right) \tag{3}$$

where γ are the numbers defining the spectrum of critical Rayleigh numbers.



(a) (b)

Figure 2. (a) Dependence of the amount of diffused propane on the pressure in the system 0.220 C_3H_8 + 0.780 CO_2 – N_2O, T = 298.0 K. (b) diffusion and convection region for the system 0.570 He + 0.430C_3H_8 – 0.597 N_2O + 0.403 C_3H_8, T = 298.0 K: MM-boundary line; $\Delta\rho$ – zero density gradient line; 1-9 – experimental data at different values of p: 1 – 0.49; 2 – 0.98; 3 – 1.47; 4 – 2.45; 5 – 2.94; 6 – 3.03; 7 – 3.92; 8 – 4.90; 9 – 5.89 MPa.

Equation (3) on the plane (Ra_1, Ra_2) defines a boundary line separating the regions of damping (diffusion) and increasing (concentration convection) perturbations. Equating the density gradient to zero, and using the definition of Rayleigh partial numbers (1) we obtain the corresponding equation of the line on the plane (Ra_1, Ra_2):

$$\tau_{11}\mathrm{Ra}_1 = -\mathrm{Ra}_2 \tag{4}$$

The relative position of boundary lines (3) and (4) shows the existence of a region (the sector between lines 1 and 2 in figure (2b)), when the state of the mixture is unstable, although the density at the top of the diffusion cell is less than at the bottom, which, at first glance, corresponds only to the diffusion mixing type. A similar situation was observed for the non-isothermal case in a binary liquid mixture [16].

We restore the experimental data corresponding to the system 0.570 He (1) + 0.430 C_3H_8 (2) – 0.597 N_2O (3) + 0.403 C_3H_8 (2) (figure 2(b)) through the Rayleigh partial numbers and display them on the coordinate plane (Ra_1, Ra_2). The numbers after the chemical element determine the component number in the calculation scheme (1) – (4). From the experiment it is known what type of mixing takes place: stable (diffusion) or unstable (convection). The points corresponding to the unstable mode will be denoted by the signs ●, and the diffusion will be determined by the icons ○. The set of such points on the plane of Rayleigh numbers (figure 2(b)) determines the transition from the diffusion region to the convective one through the boundary line of stability (3). From figure 2b it can be seen that, starting with a pressure of 2.45 MPa (point 4), convective mixing conditions are realized in the system. A further increase in pressure leads to an increase in Rayleigh partial numbers, which corresponds to an increase in the intensity of the unstable mode. Comparison of experimental data, shown in figure 1b, with the results of the calculation presented in figure 2(b), indicates a satisfactory agreement between them. Similar cartograms in terms of Rayleigh diffusion numbers can be reconstructed for other mixtures containing hydrocarbon components

4. Conclusion

The experimental investigation of diffusion mixing in gas mixtures containing hydrocarbon components at different pressures has proved the occurrence of the convective flows. The resulting convective instability of the system significantly intensifies the total mass transfer. Pressure influences the transition of the system from the diffusion to the convective region. Comparison of experimental data with the results of theoretical calculation has shown a satisfactory agreement between the theory and experiment in determining the diffusion and convection regions.

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