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CONVECTIVE REGIMES OF MIXING IN BINARY SYSTEMS WITH THE MECHANICAL EQUILIBRIUM INSTABILITY OF A GAS MIXTURE

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Experimental data on pressure that correspond to the diffusion–concentration-induced convection are presented. Stability analysis carried out with allowance for the channel geometry realized in the experiment shows the existence of various modes of perturbations, in good agreement with experimental data.

Introduction. Experimental investigations [1–4] have shown that in certain ternary gas systems at definite critical parameters there exists the mechanical equilibrium instability of a mixture followed by the occurrence of concentration-induced convection. The description of such types of problems related to natural thermal gravitational convection is carried out within the framework of the stability theory [5–7]. It was applied to isothermal analogs in the case of three-component gas systems [8]. It is evident that the characteristic features of mixing in both binary and multicomponent systems upon a change in pressure can be revealed by comparing the corresponding regimes of mixing. Below, we present experimental data on mixing in the isothermal binary systems Ar–He, N_2–He, and Ar–N_2 and analyze them with allowance for the real geometry of a diffusion channel in the case of unstable stratification of the gas mixture density. The dependence of the transferred portion of the mixture on pressure is determined.

Experimental Setup and Technique. Experiments were run in the pressure range 0.33–6.58 MPa at a temperature of 298 K on an experimental setup with application of the double-flask method [8]; the schematic diagram of the setup is shown in Fig. 1. The geometric dimensions of the diffusion apparatus are: the volumes of the upper, V_{I}, and lower, V_{II}, flasks are the same and equal to $55.6 \cdot 10^{-6}$ m^3; the length of the diffusion channel is $L = 6.4 \cdot 10^{-2}$ m, and its diameter is $d = 0.4 \cdot 10^{-2}$ m. The duration of all the experiments was 17 min except for the point depicted in Fig. 2 at a pressure of $p = 0.29$ MPa, where the experiment lasted for 60 min. The choice of the interval of 17 min was optimal as proved by search experiments. This was prompted by the convective process itself and by the analysis of gases after mixing.

When filling the flasks and controlling pressure during experiments, standard pressure gauges of accuracy class 0.4, with corresponding measurement limits and specially made membrane separators, were used. The separators 2 cut a rather large volume of the cavity of gauge 3 from the corresponding flask, thus excluding a considerable parasitic capacity. All gas pipes had an inner diameter of 1 mm.

The diffusion apparatus was made from Kh18N10T stainless steel and was located directly in the thermostat bath. The accuracy with which the thermostat maintained temperature was ±0.1 K. The construction of the shutting-off device in the apparatus did not change the volumes of the flasks at the time of opening and closing of the diffusion channel.

The experimental procedure was as follows. The flasks were disconnected and a roughing-down pump evacuated, e.g., the upper flask which was then flushed with a respective gas from a cylinder and thereafter was filled with this gas up to the experimental pressure. After the attainment of the prescribed temperature regime (in our case $T = 298.0 \pm 0.1$ K) the pressure in the flasks was equalized via a special vessel 4. The pressure in the flasks was recorded by standard pressure gauges. After this the diffusion channel was opened and simultaneously a timing device was switched on. After the end of the experiment the flasks were disconnected, and the time of mixing was noted. An

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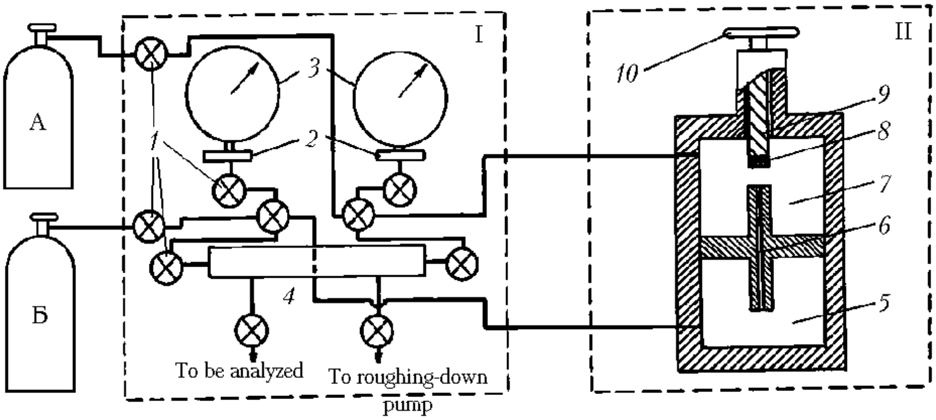

Fig. 1. Schematic of the experimental setup of the two-flask method: A and B, cylinders with a heavy and light gases; I, block of preparation of gases; II, thermostat with the two-flask apparatus; 1) taps; 2) membrane separators; 3) standard gauges; 4) leveling vessel; 5) lower flask; 6) diffusion channel; 7) upper flask; 8) Teflon preform; 9) rod; 10) flywheel.

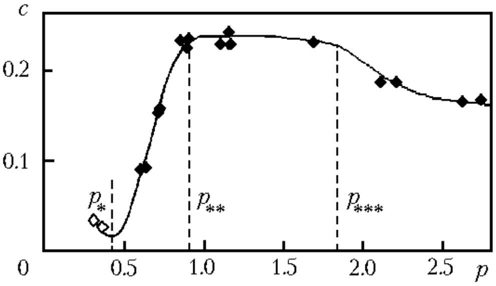

Fig. 2. The quantity of argon that passed from the upper flask into the lower one depending on pressure and different regimes of mixing: up to $p_* \approx 0.4$ MPa — molecular diffusion; from p_* to $p_{**} \approx 0.9$ MPa — inception and development of convection; from p_{**} to $p_{***} \approx 1.8$ MPa — region of transition from one type of convective mixing to other; above p_{***} — drop in convection intensity. Points — experimental data; curve — approximation by a polynomial.

analysis of the mixture of gases after mixing from both the upper and lower flasks was made by a chromatograph. Having found the concentration of components after mixing, its regime was determined (molecular or convective) by comparing experimental data with those calculated by the formula from [9]:

$$D_{12} = \frac{B}{\tau} \ln \frac{\Delta c_0}{\Delta c_\tau}, \tag{1}$$

where $B = LV_{\text{I}}V_{\text{II}}/S(V_{\text{I}} + V_{\text{II}})$; Δc_0 and Δc_τ are the differences in the concentrations of components in the flasks of the apparatus at the initial and final instants of time.

Depending on the condition of the comparison the diffusion coefficients could be converted to a normal pressure. If the experimental value of the coefficient corresponded, within the experimental error (of the order of 3%), to the literature data [9, 10], we could state that in the system investigated there are fluxes only of diffusional nature. In the case of larger discrepancies there could be only disturbance of molecular mass transfer by microflows, i.e., by gravitational free convection. All investigations were carried out with a denser component being poured into the upper flask from cylinder A and a lighter component in the lower flask poured from cylinder B. All gases were considered ideal up to a pressure of 7 MPa, since deviation from ideality amounted to 2–4%.

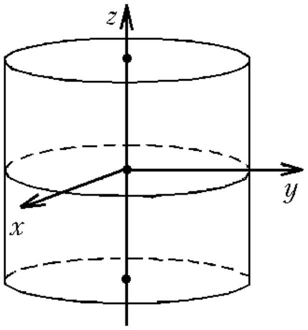

Fig. 3. Geometry of the problem. Coordinate system.

Experimental data for the Ar–N$_2$ system at different pressures are presented in Fig. 2. The quantity of gas that passed from the upper flask is similar to that passed in the opposite direction. The points that correspond to the unstable regime are denoted by black symbols, whereas those corresponding to diffusion — by open ones.

The analysis of experimental data allows the conclusion that at a pressure lower than $p_* \cong 0.4$ MPa the Ar–N$_2$ system is in the region of diffusion (and this is despite the fact that the heavy gas is above the light one). Note that we conducted a special series of experiments (for this region of pressures) in which we changed their duration from test to test. The values of the coefficients of mutual diffusion recovered from experimental data were compared with those given in the literature. The coincidence of the measured data with those taken from literature sources confirmed that the processes in progress in different time intervals is precisely the diffusional one.

When $p \geq p_*$, the stability of the mechanical equilibrium of the system is disturbed and convective flows appear in it, with their intensity being increased with pressure. However, at a pressure p_{**} (equal for the given mixture to 0.9 MPa) this tendency is disturbed. Evidently the number of convective flows undergoes a change in the system, which cannot but influence the intensity of mass transfer. With further rise in pressure the mass transfer intensity virtually does not change. And, finally, beginning from the pressure $p_{***} \approx 1.8$ MPa the mass transfer intensity decreases with increase in pressure.

Statement and Solution of the Problem. We will try to explain the results obtained within the framework of an analysis of the mechanical equilibrium stability of a gas mixture [5–7].

The microscopic motion of a binary mixture of ideal gas under isothermal conditions is described by a general system of hydrodynamic equations which includes the Navier–Stokes equations of motion, continuity, and diffusion. Neglecting in them the terms responsible for barodiffusion, since the pressure drop occurring under its influence is small in comparison with the pressure values ($\Delta p \ll p$), and taking into account the condition $c_1 + c_2 = 1$, we have the following system of equations [6, 7]:

$$\rho \left[\frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u}\nabla)\, \mathbf{u} \right] = - \nabla p + \eta \Delta \mathbf{u} + \rho \mathbf{g} \,, \quad \rho \left(\frac{\partial c}{\partial t} + \mathbf{u} \nabla c \right) = - \operatorname{div} \mathbf{j} \,, \quad \mathbf{j} = - \rho D_{12} \nabla c \,, \quad \frac{\partial \rho}{\partial t} + \operatorname{div} (\rho \mathbf{u}) = 0 \,. \tag{2}$$

System (2) is supplemented with the equation of state of the medium:

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

We consider that the thermodynamic variables c and pressure p have the form

$$c = \langle c \rangle + \tilde{c}\,, \quad p = \langle p \rangle + \tilde{p}\,,$$

where $\langle c \rangle$ and $\langle p \rangle$ are the average values taken as the start of reading, and $\tilde{c}$ and $\tilde{p}$ are small perturbations. The change in the density caused by the pressure nonuniformity is small as compared to the changes due to the inhomogeneity of the composition and indicates that the pressure along the gas mixture should not change substantially. Then, by linearizing the initial system, neglecting the terms of second order of smallness, and dimensionalizing as in [6], the problem can be reduced to a system of equations for perturbed quantities (here and hereafter the signs "~" are omitted):

$$\frac{\partial \mathbf{u}}{\partial t} = -\nabla p + \Delta \mathbf{u} + \mathrm{Ra}\, c\boldsymbol{\gamma}\,, \quad \mathrm{Pr}\frac{\partial c}{\partial t} - (\mathbf{u}\boldsymbol{\gamma}) = \Delta c\,, \quad \mathrm{div}\, \mathbf{u} = 0\,, \tag{3}$$

here d is the characteristic scale of distance; d^2/ν, of time; D_{12}/d, of velocity; Ad, of concentration (A is given by the expression $\nabla c_0 = -A\gamma$, ∇c_0 is the concentration gradient in the absence of perturbations); $\rho_0 \nu D_{12}/d^2$, of pressure; $\beta = \frac{1}{\rho_0}\left(\frac{\partial \rho}{\partial c}\right)_{p,T}$, $\rho_0 = \rho(\langle c\rangle, \langle p\rangle)$, $\nu = \eta/\rho_0$, and the diffusion Rayleigh number $\mathrm{Ra} = g\beta A d^4/(\nu D_{12})$ and Prandtl number $\mathrm{Pr} = \nu D_{12}$ are the similarity numbers.

First, we consider the case of an infinite vertical cylinder (Fig. 3). By analogy with [6], we will consider that $\mathbf{u}(\mathbf{r}, t) = u(\mathbf{r}) \exp[-\lambda t]$, $c(\mathbf{r}, t) = c(\mathbf{r}) \exp[-\lambda t]$, $\nabla p = 0$, $u_r = 0$, $u_\varphi = 0$, $u_z = u(r)\cos(n\varphi)$, $c = c(r)\cos(n\varphi)$. Then, taking into account the conditions of vanishing of the velocity and flux of substance at the boundaries of the cylindrical layer ($r = 1$)

$$\mathbf{u}(1) = 0\,, \quad c'(1) = 0\,, \tag{4}$$

where the prime denotes a derivative with respect to r, we reduce (3), if (4) is satisfied, to a system of equations for perturbations:

$$-\lambda u = u'' + \frac{1}{r}u' - \frac{n^2}{r^2}u + \mathrm{Ra}\, c\,, \quad -\lambda\, \mathrm{Pr}\, c = c'' + \frac{1}{r}c' - \frac{n^2}{r^2}c + u\,. \tag{5}$$

In order to obtain equations for neutral perturbations (i.e., that do not decay or increase in time) we substitute $\lambda = 0$. Then the system of equations (5) will be written as follows:

$$u'' + \frac{1}{r}u' - \frac{n^2}{r^2}u + \mathrm{Ra}_{\mathrm{cr}}\, c = 0\,, \quad c'' + \frac{1}{r}c' - \frac{n^2}{r^2}c + u = 0\,, \tag{6}$$

where $\mathrm{Ra}_{\mathrm{cr}}$ corresponds to these perturbations. Solution (6) allows one to obtain the range of critical Rayleigh numbers that, in the mode with $n = 1$, determine the conditions of transition from the region of molecular diffusion to convection (the convection is induced in this case in the form of diametrically antisymmetric motion, i.e., the gas ascends in one-half of the channel and descends in the other), whereas in higher modes they determine the transition from convection of one space structure to another (with a higher number and other arrangement of convective fluxes).

The general solution (5), which remains finite when $r \to 0$, has the form [6]

$$u = C_1 J_n(Kr) + C_2 I_n(Kr)\,, \quad K = \mathrm{Ra}^{1/4}\,. \tag{7}$$

Substituting the general solution for the velocity into Eq. (5), we obtain

$$c = \frac{1}{K^2}\left(C_1 J_n(Kr) - C_2 I_n(Kr)\right). \tag{8}$$

Determining the constants C_1 and C_2 from Eqs. (7) and (8), we find the velocity and concentration profiles that correspond to critical motions [6]:

$$u_n = \frac{J_n(Kr)}{J_n(K)} - \frac{I_n(Kr)}{I_n(K)}\,, \quad c_n = \frac{1}{K^2}\left(\frac{J_n(Kr)}{J_n(K)} + \frac{I_n(Kr)}{I_n(K)}\right). \tag{9}$$

The critical Rayleigh numbers $\mathrm{Ra}_{\mathrm{cr}} = K^4$ are found as the roots of the transcendental equation

$$\frac{J_n'(K)}{J_n(K)} + \frac{I_n'(K)}{I_n(K)} = 0 \tag{10}$$

at a fixed n that determines the azimuthal structure of motion. Equation (10) has an infinite sequence of the roots $K_n^{(1)}$, $K_n^{(2)}$, ..., given in their ascending order. The corresponding motions differ in the radial structure; for the higher value of the superscript there corresponds a greater number of nodes of velocity and concentration. Thus, any critical Rayleigh number for motions of the form considered has two indices $\mathrm{Ra}_n^{(l)} = \left[K_n^{(l)}\right]^4$.

Having solved Eq. (10), we obtain [6]: $K_1^{(1)} = 2.871$, $K_1^{(2)} = 6.145$, $K_1^{(3)} = 9.333$, ..., $K_2^{(1)} = 4.259$, $K_2^{(2)} = 7.572$, ..., $K_3^{(1)} = 5.541$, $K_2^{(3)} = 8.932$, ..., $K_4^{(1)} = 6.771$, ... Correspondingly, $\mathrm{Ra}_1^{(1)} = 67.95$, $\mathrm{Ra}_1^{(2)} = 1426$, $\mathrm{Ra}_1^{(3)} = 7586$, ..., $\mathrm{Ra}_2^{(1)} = 329.1$, $\mathrm{Ra}_2^{(2)} = 3286$, ..., $\mathrm{Ra}_3^{(1)} = 942.5$, $\mathrm{Ra}_3^{(2)} = 6365$, ..., $\mathrm{Ra}_4^{(1)} = 2102$, ...

Now, we will consider a cylinder of finite height. Just as in [6] we will assume that

$$u_r = z\,(h^2 - z^2)\,v(r)\cos(n\varphi)\,, \quad u_\varphi = z\,(h^2 - z^2)\,w(r)\sin(n\varphi)\,, \quad u_z = \frac{1}{4}(h^2 - z^2)^2\,u(r)\cos(n\varphi)\,, \tag{11}$$

where $n = 0, 1, 2, ..., h = L/d$. The radial functions u, v, and w should vanish on the solid side surface of the cylinder (at $r = 1$). These requirements can be satisfied by assuming that [6]

$$u = \frac{J_n(Kr)}{J_n(K)} - r^n\,, \quad v = -\frac{1}{KJ_n(k)}\left[J_n'(Kr) - J_n'(K)\,r^{n+1}\right], \quad w = \frac{n}{K^2J_n(K)}\left[\frac{1}{r}J_n(Kr) - J_n(K)\,r^{n+1}\right],$$

where the parameter K satisfies the condition [6]

$$KJ_n''(K) = (n+1)\,J_n'(K)\,.$$

To find c it is necessary to solve the equation $\Delta c = -u_z$. Assuming, just as in [6], that $c = f(r, z)\cos(n\varphi)$, for f we obtain the equation

$$\frac{\partial^2 f}{\partial r^2} + \frac{1}{r}\frac{\partial f}{\partial r} - \frac{n^2}{r^2}f + \frac{\partial^2 f}{\partial z^2} = -\frac{1}{4}\left[\frac{J_n(Kr)}{J_n(K)} - r^n\right]\left(h^2 - z^2\right)^2. \tag{12}$$

We assume that perturbations of concentration on the end-face surfaces vanish. Taking into account the supplementary conditions that follow from (12), that is, the vanishing of the second derivative $\partial^2 f/\partial z^2$ on the end-faces, we may select the following approximation for the function f [6]:

$$f(r, z) = (h^2 - z^2)\,(5h^2 - z^2)\,c(r)\,. \tag{13}$$

To determine $c(r)$ we will employ the Kantorovich method. Just as in [6], substituting (13) into (12), multiplying the part of the function $f(r, z)$ that depends on z, and integrating it over z within the limits from $-h$ to h, we obtain

$$c'' + \frac{1}{r}c' - \left(\frac{n^2}{r^2} + \alpha^2\right)c = -\frac{11}{248}\left[\frac{J_n(Kr)}{J_n(K)} - r^n\right],$$

TABLE 1. Comparison of Experimental and Theoretical Data for Three Systems

Pressure, MPa	Mixture		
	Ar–He	Ar–N_2	N_2–He
		$n = 1$	
p_*	0.54	0.47	0.58
p_{theor}	0.55	0.48	0.62
		$n = 2$	
p_{**}	1.30	0.90	1.40
p_{theor}	1.24	1.03	1.37

where $\alpha^2 = 153/62h^2$. Having solved this equation for c, provided that the side surfaces are impermeable for the substance flux $\left.\frac{\partial c}{\partial r}\right|_{r=1} = 0$, we have

$$c(r) = \frac{11}{248\alpha^2 (K^2 + \alpha^2)} \left[\alpha^2 \frac{J_n(Kr)}{J_n(K)} + \frac{K^2 (n^2 + \alpha^2) + 2n (K^2 + \alpha^2)}{n+2} \frac{I_n(\alpha r)}{I_n'(\alpha)} - (K^2 + \alpha^2) r^n \right]. \tag{14}$$

The resulting expressions for u and c allow one to calculate the critical Rayleigh number with the aid of the integral Galerkin relation:

$$\text{Ra} = -\frac{\int u_z \Delta u_z dV}{\int u_z c dV}.$$

For the given diffusion apparatus $h = \frac{6.4 \cdot 10^{-2}}{0.4 \cdot 10^{-2}} = 16$, therefore we find that $\text{Ra}_1^{(1)} = 70.1$, $\text{Ra}_2^{(1)} = 338.1$, $\text{Ra}_3^{(1)} = 967.3$, etc. As is known, if the value of the Rayleigh number in the system exceeds the critical Rayleigh number for the first, most dangerous, mode of perturbations, the system must develop a critical flow corresponding to this Rayleigh number. The critical motions that correspond to Rayleigh numbers for higher modes are inaccessible until conditions are created in the system that prevent the development of the first critical motion.

The investigations carried out by us were aimed at studying the behavior of the system under conditions that greatly exceeded the critical ones, that is, conditions were created that favored the development of motions for both the main mode and much higher modes, with perturbations developing simultaneously and the main mode having no advantages in time; therefore the motion corresponding to it cannot be regarded as a developed one. Mixing in the system with conditions available for origination of both motions (for the main and higher mode) had to differ somehow from mixing in the system in which only motion in the main mode is possible. The appearance of this motion should have an influence on the character of mixing.

According to (3), the experimental Rayleigh number in the case of a diffusion channel with a radius r and length L that connects the upper flask I of the apparatus with the lower one II, was determined in terms of the channel radius and is written as follows: $\text{Ra} = \frac{gbr^4 \Delta m \Delta c}{\rho_0 \bar{\nu} D_{12} L}$, where $\Delta m = m_1 - m_2$ is the difference between the masses of the molecules of components, $\Delta c = c_{1\text{I}} - c_{1\text{II}}$, $b = p/kT$. With the experimental conditions being known (pressure, temperature, composition of the mixture in each of the flasks, geometrical dimensions of the diffusion channel), it is possible to find the experimental Rayleigh number and subsequently the transition boundary. It is seen from Fig. 2 that for the Ar–N_2 mixture at the pressure $p_* \approx 0.47$ MPa one can observe the transition from molecular diffusion to the beginning of convection which rapidly increases with pressure, with the diffusional transfer still being present, though not playing a marked role. The transition pressure $p = 0.47$ (0.48) MPa corresponds to the theoretical Rayleigh number $\mathrm{Ra}_1^{(1)} = 67.95$ (70.1) obtained. (Here, the first value corresponds to the infinite vertical cylinder and the second (in brackets) — to a cylinder of finite height). At the pressure $p_{**} \approx 0.9$ MPa the character of flow in the system is rearranged, influencing the intensity of mass transfer. For the predicted Rayleigh number $\mathrm{Ra}_2^{(1)} = 329.1$ (338.1) there corresponds the transition pressure $p = 1.0$ (1.0) MPa. At a pressure of 1.8 MPa that corresponds to the Rayleigh number $\mathrm{Ra}_3^{(1)} = 942.5$ (967.3), the character of flow changes once again.

For each number of periodicity there corresponds its own character of flow. It would be desirable to cite several values of Rayleigh number at $n = 3$, but a maximum coincidence between the theory and experiment is demonstrated precisely by that number which is given in the present work. We assume that convection of precisely such spatial structure that corresponds to this number is realized experimentally.

The results for all three mixtures are presented in Table 1, from which satisfactory agreement between experimental and predicted values for the problems considered by us is seen (an infinite vertical cylinder and a cylinder of finite height). It is seen that for the Ar–He and N_2–He systems the difference between theory and experiment for the first and second modes can be explained by the still rather great influence of purely molecular diffusion on the boundaries of transition from one type of mixing to another.

Conclusions. As a result of the investigations carried out, it has been found that, depending on the pressure during mixing of the two gases, different forms of convective motions appear in the systems investigated, and this exerts an effect on the intensity of mixing.

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NOTATION

A, dimensionless gradient of concentration; B, instrumental constant, m^2; b, concentration, m^{-3}; C_1 and C_2, integration constants; c, c_1, and c_2, concentration of a heavy component, concentration of the first (heavy) and second (light) gases, respectively; d, diameter of the diffusion channel, characteristic scale of the measurement of distance, m; D_{12}, coefficient of mutual diffusion, m^2/sec; $\langle f \rangle$, average value of f taken for the reference point; $\tilde{f}$, perturbation of quantity f; g, $\mathbf{g}$, free fall acceleration (scalar and vector), m/sec^2; h, dimensionless height for the cylinder of finite height; I_n, J_n, Bessel function of the first kind, order n for the imaginary and real argument, respectively; $\mathbf{j}$, diffusion flux density of a heavy component, kg·m^{-2}·sec^{-1}; k, Boltzmann constant, J/K; K, dimensionless parameter; L, length of the diffusion channel, m; l, number that characterizes the radial structure of spatial perturbations; m_i, mass of a molecule of the ith component, kg; n, mode of perturbations; Pr, Prandtl number; p, pressure, N/m^2; p_*, p_{**}, and p_{***}, the first, second, and third pressures of transition, N/m^2; Ra and $\mathrm{Ra}_{\mathrm{cr}}$, Rayleigh number and critical Rayleigh number; r, $\mathbf{r}$, radius of diffusion channel (scalar and vector ones), m; S, area of the diffusion channel, m^2; T, temperature, K; t, time, sec; $\mathbf{u}$, mean mass velocity of mixture, m/sec; $u(r)$, $v(r)$, and $w(r)$, radial functions that characterize velocities over the axes z, r, and φ of the cylindrical coordinate system, respectively; V_{I} and V_{II}, volumes of the upper and lower flasks, respectively, m^3; α, dimensionless parameter that characterizes the type of mixing, parameter in equation for the cylinder of finite height; β, coefficient of thermal expansion, m/K; γ, unit vector directed vertically upward; η, coefficient of dynamic viscosity, kg·m^{-1}·sec^{-1}; λ, damping constant, sec^{-1}; ν and $\bar{\nu}$, coefficients of kinematic viscosity and mixture-average coefficient of kinematic viscosity, m^2/sec; ρ and ρ_0, local and average density of a mixture, respectively, kg/m^3; τ, time of carrying out a test, sec; φ, angle.

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