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RAYLEIGH NUMBERS FOR CRITICAL INSTABILITY LEVELS IN THE CASE OF ISOTHERMAL THREE-COMPONENT DIFFUSION IN A CYLINDRICAL CHANNEL

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Within linear analysis of stability, a relation of wave numbers (perturbation modes) that determine various kinds of convective flows in a cylindrical channel with Reyleigh numbers is found for ideal three-component gas mixtures.

Introduction. Diverse convective forms of motion induced by a temperature gradient manifest themselves in numerous natural phenomena and in various kinds of chemical engineering processes. The main feature of free convection are described rather thoroughly for the case of a single-component liquid [1, 2]. In spite of the long history of research in this phenomenon (for example, see [3, 4]), it is still one of the most urgent and complicated problems of contemporary physics of continua that at present still attract the attention of experimentors and theoreticians [5-7].

Description of existing convection modes in which arising turbulent structures dominate is difficult in itself. The situation becomes much more complicated in studies of multicomponent media in which diffusion is of basic importance. The classic experiment with Benard convection cells in a single-component homophase medium (liquid) distinctly showed one of the key difficulties in studies of the effect. On the one hand, there exists high reproducibility of cellular structures that are governed by certain periodic laws; on the other, in the case of convective motion forms occurring in the gravity field (for example, rotation), a structured liquid material disturbs this periodicity and the process becomes almost stochastic [8, 9]. In this case dualism of the laws governing the appearance and existence of Benard cells is exhibited quite clearly. This specific form of ordering (self-organization) of the system occurring far from equilibrium gives rise to a new physical system, a dissipative structure that in its evolution is characterized by certain forms of disturbance of symmetry, which is reflected in the presence of bifurcations [9, 10].

A similar approach can be also applied to the study of diffusion mass transfer in multicomponent gas mixtures. In the limiting isothermal case for some of them, due to differences in the diffusion coefficients of the component (provided satisfaction of certain parameters such as pressure, temperature, initial composition of the mixture, etc.), conditions appear for the existence of concentration-induced convection caused by Archimedean forces [11, 12]. The structural form of convective flows of components in the mixture is very complicated and unpredictible, which is revealed by multiple visual and katharometric studies [11, 13]. Determination of relations governing their appearance seems important even in a linear approximation by introduction of the number of structural components (periodicity numbers) [14]. In the present work, within linear stability theory, a relation is found between diffusion Raylaigh numbers and periodicity numbers whose values simulate a broad diversity of structural forms of motion in a cylindrical channel, both along and across it.

Linear Stability Analysis. At a constant pressure under isothermal conditions, the state of an ideal three-component gas mixture is described by the system of equations [14]:

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$$\rho\left[\frac{\partial \mathbf{u}}{\partial t}+(\mathbf{u}\nabla)\,\mathbf{u}\right]=-\nabla p+\eta\nabla^2\mathbf{u}+\rho g\boldsymbol{\gamma}\,,\ \ \operatorname{div}(\rho\mathbf{u})=0\,;$$

$$\frac{\partial \rho_i}{\partial t}+(\nabla\rho_i\mathbf{u})=\operatorname{div}\mathbf{j}_i\,,\ \ \mathbf{j}_i=-\rho\sum_{j=1}^{3}D_{ij}\nabla c_j\,,\ \ i=\overline{1,3}\,. \tag{1}$$

With the condition of independent diffusion and the imposed constraints

$$\sum_{i=1}^{3}\mathbf{j}_i=0\,,\ \ \sum_{i=1}^{3}c_i=1\,,$$

and in view of the smallness of the diffusion cross-coefficients in comparison with the diagonal ones $D_{ij} << D_{ii}$ [15], the system of equations (1) is transformed as follows

$$\frac{\partial c_1}{\partial t}+\mathbf{u}\nabla c_1=D_1\nabla^2c_1\,,\ \ \frac{\partial c_2}{\partial t}+\mathbf{u}\nabla c_2=D_2\nabla^2c_2\,,$$

$$\frac{\partial \mathbf{u}}{\partial t}+(\mathbf{u}\nabla)\,\mathbf{u}=-\frac{1}{\rho_0}\nabla p+\nu\nabla^2\mathbf{u}+g\frac{\rho}{\rho_0}\boldsymbol{\gamma}\,, \tag{2}$$

where D_i is the partial diffusion coefficient (PDC) of the i-th component. Equations (2) are linearized in a Boussinesq approximation [1, 14] with the use of perturbations for the following quantities: $c_i=c_{0i}+c_i'$, $p=p_0+p'$, where c_{0i} and p_0 are constant average values taken as a reference point. The perturbed quantities c_i' and p' are small, and deviations of the density ρ' induced by them are insignificant in comparison with the average ρ_0.

The equation of state has the form

$$\rho=\rho_0\left(1-\sum_{i=1}^{3}\beta_i c_i'\right),\ \ \beta_i=-\frac{1}{\rho}\left(\frac{\partial\rho}{\partial c_i}\right)_{p,T}\,,$$

$$\sum_{i=1}^{3}\beta_i c_i'=\xi_{3i}\beta_i c_i'+\xi_{3j}\beta_j c_j'\,,\ \ \xi_{3i}=1-\frac{\beta_3}{\beta_i}\cong 1-\frac{\delta c_{03}}{\delta c_{0i}}\,,\ \ i=1,2\,, \tag{3}$$

where δc_{0i} is the concentration difference of the i-th component.

Having made Eqs. (2) and (3) dimensionless relative to the scale of length d, time d^2/ν, velocity D_i/d, concentration $B_i d$, pressure $\rho_0\nu D_i/d^2$, after some manipulations, neglecting values of the second order of smallness, we obtain the equations for perturbations (primes are omitted)

$$\frac{\partial \mathbf{u}}{\partial t}=-\nabla p+\nabla^2\mathbf{u}+(\overline{\mathrm{R}}_1c_1+\overline{\mathrm{R}}_2c_2)\boldsymbol{\gamma}\,,\ \ \overline{\mathrm{R}}_i=\tau_i\xi_{3i}\mathrm{R}_i\,,$$

$$\frac{\partial c_i}{\partial t}-(\mathbf{u}\boldsymbol{\gamma})=\frac{1}{\mathrm{Pr}_i}\nabla^2c_i \tag{4}$$

We introduce cylindrical coordinates (r,φ,z) with the axis z directed upward along the cylindrical axis. Then, in the steady-state case, system of equations (4) becomes

$$\Delta u+\overline{\mathrm{R}}_1c_1+\overline{\mathrm{R}}_2c_2=0\,,\ \ \frac{1}{\mathrm{Pr}_i}\Delta c_i+u=0\,,$$

$$\Delta=\frac{\partial^2}{\partial r^2}+\frac{1}{r}\frac{\partial}{\partial r}+\frac{1}{r^2}\frac{\partial^2}{\partial\varphi^2}+d^2\frac{\partial^2}{\partial z^2}\,, \tag{5}$$

where $d = r/L$ is the inverse of the diameter; r and L are the radius and length of the cylinder, respectively.

Boundary conditions are

$$\frac{\partial u}{\partial r} = \frac{\partial c_i}{\partial r} = 0 \quad \text{at} \quad r = 1\,,$$
$$\frac{\partial u}{\partial z} = \frac{\partial c_i}{\partial z} = 0 \quad \text{at} \quad z = \pm 1\,, \tag{6}$$

u and c_i are finite values at $r = z = 0$ and arbitrary φ.

A solution of (5) will be sought in the form:

$$u\,(r, \varphi, z) = U\,(r) \cos\,(n\varphi)\,\mathrm{ch}\,(jm\pi z)\,,$$
$$c_i\,(r, \varphi, z) = C_i\,(r) \cos\,(n\varphi) \sin\,(m\pi z)\,, \quad j = \sqrt{-1}\,, \tag{7}$$

where $U(r)$ and $C_i(r)$ are arbitrary constants. Substituting (7) into (5), with (6) taken into consideration, we obtain the following system of equations

$$\Delta_1 u + \overline{\mathrm{R}}_1 c_1 + \overline{\mathrm{R}}_2 c_2 = 0\,, \quad \Delta_2 c_i + \mathrm{Pr}_i u = 0\,,$$
$$\Delta_1 = \Delta_r + \lambda^2\,, \quad \Delta_2 = \Delta_r - \lambda^2\,, \quad \Delta_r = \frac{\partial^2}{\partial r^2} + \frac{1}{r}\frac{\partial}{\partial r} - \frac{n^2}{r^2}\,, \tag{8}$$

where $\lambda = m\pi d$ is the wave complex. Multiplying the equation for velocity in (8) by the operator Δ_2 and using equations for concentrations with account of the fact that

$$\Delta_1 \Delta_2 = (\Delta_r + \lambda^2)\,(\Delta_r - \lambda^2) = \Delta_r \Delta_r - \lambda^4\,,$$

we obtain the biharmonic equation in velocity

$$E^2 u = 0\,, \quad E^2 = \Delta_r\,\Delta_r - \lambda^4 - \overline{\mathrm{R}}_1 \mathrm{Pr}_1 - \overline{\mathrm{R}}_2 \mathrm{Pr}_2\,. \tag{9}$$

The general solution of (9) has the form

$$u = A_1 Y_n\,(Br) + A_2 I_n\,(Br)\,, \tag{10}$$

where Y_n and I_n are first-kind Bessel functions; A_1 and A_2 are constants; B is a constant depending on R. It should be noted that as in the case of a single-phase liquid (Ostroumov's problem [14], in the case of a diametrically antisymmetric structural mixture flow in the azimuth direction ($n = 1$), problem (9)-(10) gives the critical Rayleigh number

$$\mathrm{R}^* = 67.95\,. \tag{11}$$

In the present formulation for a three-component mixture, Eq. (11) has a slightly modified form

$$\mathrm{R}^* = \lambda^4 + \overline{\mathrm{R}}_1\,\mathrm{Pr}_1 + \overline{\mathrm{R}}_2\,\mathrm{Pr}_2\,. \tag{12}$$

Using the conditions of closeness and independent diffusion, after some manipulations we obtain the partial critical Rayleigh number of the i-th component

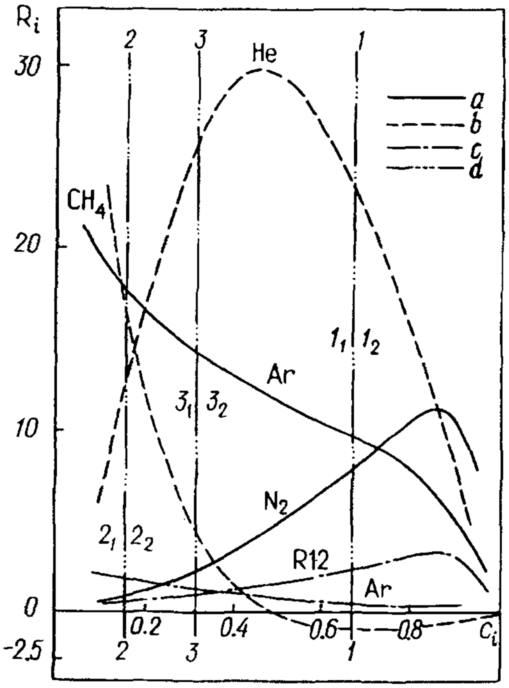


Fig. 1. Plot of critical Rayleigh number versus concentration of heavy component in mixture. Systems: 1) He + Ar – N_2, 2) H_2 + CH_4 – He, 3) H_2 + R12 – Ar, lines: a) Ar and N_2 for system 1, b) CH_4 and He for system 2; c) R12 and Ar for 3, d) of equal density for systems presented; 1_1, 1_2, 2_1, 2_2, 3_1, 3_2) of diffusion and diffusion mixing, respectively. $p = 0.1$ MPa, $T =$ 298 K, $L = 7 \cdot 10^{-2}$ m, $d = 4 \cdot 10^{-3}$ m. c_i, mole fractions.

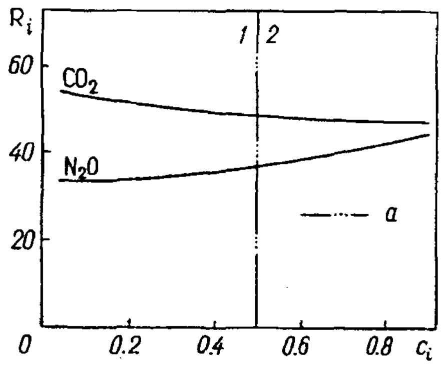
$$\mathrm{R}_i = \frac{67.95 - m^4 \pi^4 d^4}{\tau_i \left[\xi_{3i} \mathrm{Pr}_i - \xi_{3j} \mathrm{Pr}_j \sigma_{ij}\right] \left(\dfrac{\tau_i - 1}{\tau_j - 1}\right)}, \quad \sigma_{ij} = \frac{\delta c_{0i}}{\delta c_{0j}}. \tag{13}$$

of boundary-value stability problem (6)-(13) for ideal isothermal three-component gas mixtures. First of all, we failed to reduce the problem to one integral concentration Rayleigh number. For each of the components, its own critical Rayleigh number is obtained with a different behavior as a function of thermodynamic quantities, which suggests structured properties of the hydrodynamic flows of each of the components. As the equation contains two periodicity numbers (in the asimuthal n and vertical m directions), it is possible to model a flow profile that can be identified with the number of convective structures that are formed in a cylindrical channel. Comparing the Rayleigh numbers of the components at different m and n (the other parameters are invariable), we easily obtain the following dependence

$$\frac{m_1}{m_2} = \frac{2r_1}{2r_2},$$

which shows that the larger the diameter of the diffusion channel, the smaller the number of structured elements that form the flow along the vertical of the cylinder required for realization of convective mass transfer. As the diameter decreases, the density of the number of structures increases relative to the cross-section of the channel, and when the diameter is lower than the threshold value, development of perturbations in the system is impossible, which leads to stable diffusion transfer.

Now, we characterize the nature of convective flow in accordance with equation (13) with the following experimental ideal gas mixtures studied as examples: H_2 + CH_4 – He, He + Ar – N_2, H_2 + R12 – Ar, He + R12 – Ar, CH_4 + Ar – N_2 [5, 16, 17]. In the first four systems, in the order of increasing ratios of molecular



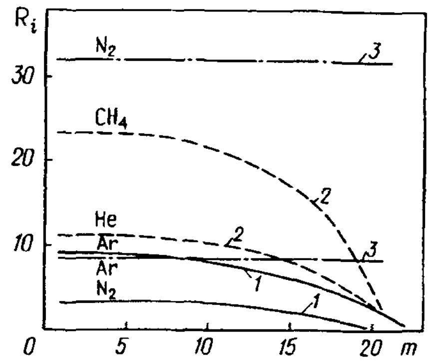


Fig. 2. Plot of critical Rayleigh number versus concentration of CO_2 in mixture for system $C_3H_8 + CO_2 - N_2O$: a) line of equal density; 1 and 2) regions of diffusion and diffusion mixing, respectively. $p = 0.1$ MPa, $T = 298$ K, $L = 7\cdot10^{-2}$ m, $d = 4\cdot10^{-3}$ m.

Fig. 3. Critical Rayleigh numbers of components with different perturbation modes. Systems: 1) $0.5He + 0.5Ar - N_2$; 2) $0.9H_2 + 0.1CH_4 - He$; 3) $0.9CH_4 + 0.1Ar - N_2$. $p = 0.1$ MPa, $T = 298$ K, $L = 7\cdot10^{-2}$ m, $d = 2r = 4\cdot10^{-3}$ m.

masses in the binary systems, diffusion instability is observed at a certain percentage of the heaviest component. However, the conditions of unstable diffusion transfer are different in different systems (see Fig. 1).

For the system $H_2 + CH_4 - He$, typical concentrations are found, at which critical Rayleigh numbers of the components are maximal (minimal), which suggests a change of instability forms. At a low ("trace") content of methane in the mixture (up to 0.1 mole fraction) ordinary diffusion of hydrogen into helium is observed, which is indicated by relative growth of the critical Rayleigh number for the heaviest component in the mixture. A further increase in its concentration results in diffusion mixing, which can be inferred from a decrease in the critical Rayleigh number R. Starting from $c_{CH_4} \cong 0.7$ mole fraction, light (the most mobile) components of the mixture make the main contribution to development of instability, resulting in a decrease in the critical Rayleigh number to its value for helium. For the system $He + Ar - N_2$, as the concentration of argon in the mixture increases, its critical Rayleigh number is found to decrease, which indicates an increase in the intensity of the unsteady process (which was shown experimentally in [11]). However, relative to nitrogen, the system is stabilized to some extent (since $D_{N_2-Ar} < D_{N_2-He}$) and the unstable process induced by the action of Archimedian forces can appear at a certain percentage of argon in the three-component system. For the mixture $H_2 + R12 - Ar$, critical Rayleigh numbers of the main diffusing components remain almost invariable (the slight trend of increasing R_{Ar} and decreasing R12 can be explained by reasons similar to those suggested for the previous mixture). Meanwhile, in the diffusion region, for these gases the Rayleigh numbers are much lower ($0.2 < R_{Ar}, R_{R12} < 2.8$) as compared with the previous systems. This indicates that the mixtures that contain R12 are potentially the most "unstable," if we compare, for example, the experimental data of [18] and [11, 16]. For the systems $CH_4 + Ar - N_2$ and $C_3H_8 + CO_2 - N_2O$ (Fig. 2), the PDC of the components are comparable with each other, and the critical Rayleigh numbers are almost invariable in the diffusion region, which suggests the absence of changes of regimes. This is also confirmed by experimental data of [17], which do not show concentration-induced convection.

In [14] it is found that the wave numbers (perturbation modes) that determine the flow patterns and the critical Rayleigh number for the case of heat convection are interrelated. A similar situation is found for diffusion instability. An increase in the wave number leads to a decrease in the critical Rayleigh number for all components involved in the transfer (Fig. 3). This can be explained as follows. Penetrating concentration perturbations expand the regions of their "existence" and involve more and more of the ambient gas mixture in convective flows, which results in an increase in the intensity of unstable transfer. For systems in which the PDC of the components are comparable with each other, conditions can be selected (for example, geometrical dimensions of the diffusion channel) under which an increase in the wave number has almost no effect on the behavior of the critical Rayleigh number. In such cases, it is likely that transition from the stability state to the state of diffusion instability is impossible.

Thus, with the present model for various wave numbers it is possible to determine the range of critical Rayleigh numbers that characterize the region of stable (unstable) diffusion and to demonstrate the diversity of convective forms of motion that can be realized in a cylindrical channel in the case of unstable mass transfer in isothermal three-component gas mixtures.

NOTATION

u, convection rate; p, pressure; ρ_i, ρ, density of i-th component and density of mixture; η, ν, dynamic and kinematic viscosities of mixture; c_i, molecular concentration of i-th component; $D_{i,j}$, diffusion coefficients; g, gravitational acceleration; $\boldsymbol{\gamma}$, upward vertical unit vector; D_i, partial diffusion coefficient; $\mathrm{Pr}_i = \nu/D_i$, diffusion Prandtl number of i-th component; $\mathrm{R}_i = g\beta_i B_i r^4/D_i\nu$, diffusion Rayleigh number of i-th component; $\tau_i = D_i/D_3$, parameter that characterizes ratio of diffusion coefficients; n, periodicity number in asimuthal direction; m, periodicity number along vertical axis z.

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