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THEORETICAL
AND MATHEMATICAL PHYSICS

Instability of Mechanical Equilibrium during Diffusion in a Three-Component Gas Mixture in a Vertical Cylinder with a Circular Cross Section

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Abstract—The problem of the emergence of instability of mechanical equilibrium of a three-component gas mixture during diffusion in a vertical channel with the wall impenetrable for the mass flux is considered in the case when the channel has a circular cross section. The critical Rayleigh numbers are determined, and the neutral stability lines are found in analytic form. The results are compared with experimental data.

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INTRODUCTION

Experimental studies of diffuse mixing of multicomponent gas mixtures show that instability of mechanical equilibrium of the mixture followed by the emergence of gravitational concentration convection may take place in systems under certain conditions [1–6]. Such problems associated with the formation of convective flows can be described using the stability theory [7, 8]. The main regularities in such an approach were obtained for the cases describing thermal convection of incompressible one-component [7, 9] or two-component liquids [10]. In the present paper, the formalism developed in [7–11] is extended to the case of diffuse mixing of isothermal three-component gas systems in a cylindrical channel with a circular cross section and with the walls impermeable for the mass flux.

1. BASIC EQUATIONS

The macroscopic motion of an isothermal three-component gas mixture can be described by the general system of equations including the Navier–Stokes equation as well as the equation of the conservation of the number of particles in the mixture and its components. Taking into account the conditions independent diffusion during which $\sum_{i=1}^{3}\mathbf{j}_i = 0$ and $\sum_{i=1}^{3} c_i = 1$ for an isothermal gas mixture, this system of equations has the form [7, 12]

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

$$\frac{\partial n}{\partial t} + \operatorname{div}(n\mathbf{v}) = 0, \quad \frac{\partial c_i}{\partial t} + \mathbf{v}\nabla c_i = -\operatorname{div}\mathbf{j}_i,$$

$$\mathbf{j}_1 = -(D_{11}^*\nabla c_1 + D_{12}^*\nabla c_2),$$

$$\mathbf{j}_2 = -(D_{21}^*\nabla c_1 + D_{22}^*\nabla c_2). \qquad (1)$$

Here, $\mathbf{u}$ is the average mass velocity vector, $\mathbf{v}$ is the vector of numerical average velocity, ρ is the density, p is the pressure, η and ξ are the shear and bulk viscosity coefficients, $\mathbf{g}$ is the free-fall acceleration vector, n is the number density, t is the time, c_i is the concentration of the ith component, $\mathbf{j}_i$ is the diffusion flux density vector for the ith component, and D_{ij}^* are the actual diffusion coefficients determined in terms of the mutual diffusion coefficient (MDCs) [13].

Equations (1) should be supplemented with the equation of state for the medium:

$$\rho = \rho(c_1, c_2, p), \quad T = \text{const}.$$

In the solution of system (1), we used the small-perturbation method [7], which presumed that the concentration c_i of the ith component and pressure p can be presented in the form

$$c_i = \langle c_i\rangle + c_i', \quad p_i = \langle p\rangle + p',$$

where $\langle c_i\rangle$ and $\langle p\rangle$ are the constant mean values taken as the origin.

Considering that for $L \gg r$ (L and r are the length and the radius of the diffusion channel, respectively), the difference between the perturbations in numerical average velocity **v** and average mass velocity **u** in the Navier–Stokes equation is insignificant [12], assuming that nonstationary perturbations of mechanical equilibrium are small, disregarding the terms quadratic in perturbations, and choosing appropriate scales for the units of measurements (distance d, time $d^2/\nu\nu$, velocity D_{22}^*/d, concentration $A_i d$ of the ith component, and pressure $\rho_0 \nu D_{22}^*/d^2$), we obtain the following system of equations describing gravitational convection for perturbed values in dimensionless units (primes are omitted):

$$p_{22}\frac{\partial c_1}{\partial t} - (\mathbf{u}\mathbf{e}_z) = \tau_{11}\nabla^2 c_1 + \frac{A_2}{A_1}\tau_{12}\nabla^2 c_2,$$

$$p_{22}\frac{\partial c_2}{\partial t} - (\mathbf{u}\mathbf{e}_z) = \frac{A_1}{A_2}\tau_{21}\nabla^2 c_1 + \nabla^2 c_2,$$

$$\frac{\partial \mathbf{u}}{\partial t} = -\nabla p + \nabla^2\mathbf{u} + (R_1\tau_{11}c_1 + R_2 c_2)\mathbf{e}_z, \tag{2}$$

$$\operatorname{div}\mathbf{u} = 0.$$

where $\mathbf{e}_z$ is the unit vector in the direction of the z axis, $p_{ii} = \nu/D_{ij}^*$ is the diffusion Prandtl number, $R_i = g\beta_i A_i d^4/\nu D_{ii}^*$ is the partial Rayleigh number, $\tau_{ij} = D_{ij}^*/D_{22}^*$ are the parameters determining the ratio of the actual diffusion coefficients, $\beta_i = -\frac{1}{\rho_0}\left(\frac{\partial\rho}{\partial c_i}\right)(p, T)$, and $A_i\mathbf{e}_z = -\nabla c_{i0}$ (subscript "0" corresponds to mean values).

2. MONOTONIC INSTABILITY NEUTRAL LINE FOR A VERTICAL CYLINDER WITH A CIRCULAR CROSS SECTION

Let us consider a three-component gas mixture filling an infinitely long vertical channel of a circular cross section shown in Fig. 1. We will analyze the critical motion parallel to the channel axis, assuming that perturbations of the concentration are independent of z: $u_x = u_y = 0$, $u_z = u(x, y)$, and $c_i = c_i(x, y)$. Then, we can write system of equations (2) in the form

$$\nabla^2 u + R_1\tau_{11}c_1 + R_2 c_2 = 0,$$

$$\tau_{11}\nabla^2 c_1 + \frac{A_2}{A_1}\tau_{12}\nabla^2 c_2 + u = 0,$$

$$\frac{A_1}{A_2}\tau_{21}\nabla^2 c_1 + \nabla^2 c_2 + u = 0, \tag{3}$$

$$\operatorname{div}\mathbf{u} = 0.$$

Passing to this system of equations, we disregard possible perturbations of pressure because the pressure relaxation time is shorter than the concentration relaxation time.

We introduce cylindrical coordinates (r, φ, z) and direct the z axis upwards along the axis of the cylinder.

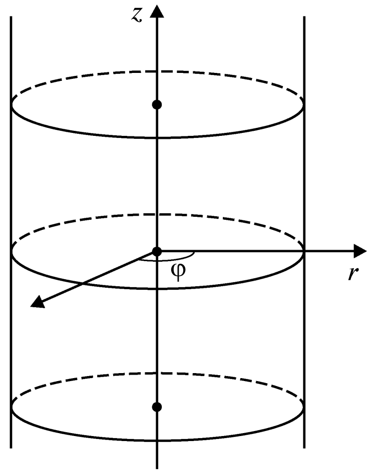

Fig. 1. Infinitely long vertical cylinder (coordinate system).

The problem under consideration has particular solutions, in which velocity u and concentration c_i of the components depend on angle φ in accordance with the harmonic law:

$$u(r, \varphi) = u^0(r)\cos n\varphi, \quad c_i(r, \varphi) = C_i^0(r)\cos n\varphi, \tag{4}$$

where n assumes integer values $n = 0, 1, 2, \ldots$.

Substituting relations (4) into (3), we obtain the following equations for radial functions u^0 and C_i^0:

$$u^0 + \tau_{11}\left(C_1^{0''} + \frac{1}{r}C_1^{0'} - \frac{n^2}{r^2}C_1^0\right)$$

$$+ \frac{A_2}{A_1}\tau_{12}\left(C_2^{0''} + \frac{1}{r}C_2^{0'} - \frac{n^2}{r^2}C_2^0\right) = 0,$$

$$u^0 + \frac{A_1}{A_2}\tau_{21}\left(C_1^{0''} + \frac{1}{r}C_1^{0'} - \frac{n^2}{r^2}C_1^0\right) \tag{5}$$

$$+ C_2^{0''} + \frac{1}{r}C_2^{0'} - \frac{n^2}{r^2}C_2^0 = 0,$$

$$u^{0''} + \frac{1}{r}u^{0'} - \frac{n^2}{r^2}u^0 + R_1\tau_{11}C_1^0 + R_2 C_2^0 = 0.$$

Eliminating the radial functions of the concentrations of the component from Eqs. (5), we obtain the equation containing only the velocity,

$$(D^2 - R)u^0 = 0, \tag{6}$$

where

$$D = \frac{d^2}{dr^2} + \frac{1}{r}\frac{d}{dr} - \frac{n^2}{r^2}, \quad R = R_1\tau_{11}K_1 + R_2K_2,$$

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

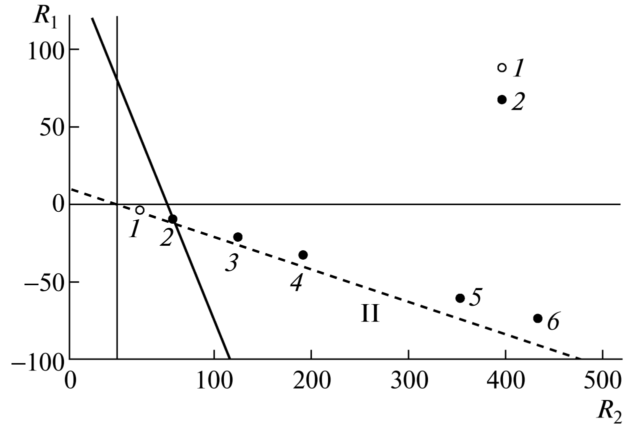

Fig. 2. Boundary monotonic instability (I) and zero density gradient (II) lines for the three-component system 0.7796He + 0.2204R12 – Ar at $T = 298.0$ K; points *1* and *2* correspond to experimental data.

The general solution to Eq. (6) has the form

$$u^0 = a_1 J_n(\gamma r) + a_2 I_n(\gamma r). \quad (7)$$

Here, J_n and I_n are the Bessel functions of the fist kind, a_1 and a_2 are arbitrary constants, and

$$\gamma = R^{1/4}, \quad \text{i.e.,} \quad \gamma = (R_1\tau_{11}K_1 + R_2K^2)^{1/4},$$

where dimensionless parameter γ] can be treated as an analog of the generalized Rayleigh number.

The concentrations of the components can be determined from the equations

$$\nabla^2 C_i^0 = -u^0 K_i, \quad i = 1, 2. \quad (8)$$

When calculating the concentrations, we must take into account only the solution to the inhomogeneous equation obtained as a result of substitution of expression (7) into (8). This gives

$$C_i^0 = \frac{K_i}{\gamma^2}(a_1 J_n(\gamma r) - a_2 I_n(\gamma r)). \quad (9)$$

At the boundary of the horizontal cross section of the channel, the velocity perturbations and the mass flux normal to the boundary vanish:

$$u = 0, \quad \frac{\partial c_i}{\partial t} = 0, \quad i = 1, 2 \text{ for } \quad r = \pm 1. \quad (10)$$

These boundary conditions lead to the following homogeneous system of equations for determining two integration constants:

$$a_1 J_n(\gamma) + a_2 I_n(\gamma) = 0,$$
$$a_1 J'_n(\gamma) - a_2 I'_n(\gamma) = 0. \quad (11)$$

The condition for the existence of a nontrivial solution to this system implies that its determinant vanishes. Expanding the determinant, we obtain the characteristic relation determining the spectrum of the critical values of the Rayleigh number:

$$\frac{J'_n(\gamma)}{J_n(\gamma)} + \frac{I'_n(\gamma)}{I_n(\gamma)} = 0. \quad (12)$$

Having determined constants a_1 and a_2 from system (11), we can find the velocity and concentration profiles corresponding to critical motion:

$$u = \left[\frac{J_n(\gamma r)}{J_n(\gamma)} - \frac{I_n(\gamma r)}{I_n(\gamma)}\right]\cos n\varphi, \quad (13)$$

$$c_i = \frac{K_i}{\gamma^2}\left[\frac{J_n(\gamma r)}{J_n(\gamma)} + \frac{I_n(\gamma r)}{I_n(\gamma)}\right]\cos n\varphi, \quad (14)$$

Critical Rayleigh numbers $R = \gamma^4$ are defined as the roots of transcendental equation (12). For a fixed number n (azimuthal component of the structure of motion) in Eq. (12), we have an infinite set of roots $\gamma_n^{(1)}$, $\gamma_n^{(2)}$, $\gamma_n^{(3)}$, ..., numbered in increasing order. The corresponding movements differ in the radial structure; a higher value of the superscript corresponds to a larger number of nodes of velocity and concentration [7]. Any critical Rayleigh number for the movements considered here has two indices: $R_n^{(l)} = [\gamma_n^{(l)}]^4$. The numerical solution of Eq. (12) for $l = 1$ gives the following roots:

$$\gamma_1 = 2.871, \quad \gamma_2 = 4.259, \quad \gamma_3 = 5.541,$$
$$\gamma_4 = 6.6771, \ldots$$

and, accordingly, the following Rayleigh numbers:

$$R_1 = 67.95, \quad R_2 = 329.1,$$
$$R_3 = 942.5, \quad R_4 = 2102. \quad (15)$$

To determine the line of monotonic perturbations, we form the scalar product of the first equation in system (3) and **u** and integrate over the cylinder cross section:

$$\int \mathbf{u}\nabla^2\mathbf{u}dS + R_1\tau_{11}\int uc_1 dS + R_2\int uc_2 dS = 0. \quad (16)$$

Substituting the expressions for the velocity and concentration distributions into Eq. (16) and performing integration, we obtain

$$\tau_{11}\left(1 - \frac{A_2}{A_1}\tau_{12}\right)R_1 + \left(\tau_{11} - \frac{A_1}{A_2}\tau_{21}\right)R_2$$
$$= \gamma^4(\tau_{11} - \tau_{12}\tau_{21}). \quad (17)$$

Figure 2 shows neutral line (17) on the (R_1, R_2) plane for system 0.7796He + 0.2204R12(2) – Ar(3) (here and below, the coefficient of the chemical element index indicates the concentration in molar fractions; the numbers in the parentheses following the element symbols reflect the numeration of gases). The instability region is located above straight line I, and the region corresponding to diffusion lies below this line. The critical concentration of Rayleigh numbers for the mode most hazardous from the stability point of view ($l = 1$, $n = 1$) have values $R_1 = 77.7936$ and $R_2 = 50.2471$ for $\gamma = 2.8712$. Figure 2 also shows line II corresponding to zero density gradient, which is defined as [12]

$$\tau_{11}R_1 = -R_2. \quad (18)$$

It can be observed that there are regions on the (R_1, R_2) plane, in which neutral line I lies below straight line II. The state of the mixture in such a region is unstable although the density of the mixture in the lower part of the channel is higher than in its upper part. This region corresponds to partial diffusion Rayleigh numbers with opposite signs ($R_1 < 0$ and $R_2 > 0$). Such a situation is typical of a nonisothermal binary mixture of liquids [7, 8].

3. COMPARISON WITH THE EXPERIMENT

We will compare the theoretical results with experimental data using the pressure dependence of diffuse mixing of a binary mixture of helium and Freon-12 with argon as an example. It was shown in [12, 14] that in such mixtures (e.g., 0.7796He + 0.2204R12(2) – Ar(3) at $T = 298.0$ K), an increase in the pressure may induce a transition from the diffusive state to the convective one. The stability test for such a system performed in [12] under the assumption of a linear distribution of the concentration components over the diffusion channel length (planar vertical layer, perturbations of the concentration components vanish at the boundaries) revealed a considerable discrepancy in the positions of the theoretical and experimental stability boundaries. The reason for such a discrepancy is that a substantial difference in the MDCs of the components leads to the situation in which the partial flux of the heaviest component is almost zero (i.e., a diffusion "gate" is formed [14]). In this case, the concentration distribution of the components over the channel length is apparently more intricate than the linear approximation. The theoretical calculations represented in Fig. 2 indicate that the allowance for the concentration component distribution in form (14), the investigation of the diffusion channel in the form of a vertical cylinder, and the fulfillment of the condition of zero mass flux at the channel boundaries eliminate the above-mentioned contradictions and lead to a quantitative agreement with experimental data.

To compare the results of determination of the stability and instability region, we can represent experimental data on the (R_1, R_2) plane in the form of partial Rayleigh numbers. As applied to diffusion channel of radius r and length L, we can write the partial Rayleigh numbers, in accordance with relations (2), in the form

$$R_1 = \frac{gnr^4\Delta m_1}{\rho\nu D_{11}^*}\frac{\partial c_1}{\partial z}, \quad R_2 = \frac{gnr^4\Delta m_2}{\rho\nu D_{22}^*}\frac{\partial c_2}{\partial z}, \quad (19)$$

where $\Delta m_1 = m_1 - m_2$ and $\Delta m_2 = m_2 - m_3$, m_i being the mass of a molecule of the ith species. If the experimental conditions (pressure, temperature, composition of the mixture in each flask, and the size of the diffusion channel) are known, formulas (19) can be used for determining R_1 and R_2 and, hence, for finding the point on the (R_1, R_2) plane, which represents the given experiment. It is well known from experiments that such a regime (diffusion or convection) is observed for the given conditions. We will mark the points corresponding to the unstable regime by symbols *2*; symbols *1* correspond to diffusion. The aggregate of such points on the plane of the Rayleigh numbers determines the transition from the diffusive to the convective region via the neutral stability line. Figure 2 shows experimental results in terms of the Rayleigh numbers, which were obtained for system 0.7796He + 0.2204R12 – Ar at $T = 298.0$ K by varying pressure. Points *1–5* correspond to the following values of pressure: 0.12 (*1*), 0.19 (*2*), 0.29 (*3*), 0.36 (*4*), 0.49 (*5*), and 0.54 MPa (*6*). Point *1* corresponds to the diffuse regime. Points *2–5* characterize the unstable regime. It can easily be seen that a satisfactory agreement between the theory and experiment is observed in the location of diffusion and concentration convection regions.

CONCLUSIONS

Thus, based on the stability theory, the possibility of instability in mechanical equilibrium is demonstrated for diffusion in isothermal three-component mixtures in an infinitely long cylindrical section of a circular cross section under the boundary conditions corresponding to zero mass flux though the channel walls. Comparison with experiment demonstrates a satisfactory agreement in the positions of the boundaries at which diffusion changes to concentration gravitational convection.

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