

tions leads to a considerable decrease (by up to 2-3 times) in the minimum irrigation densities.

It is important to note that the results which have been obtained were achieved with relatively small energy consumptions for exciting the fluctuations, namely, 3-8% of the energy required for establishing the film flow.

NOTATION

G_0 , liquid flow rate, m^3/sec ; Γ_0 , irrigation density, m^2/sec ; f_n , frequency of imposed fluctuations, Hz; δ , liquid film thickness, mm; D , dispersion of the wavy flow with superimposition of fluctuations, mm^2 ; D_0 , dispersion of steady-state wavy flow, mm^2 ; Γ_1 , irrigation density for the beginning of the appearance of dry patch, m^2/sec ; Γ_2 , irrigation density at moment of disappearance of dry patch in film, m^2/sec .

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VISCOUS ISOTHERMAL MOTION OF A BINARY GAS MIXTURE THROUGH AN ORIFICE

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Motion of gases through an orifice at low Knudsen number is studied.

Motion of gas mixtures through an orifice in a thin film occurs in many technological processes. From the scientific viewpoint this process is interesting because any effect of channel walls on the gas flow is absent. However, until now experimental and theoretical studies of flows through an orifice have been limited in number and have essentially considered only single-component gases.

The present study will investigate the kinetic coefficients of isothermal motion of a binary gas mixture through an orifice at Knudsen numbers much less than unity.

1. Kinetic Coefficients of Isothermal Motion of a Binary Gas Mixture in Long Channels and an Orifice. If at the ends of a channel we create a pressure difference Δp and (or) a concentration difference Δc , then motion of the gas mixture within the channel commences.

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Thermodynamic treatment of this situation permits relating the means of the component velocities across the channel section \bar{u}_1 and \bar{u}_2 to the pressure and concentration differences which cause the former in terms of kinetic coefficients L_{ij} with the expressions [1]:

$$\bar{u}_1 c_1 + \bar{u}_2 c_2 = -L_{11} \Delta p - L_{12} p \Delta c_1, \quad \bar{u}_1 - \bar{u}_2 = -L_{21} \Delta p - L_{22} p \Delta c_1. \quad (1)$$

Under concrete conditions the values of the kinetic coefficients are determined by the characteristics of the mixing gas molecules and their interaction with the channel walls, as well as the channel geometry, temperature, pressure, and mixture concentration.

For long channels the coefficients L_{ij} have been thoroughly studied, both theoretically [2] and experimentally [3]. However, for viscous motion of a gas mixture through an orifice the only kinetic coefficient completely known is L_{11}^0 , describing flow of a gas caused by a pressure difference. Using an analytical solution of the Navier-Stokes equation [4] demonstrated that the mean velocity of motion of a single-component gas in the orifice section at small Knudsen numbers and low pressure heads $\Delta p \ll p$ is given by the expression

$$\bar{u}_1^0 = -\frac{R}{3\pi\eta_1} \Delta p. \quad (2)$$

This result was confirmed experimentally in [5]. Since in a viscous flow regime the velocities of the mixture components are close to each other ($\bar{u}_1 \approx \bar{u}_2 \approx \bar{u}$), Eq. (2) is also valid for mixtures, if in place of the viscosity of the single-component gas η_1 we use the mixture viscosity η_{12} . The goal of the present study is to investigate the kinetic coefficients L_{12}^0 , L_{21}^0 , and L_{22}^0 for an orifice, for which there are at present no theoretical or experimental data available.

A round orifice can be considered the limiting case of a cylindrical channel with radius R and length ℓ as $\ell \rightarrow 0$. In view of this it can be expected that the expressions for kinetic coefficients describing viscous ($\text{Kn} \rightarrow 0$) motion of gas mixtures through an orifice will basically coincide with analogous expressions for the coefficients for extended channels of the same radius. Therefore, to establish the form of L_{ij}^0 at an orifice we will use expressions for the corresponding kinetic coefficients from [1] for the case $\ell \gg R \gg \lambda$:

$$L_{11} = \frac{R^2}{8\eta_{12}l}, \quad L_{12} = \frac{\sigma D_{12}}{lp}, \quad L_{21} = \frac{\alpha_p D_{12}}{lp}, \quad L_{22} = \frac{D_{12}}{c_1 c_2 lp} \quad (3)$$

and generalize these to the case of the orifice.

Using Eq. (2) and definition (1), we obtain the kinetic coefficient L_{11}^0 for the orifice:

$$L_{11}^0 = \frac{R}{3\pi\eta_{12}}. \quad (4)$$

Comparing the expressions for L_{11} for a long channel, Eq. (3), and an orifice, Eq. (4), we note that they may be written in one and the same form by distinguishing an "effective length" for the orifice in Eq. (3):

$$l_p^{\text{ef}} = \frac{3\pi}{8} R, \quad L_{11}^0 = \frac{R^2}{8\eta_{12}l_p^{\text{ef}}}. \quad (5)$$

It should be obvious that such introduction of l_p^{ef} is possible only in a viscous flow regime. For arbitrary numbers Kn the differences in the expressions for L_{11} and L_{11}^0 will be related to a number of additional parameters which appear upon transition from an orifice to a channel of finite length (the accommodation coefficients, Knudsen number over channel length).

The introduction of an effective length l_p^{ef} has been used previously in considering flow [6] and diffusion [7] in channels with a finite ratio of channel length to radius. The physical meaning of l_p^{ef} is that outside the channel near both its ends there exist regions of gas perturbed by the flow which produce an additional contribution to channel resistance. This change in channel resistance is equivalent to an increase in its length by an amount Δl , which is termed the end correction. Having replaced the real channel length

by its effective value, such that $\ell^{ef} = \ell + \Delta\ell$, in the calculation we may use the same relationships as for channels with $\ell \gg R$. It is obvious that near an orifice there also exist perturbed regions, with characteristic dimension ($\sim R$), which will define the "effective length" of the orifice. Since the characteristic distances of the concentration and pressure distribution fields may not coincide, we introduce two effective lengths: ℓ_p^{ef} for the coefficients L_{11}^0 and L_{21}^0 , describing flows caused by a pressure difference, and ℓ_c^{ef} for coefficients L_{12}^0 and L_{22}^0 , related to a concentration difference.

To determine ℓ_c^{ef} we find an expression for the coefficient L_{22}^0 . We will consider a system of two flasks (with characteristic dimension R_0), filled with different gases. The gases mix through an orifice in the thin membrane dividing the volumes. The baroeffect which develops in such a system leads to vanishing of the mean numerical velocity of the mixture $c_1\bar{u}_1^0 + c_2\bar{u}_2^0 = 0$. In this case the quasisteady state concentration field in the vessels with neglect of barodiffusion ($L_{21}\Delta p \ll L_{22}p\Delta c_1$) will be described by the Laplace equation $\Delta c = 0$. If the diffusion resistance of the orifice ($\sim D_{12}/R$) is much greater than the diffusion resistance of the volumes ($\sim D_{12}/R_0$), then the real problem geometry can be approximately replaced by a simpler one, corresponding to mixing through an orifice of gases occupying two semiinfinite volumes. Having solved the Laplace equation together with the boundary conditions of concentration and mixture flow continuity in the plane of the orifice, the nonpenetration conditions on the wall separating the mixing gases, and constancy of mixture composition far from the orifice, we can find the concentration field which develops upon mixing through the orifice. Omitting calculations which can be found in [8], we present the final result. In a cylindrical coordinate system with origin in the center of the orifice and z axis perpendicular to the plane of the wall, the change in concentration of the first component from the value c_1^+ , corresponding to the mixture composition $z \rightarrow \infty$, to the value c_1^- , corresponding to the composition as $z \rightarrow -\infty$, is described by the function

$$c_1 = c_1^+ - \frac{c_1^+ - c_1^-}{\pi} \arcsin \frac{2R}{\sqrt{z^2 + (R+r)^2} + \sqrt{z^2 + (R-r)^2}},$$

if $z \geq 0$ (6)

$$c_1 = c_1^- - \frac{c_1^+ - c_1^-}{\pi} \arcsin \frac{2R}{\sqrt{z^2 + (R+r)^2} + \sqrt{z^2 + (R-r)^2}},$$

if $z \leq 0$.

According to [9], the local velocities (defined at a point) follow the law

$$\mathbf{u}_1 - \mathbf{u}_2 = - \frac{D_{12}}{c_1 c_2} \nabla c_1. \quad (7)$$

Analysis of the concentration fields of Eq. (6) shows that mixture concentration remains constant at any point in the plane of the orifice. Averaging Eq. (7) over the channel section with consideration of this fact, we obtain

$$\bar{u}_1^0 - \bar{u}_2^0 = \frac{D_{12}}{c_1 c_2} \left. \frac{dc_1}{dz} \right|_{z=0}. \quad (8)$$

Neglecting in the second equation of system (1) the small contribution of barodiffusion and equating to Eq. (8), we obtain

$$L_{22}^0 = \frac{D_{12}}{c_1 c_2 p \ell_c^{ef}}, \quad \ell_c^{ef} = \left. \frac{\Delta c_1}{(dc_1/dz)} \right|_{z=0}. \quad (9)$$

The quantity ℓ_c^{ef} has the sense of an effective length, which develops upon linear extrapolation of the concentration distributions in the orifice section to values corresponding to the concentrations in the containers. The effective orifice length, calculated from Eqs. (6) and (9), is

$$\ell_c^{ef} = \frac{\pi}{2} R. \quad (10)$$

We will note that this introduction of an λ_c^{ef} , which allows derivation of an expression for the kinetic coefficient L_{22}^0 in a form analogous to L_{22} , is correct only as $Kn \rightarrow 0$.

The kinetic coefficient L_{12} describes development of a mean numerical velocity in the channel under the action of a concentration difference between the ends. The velocity of such mixture motion has been termed the diffusion slip velocity. Consequences of diffusion slip are, for example, osmotic pressure differences on a membrane separating gas or liquid mixtures, and diffusion phoresis of aerosol particles in gas mixtures with inhomogeneous composition. Separation of mixtures during motion in a channel under the action of pressure difference (barodiffusion) is a cross-phenomenon with regard to diffusion slip. In accordance with the Onsager reciprocity principle the kinetic coefficients describing cross-phenomena must be equal.

The existence of diffusion slip in long channels has been demonstrated experimentally in diffusion bridge type devices [10], as well as by indirect measurements of the diffusion baroeffect in a system of two containers [11]. Even in a viscous flow regime calculation of the kinetic coefficient L_{12} cannot be carried out by solution of the Boltzmann equations by the Chapman-Enskog method [9]. Theoretical expressions for L_{12} which agree well with experimental data have been obtained for long channels by solution of the kinetic equations with specified boundary conditions for the distribution function [12]. Since L_{12} is of a diffusion nature, it is convenient to represent it in a form proportional to L_{22} :

$$L_{12} = \sigma c_1 c_2 L_{22}. \quad (11)$$

For long channels the coefficient σ introduced in this manner is called the diffusion slip coefficient. This coefficient should vanish when a difference in component levels disappears. For mixtures of molecules whose characteristics differ little from each other the diffusion slip coefficient can be written in a difference form [12]:

$$\sigma = A \frac{m_1 - m_2}{m_1 + m_2} - B \frac{S_1 - S_2}{S_1 + S_2} + C \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + \varepsilon_2}. \quad (12)$$

To the authors' knowledge the existence of a diffusion slip velocity at an orifice has yet to be established. However, it is reasonable to propose that for mixtures with different masses or molecular collision sections of the components such a velocity should exist. This will be demonstrated experimentally below.

2. Method for Kinetic Coefficient Measurement. To measure the kinetic coefficients of mixture motion in an orifice a method based upon determination of the time dependence of the pressure difference of the diffusion baroeffect [3] was used. The diffusion baroeffect is a phenomenon of disruption of the isobaric nature of the mixing process in a two-reservoir device. It has been studied for gas mixing through long channels and porous media. Below we will consider the case of diffusion through an orifice.

Equations were presented in [3] which related the value of the pressure difference between the volumes $\Delta p(t)$ at time t with the kinetic coefficients of motion of a binary gas mixture. Since the derivation of these equations used no assumptions regarding the form or geometry of the channel, they can also be applied to description of the diffusion baroeffect at an orifice. We will write these equations for a viscous flow regime, where the Knudsen number, equal to the ratio of the mean free path length to the channel diameter, is much less than unity:

$$\frac{\Delta p(t)}{p} = \frac{L_{12}^0}{L_{11}^0} (\exp(E_1 t) - \exp(E_2 t)), \quad (13)$$

$$E_1 = -0,5pdL_{22}^0, \quad E_2 = -2pdL_{11}^0, \quad d = \pi R^2 V^{-1}.$$

The quantities E_1 and E_2 are the reciprocal relaxation times of the two characteristic processes occurring in the two-reservoir device upon mixing of the gases. A decrease in the mixture concentration difference between the volumes occurs with a characteristic time $1/E_1$, inversely proportional to the mutual diffusion coefficient. The pressure difference between the volumes equalizes with a characteristic time $1/E_2$, proportional to the mixture viscosity. If $E_2 \gg E_1$, then in a two-reservoir device a quasisteady state is established, with the pressure difference undergoing practically no change over the course of a time $\sim 1/E_2$, because

of mutual compensation of the hydrodynamic mixture flow, caused by the pressure difference, and an oppositely directed flow, caused by the concentration gradient.

Processing of the experimental dependence $\Delta p(t)$ by the method of least squares can establish the values of the three coefficients L_{ij}^0 appearing in Eq. (13). Since the values of L_{11}^0 are known quite reliably and their measurement is not of interest in the present study, the experimental $\Delta p(t)$ dependences obtained at an orifice were used to generate values of two coefficients: L_{12}^0 and L_{22}^0 .

Using Eqs. (4) and (7), it can easily be proved that the ratio $L_{12}^0/L_{22}^0 \sim Kn^2$, i.e., decreases with increase in the pressure $\sim p^2$. The value of the baroeffect pressure difference $\Delta p(t) \sim pL_{12}^0/L_{22}^0$ and decreases $\sim p^{-1}$. Therefore, from the viewpoint of increased measurement accuracy, it is desirable to perform experiments at the lowest pressure possible. At the same time pressure reduction leads to transition from the viscous flow regime to an intermediate one ($Kn \sim 1$), where Eq. (3) is invalid. The optimum pressures prove to be those corresponding to a viscous slip regime, $0.01 < Kn < 0.15$, where the quantity $\Delta p(t)$ can be recorded with an accuracy sufficient for determination of L_{12}^0 and L_{22}^0 with an uncertainty of $\sim 1\%$. At the same time it is known that the coefficients $L_{ij}(Kn)$ in long channels in this pressure interval have a simple relationship with the value in the viscous regime:

$$L_{ij}(Kn) = L_{ij}(1 - A_{ij}Kn),$$

which corresponds to expansion of $L_{ij}(Kn)$ in a series in the small parameter Kn near the value in the limiting viscous regime ($Kn = 0$). In this expansion we neglect terms proportional to Kn^2 . Nonconsideration of these terms for $Kn < 0.1$ introduces an error of less than 1% , which corresponds to the accuracy of the experimental data. It can be expected that the same relationship characterizes the coefficients for an orifice L_{ij}^0 . Thus, by measuring the value of $L_{ij}^0(Kn)$ for several values of Kn from 0.01 to 0.15, one can determine L_{ij}^0 by linear extrapolation to the value $Kn = 0$. This method of determining L_{ij}^0 gives significantly higher accuracy than measurement in a significantly viscous regime ($Kn < 0.01$), while for mixtures of gases with similar masses, for example He-D₂, in which the value of the baroeffect is small even at $Kn \sim 0.1$, it is the only possible technique.

3. Experimental Apparatus. Baroeffect measurements were performed with apparatus similar to that described in [11]. The equipment was based on a two-reservoir device consisting of two cylindrical volumes (diameter 80 mm, length 30 mm), separated by a 3-mm-thick flange. The sensitive element of a pressure difference sensor (corrugated beryllium bronze membrane 10 μ m thick) together with a film containing the orifice to be studied were attached to the flange. The orifice diameter was measured by two methods: optical, and from gas flow rate in the free molecular regime. Measurements by both methods agreed within the limits of measurement uncertainty and indicated a value of $R = 0.150 \pm 0.001$ mm. The ratio $2R/\ell > 100$, which allows description of the experiments performed with the assumption $\ell = 0$. Two conditions were maintained in attaching the orifice to the apparatus: rigidity of the attachment and smallness ($< 1\%$) of the chamber diffusion resistance as compared to the diffusion resistance of the orifice. Rigidity of the attachment was assured by gluing the film to a copper foil 0.1 mm thick with 4-mm-diameter orifice. The foil was first soldered to the flange such that the center of the orifice in the foil coincided with the center of the orifice in the flange (15-mm diameter).

The pressure difference between the chambers was measured by a capacitance differential micromanometer [13] with sensitivity of $\Delta p/\Delta F = 2.07 \cdot 10^{-5}$ Pa/Hz. Absolute pressures in the apparatus were measured by mercury or oil U-tube manometers. The apparatus was temperature stabilized by a water thermostat at room temperature.

4. Measurement Results. The function $\Delta p(t)$ was determined in the following manner. Nonequilibrium conditions $\Delta p = 0$ and $\Delta c = 1$ were created on the orifice, corresponding to filling of the device volumes by single-component gases at identical pressures. At the moment when mixing begins (opening of the closed device) automatic recording of the micromanometer indications commenced. At some time t_m the value of the pressure difference reaches a maximum $\Delta p_m = \Delta p(t_m)$, after which it slowly falls to the value $\Delta p = 0$ upon complete mixing of the gases. Recording of pressure differences was terminated upon decrease in Δp to a value of $\sim 0.1 \Delta p_m$. The characteristic form of the experimental curves is shown in Fig. 1. The data were obtained for an Ar-Kr mixture at $Kn = 0.046$. The curve shows the dependence of Eq. (13), obtained by processing of the experimental data by the nonlinear

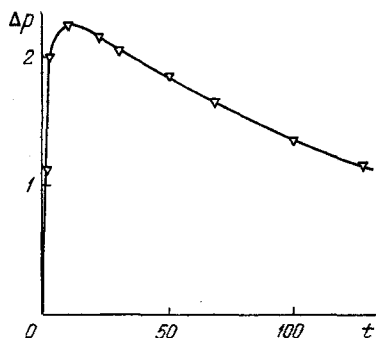


Fig. 1

Fig. 1. Baroeffect pressure difference vs time. Δp , Pa; t , sec.

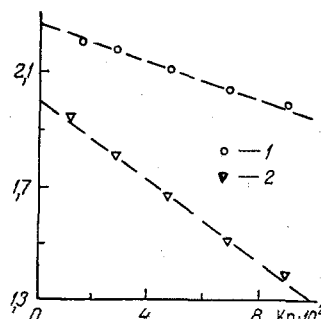


Fig. 2

Fig. 2. Kinetic coefficients of Ar-Kr mixture at orifice. Kn , dimensionless; 1) $L_{22}^0(Kn)p^2 \cdot 10^{-4}$; 2) $L_{12}^0(Kn)p^2 \cdot 10^{-3}$ mPa·sec $^{-1}$.

method of least squares. The value of the kinetic coefficient $L_{11}^0(Kn)$ was calculated by the expression presented in [5]:

$$L_{11}^0(Kn) = \frac{R}{3\pi\eta_{12}}(1 + 3,51Kn),$$

$$Kn = \frac{\sqrt{\pi}}{2} \frac{c_1\delta_2 + c_2\delta_1}{\delta_1\delta_2}, \quad \delta_i = \sqrt{\frac{m_i}{2kT}} \frac{pR}{\eta_i}, \quad i = 1, 2,$$

while the coefficients $L_{12}^0(Kn)$ and $L_{22}^0(Kn)$ were chosen for best agreement of the theoretical Eq. (13) with the experimental data. In the example presented $p^2L_{22}^0 = (2.11 \pm 0.01) \cdot 10^4$ m·Pa/sec, $p^2/L_{12}^0 = (1.68 \pm 0.01) \cdot 10^3$ m·Pa/sec. Similar measurement accuracy was found for the other gas mixtures at $Kn < 0.1$. The uncertainty in measurement of L_{ij}^0 for the He-D₂ mixture increased to 2% because of the smallness of the baroeffect pressure difference in the vapor of these gases. Because of the equal volumes of the apparatus used, during the mixing time a constant gas mixture composition existed in the plane of the orifice with concentrations $c_1 = c_2 = 0.5$. The values obtained for the kinetic coefficients were referred to this concentration value. By repeating the experiment at a different pressure value, values of $L_{12}^0(Kn)$ and $L_{22}^0(Kn)$ corresponding to a different Knudsen number were obtained.

Graphs analogous to that presented in Fig. 2 for an Ar-Kr mixture were constructed for each mixture from the experimental data. In constructing the graphs the fact was considered that the kinetic coefficients L_{12}^0 and L_{22}^0 are proportional to the diffusion coefficient D_{12} , i.e., they depend on pressure $\sim p^{-2}$, and as $Kn \rightarrow 0$ $L_{12}^0 \rightarrow 0$ and $L_{22}^0 \rightarrow 0$. Therefore, the products $L_{12}^0p^2$ and $L_{22}^0p^2$ in the limit of the viscous regime ($Kn \rightarrow 0$) are independent of pressure. The values of these complexes for the viscous regime should then be sought by linear extrapolation of their values at finite Kn to zero Knudsen numbers.

Table 1 presents values of $L_{12}^0p^2$ and $L_{22}^0p^2$ for the gas mixtures studied, determined by the method described above for a temperature $T = 293$ K. These values were then used to calculate the orifice length ℓ_c^{ef} related to diffusion in accordance with Eq. (9) and the diffusion slip coefficient on the orifice σ^0 in accordance with Eq. (11):

$$\sigma^0 = \frac{L_{12}^0p^2}{c_1c_2L_{22}^0p^2}.$$

The values of the mixture diffusion coefficient D_{12} were taken from [14] and are presented in Table 1.

As is evident from the table, within the limits of measurement accuracy the values obtained for ℓ_c^{ef} for all mixtures coincide with the value $\ell_c^{ef} = (\pi/2)R$, obtained in Sec. 1 by solution of the diffusion equation. Some difference in the ℓ_c^{ef} value for the mixture He-D₂ may be related to the relatively higher measurement uncertainty, caused by the smallness of the measured pressure difference. The agreement of the ℓ_c^{ef} values for the various

TABLE 1. Measurement Results

Mixture	$D_{12} \cdot 10^4$, m ² /sec	$L_{12}^0 \rho^2 \cdot 10^{-3}$, m · Pa/sec	$L_{22}^0 \rho^2 \cdot 10^{-4}$, m · Pa/sec	$\frac{l_c^{ef}}{R}$	σ^0	σ
Kr—Xe	0,0725	0,513±0,006	1,26±0,02	1,56±0,02	0,163±0,003	0,191
Ar—Kr	0,131	2,01±0,02	2,27±0,02	1,56±0,02	0,354±0,006	0,368
N ₂ —Ar	0,190	1,70±0,02	3,27±0,03	1,57±0,02	0,208±0,004	0,213
He—D ₂	1,27	-5,60±0,16	20,9±0,6	1,64±0,05	-0,107±0,004	-0,110

mixtures indicates the validity of introducing this parameter. Despite the fact that the theoretical model of two semiinfinite volumes is not identical to the experimental situation (closed device), it was possible to achieve agreement of experimental and theoretical results because of the smallness of the diffusion resistance of the apparatus volumes compared to the diffusion resistance of the orifice.

The diffusion slip coefficients σ^0 of the mixtures studied were found to differ from zero. This indicates the existence of a diffusion slip velocity (diffusion wind) at the orifice. The values of the diffusion slip coefficients measured at the orifice σ^0 are close to the corresponding values for a long channel. The differences that do exist apparently reflect the disappearance of any difference due to interaction of mixture components with the channel wall.

The values obtained for σ^0 can be described approximately with the semi-empirical expression

$$\sigma^0 = 1,13 \frac{m_1 - m_2}{m_1 + m_2} - 0,98 \frac{S_1 - S_2}{S_1 + S_2}.$$

The values of S_i were determined from experimental data on viscosity of the pure gases using the expression

$$S_i^2 = \frac{5}{16} \frac{\sqrt{\pi m_i k T}}{\pi \eta_i}.$$

NOTATION

Δp , pressure difference; Δc , concentration difference; \bar{u}_i , projection on z axis of mean over channel section of velocity u_i of mixture component i; n_i , n, partial and total numerical molecular densities; $c_i = n_i/n$, relative concentration of mixture component i; $L_{ij}(\text{Kn})$, L_{ij} , kinetic coefficients for finite Kn and Kn = 0 respectively; p, pressure; T, temperature; k, Boltzmann's constant; l , channel length; R, radius; η_{12} , η_i , viscosities of mixture and single-component gas i; D_{12} , mutual diffusion coefficient; σ , diffusion slip coefficient; α_p , barodiffusion coefficient; Δl , end correction; λ , free path length; l_p^{ef} , l_c^{ef} , effective orifice lengths for pressure and concentration heads; m_i , S_i^2 , mass and collision cross section for type i molecules; ϵ_i , accommodation coefficient for type i molecules; A, B, C, A_{ij} , constants; t, time; V, chamber volume; Kn, Knudsen number; ΔF , change in indication of device recording Δp . Superscript 0 indicates values at orifice; z, r, variables of cylindrical coordinate system.

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