MEASUREMENT OF KINETIC COEFFICIENTS OF THE ISOTHERMAL MOTION OF A GAS MIXTURE IN CHANNELS WITH ARBITRARY NEGATIVE PRESSURE

V. D. Seleznev

UDC 533.6.011:082.32:541.128.13

A method is proposed for measuring three kinetic coefficients needed to fully describe the motion of a binary gas mixture in channels caused by pressure and concentration gradients in the case of arbitrary negative pressure. Measurement results are also presented.

The rate of many industrial processes connected with solid fuel combustion, catalysis, and adsorption on porous bodies is limited by the delivery of the active component through channels formed by pores filled with an inert gas. The characteristic cross-sectional dimension of the pores d in this case may be either greater or smaller than the mean free path λ in the gas mixture. The velocities of the two gaseous components under these conditions are proportional to the pressure and concentration gradients and are generally characterized by four kinetic coefficients of isothermal diffusion. The nonequilibrium thermodynamics of discontinuous systems [1, 2] gives the following definitions of these coefficients for low-density gases:

$$\omega = c_1u_1 + c_2u_2 = -L_{11}\frac{\Delta p}{L} - L_{12}p\frac{\Delta c_1}{L}, \tag{1}$$

$$u_1 - u_2 = -L_{21}\frac{\Delta p}{L} - L_{22}p\frac{\Delta c_1}{L}, \tag{2}$$

where $c_i = n_i/(n_1 + n_2) = n_i/n$, n_i, u_i are the concentration, numerical density, and longitudinal projection of the mid-channel velocity (in the laboratory coordinate system) of the i-th component; Δp, Δc_1, gradients of pressure and concentration at the ends of a channel of length L; L_{11}, L_{12}, L_{21}, L_{22}, kinetic coefficients of the isothermal motion of the binary mixture in capillary tubes and porous barriers, respectively describing the permeability, diffusion drift (slip), separation, and mixing of the components.

In accordance with the Onsager reciprocal relations, $L_{12} = L_{21}$, so that the matrix of coefficients L_{ij} will contain three independent elements.

A study which examined the motion of gas mixtures in porous media during heterogeneous catalysis [3] used one coefficient instead of three. Here, the effective diffusion coefficient D_{12}^{ef} is related as follows to L_{22}:

$$L_{22} = D_{12}^{ef}/c_1c_2p. \tag{3}$$

It is obviously impossible to correctly predict the velocities of the components in this case. The effect of the first term in the right side of Eq. (1) may prove quite substantial in view of the fact that, in the presence of reactions in the intermediate range, the pressure drop along the pores may be of the same order as the absolute pressure in the system. The customary neglect of the second term in the right side of (1) is correct only for self-diffusion. In the general case of the diffusion of two markedly dissimilar components, this term — accounting for diffusion drift of the mixture — must be considered.

The present work investigates the Knudsen number (Kn = λ/d) dependence of all three kinetic coefficients L_{ij}, giving a complete description of the motion of a binary gas mixture in channels. A method is proposed and realized for simultaneous measurement of the coeffi-

---

S. M. Kirov Ural Polytechnic Institute, Sverdlovsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 41, No. 4, pp. 702-707, October, 1981. Original article submitted June 23, 1980.

0022-0841/81/4104-1142$07.50

cients L_{11}, L_{12}, and L_{22}. The tests were conducted in the intermediate range of Knudsen numbers (Kn = 0.05-10) on five pairs of gases He–Ar, H_2–Ar, H_2–He, H_2–Ne, and He–Ne in a round glass capillary tube. As far as we know, no other tests have been conducted to determine L_{12} and L_{22} in the intermediate range for a channel of regular geometry.

The proposed method of measuring the coefficients L_{ij} is based on recording of the time dependence of the diffusion barrier effect which arises in the mixing of the components in a system of two columns connected by a capillary tube. To analytically connect the empirical time dependence of the barrier effect with the kinetic coefficients L_{ij}, we will examine the mixing of gases in a system of two columns with volumes $V_I = V_{II}$ connected by a capillary tube of length L and radius R. Ignoring the diffusion resistance of the columns, we may write the law of conservation of the number of molecules of each component in the respective columns as follows:

$$\frac{dn_{1\,\mathrm{I}}}{dt} = -\frac{\pi R^2}{V_{\mathrm{I}}} n_{01}u; \quad \frac{dn_{2\,\mathrm{II}}}{dt} = \frac{\pi R^2}{V_{\mathrm{II}}} n_{02}u_2. \tag{4}$$

Substituting the expressions for the velocities u_i from (1) and (2) into (4) and solving the resulting system of equations with the initial conditions $n_{1\mathrm{I}}(0) = n_{2\mathrm{II}}(0) = 2n_{01} = 2n_{02}$, we easily obtain

$$\frac{\Delta p\,(t)}{p} = a\,[\exp(S_1 t) - \exp(S_2 t)], \tag{5}$$

$$a = \frac{2\pi R^2 p L_{12}}{LV_1(S_2 - S_{\mathrm{I}})}, \tag{6}$$

where

$$S_{1,2} = \frac{\pi R^2 p}{V_{\mathrm{I}}\, L}\left[-L_{11} - \frac{L_{22}}{4} \pm \sqrt{L_{12}^2 + \left(L_{11} - \frac{1}{4}L_{22}\right)^2}\right].$$

In the tests, the pressure gradient in the columns Δp_i at successive moments of time t_i was recorded by a frequency–capacitance pressure transducer, the operation of which was described in detail in [4]. A basic diagram of the experimental unit is presented in [5]. Proceeding on the basis of the condition of minimum deviation of the theoretical relation (5) from the experimental set of points $\Delta p_i(t_i)$, we used the nonlinear method of least squares to determine the parameters a, S_1, and S_2. In accordance with (6), we expressed the kinetic coefficients L_{ij} through the free parameters of Eq. (5) as follows:

$$L_{11} = -\frac{(S_2 + S_1)V_{\mathrm{I}}\, L}{4\pi R^2 p} + \frac{1}{2}\sqrt{\frac{(S_1 - S_2)^2 V_{\mathrm{I}}^2 L^2}{4\pi^2 R^4 p^2} - L_{12}^2},$$

$$L_{22} = -\frac{(S_2 + S_1)V_{\mathrm{I}}\, L}{\pi R^2 p} - \sqrt{\frac{(S_1 - S_2)^2 V_{\mathrm{I}}^2 L^2}{\pi^2 R^4 p^2} - 4L_{12}^2},$$

$$L_{12} = \frac{a(S_2 - S_1)LV_{\mathrm{I}}}{2\pi R^2 p}.$$

The random error of L_{ij} depends on the magnitude of the barrier effect and varies from 2% at Kn = 0.3-3 to 7% with approach of the viscous and free-molecular limits.

We used the above method to measure three coefficients L_{ij} on the gas pairs He–Ar, H_2–Ar, H_2–He, He–Ne, and H_2–Ne at a concentration c = 0.5 and temperature T = 296°K in the range of numbers Kn = 0.03-10. The test results are shown in Fig. 1 in the form of corrected coefficients independent of the channel radius. This coefficient correction was performed in accordance with the following formulas:

$$L_{11}^{\mathrm{cr}} = \frac{L_{11}}{L_{11}^{\mathrm{Kn}}}; \quad L_{12}^{\mathrm{cr}} = \frac{L_{12}}{L_{12}^{\mathrm{Kn}}}; \quad L_{22}^{\mathrm{cr}} = \frac{L_{22}}{L_{22}^{\mathrm{Kn}}}, \tag{7}$$

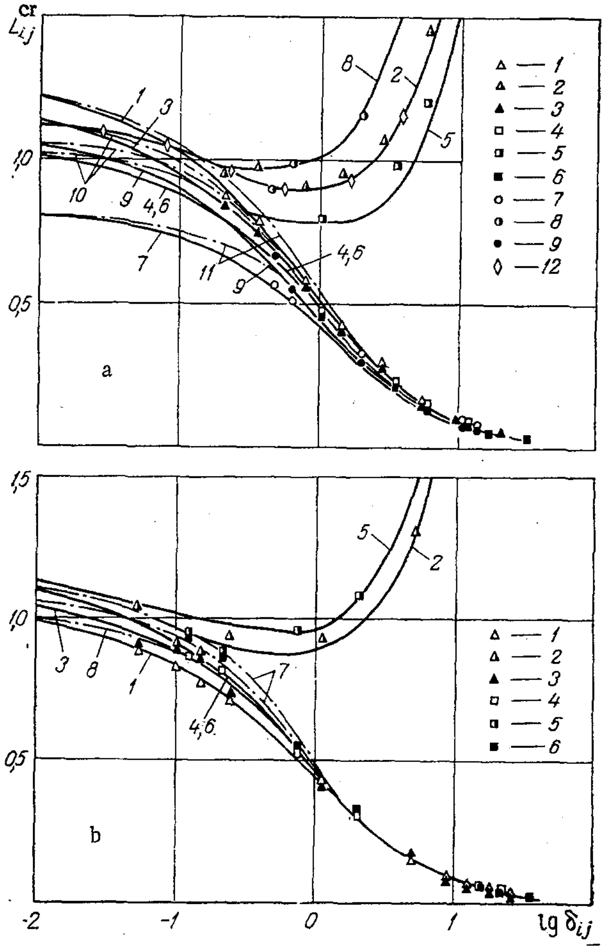

Fig. 1. Corrected kinetic coefficients of the isothermal motion of a binary mixture in a capillary tube as a function of the logarithm of the reciprocal Knudsen number: a) 1 — L_{12}^{cr}; 2 — L_{11}^{cr}; 3 — L_{22}^{cr} for the He—Ar pair; 4 — L_{12}^{cr}; 5 — L_{11}^{cr}; 6 — L_{22}^{cr} for the H_2—Ar pair; 7 — L_{12}^{cr}; 8 — L_{11}^{cr}; 9 — L_{22}^{cr} for the H_2—He pair; 10, 11 — Eqs. (9), (10); 12 — data from [8]; b) 1 — L_{12}^{cr}; 2 — L_{11}^{cr}; 3 — L_{22}^{cr} for the H_2—Ne pair; 4 — L_{12}^{cr}; 5 — L_{11}^{cr}; 6 — L_{22}^{cr} for the He—Ne pair; 7, 8 — Eqs. (9), (10).

where

$$L_{11}^{\text{Kn}} = \frac{2}{3}\frac{R}{p}(c_1 v_{t1} + c_2 v_{t2});$$

$$L_{12}^{\text{Kn}} = \frac{2}{3}\frac{R}{p}(v_{t1} - v_{t2}); \quad v_{ti} = \sqrt{\frac{8kT}{\pi m_i}};$$

$$L_{22}^{\text{Kn}} = \frac{2}{3}\frac{R}{p}\left(\frac{v_{t1}}{c_1} + \frac{v_{t2}}{c_2}\right);$$

m_i is the mass of a molecule of the i-th component.

The reciprocal Knudsen number of the mixture δ_{ij} was calculated from the relation [6]

$$\delta_{ij} = \frac{Rp}{\eta_1\sqrt{\frac{2kT}{m_1}} + \eta_2\sqrt{\frac{2kT}{m_2}}}, \tag{8}$$

where η_i is the partial viscosity of the i-th component [7].

For comparison with the test results obtained here, Fig. 1a shows experimental values of L_{11}^{cr} obtained in [8] by direct measurement of the rate of flow of an He—Ar mixture under similar conditions. The good agreement between the empirical results confirms the correctness of the method proposed in the present work for simultaneously determining three kinetic coefficients of isothermal diffusion. Use of the method can be recommended for studying the motion of a binary gas mixture in various types of porous membranes. The solid lines in Fig. 1 denote the results of numerical calculation of the coefficients L_{ij}^{cr} [9] with allowance for incomplete accommodation of the molecules on the surface [10]. The discrepancy between the theoretical and experimental values of L_{ij}^{cr} for the investigated pairs of gases does not exceed the experiment error.

In determining the velocity of the components of a binary mixture in porous media at $Kn \sim 1$, a semiempirical formula was adopted for the effective interdiffusion coefficient D_{12}^{ef} [3]. Using the notation employed here in Eq. (3), this formula for D_{12}^{ef} converts to the form

$$\frac{1}{L_{22}}=\frac{1}{L_{22}^{V}}+\frac{1}{L_{22}^{Kn}}, \tag{9}$$

where L_{22}^{V} and L_{22}^{Kn} are the viscous and free-molecular limits of the coefficient L_{22}.

It would be of interest to check the validity of this expression. The dot—dash line in Fig. 1 shows the results of calculations with Eqs. (9) and (7) with allowance for incomplete accommodation [10]. The discrepancy between the semiempirical values of L_{12}^{cr} and the nearly identical theoretical and empirical values is greatest in the region of $\delta_{ij} = 0.2$ and reaches 10%.

The kinetic coefficient function $L_{12}^{cr}(\delta_{ij})$ is similar to the function $L_{22}^{cr}(\delta_{ij})$ for the gas pairs investigated. Making use of this fact, we may propose a formula similar to (9) to approximately calculate $L_{12}(\delta_{ij})$

$$\frac{1}{L_{12}}=\frac{1}{L_{12}^{V}}+\frac{1}{L_{12}^{Kn}}, \tag{10}$$

where $L_{12}^{V} = \sigma D_{12}/P$, L_{12}^{Kn} are the viscous and free-molecular limits of L_{12}; σ is the coefficient of diffusion slip [9].

Comparison with the strictly theoretical results and the experiment shows that Eq. (10) is valid.

NOTATION

R, radius of capillary tube; ω, mean velocity; λ, mean free path length; c_i, n_i, u_i, molar fraction, numerical density, and mean (across the channel) velocity of the i-th component; Δp, Δc, gradients of pressure and molar fraction at ends of channel; L, channel length; L_{ij}, kinetic coefficients; D_{12}^{ef}, effective interdiffusion coefficient; V_I, V_{II}, volumes of columns; n_{ij}, n_{oi}, density of molecules of the i-th component in the j-th column and in the middle of the capillary tube; $\delta_{ij} = (Kn)^{-1}$, reciprocal Knudsen number of the mixture; η_i, partial viscosity of the i-th component of the mixture; σ, coefficient of diffusion slip.

LITERATURE CITED

1. S. R. De Groot and P. Mazure, Non-Equilibrium Thermodynamics, Elsevier (1962).
2. V. D. Seleznev, P. E. Suetin, and E. V. Kalinin, "Isothermal diffusion of a gas or liquid mixture in a channel," Manusc. dep. at VINITI (All-Union Institute of Scientific and Technical Information), 1977, No. 4169-77. Dep. Ref. Zh. Fiz. (1978), 2I92DEP.
3. G. K. Boreskova (editor), Porous Structure of Catalysts and Transport Processes in a Heterogeneous Catalyst [in Russian], Nauka, Novosibirsk (1970).
4. S. F. Borisov, B. A. Kalinin, B. T. Porodnov, and P. E. Suetin, "Micromanometer with digital reading," Prib. Tekh. Eksp., No. 4, 209-213 (1972).
5. P. E. Suetin and V. D. Seleznev, "Diffusion barrier effect at low temperatures," in: Diffusion in Gases and Liquids [in Russian], Glavpoligrafprom Kazakhskoi SSR, Alma Ata (1972), pp. 37-44.

6. V. D. Seleznev, P. E. Suetin, and N. A. Smirnov, "Separation of a binary gas mixture throughout the Knudsen number range," Zh. Tekh. Fiz., 45, No. 7, 1499-1502 (1975).
7. V. Zhdanov, Yu. Kagan, and A. Sazykin, "Effect of viscous momemtum transfer on diffusion in a gas mixture," Zh. Eksp. Teor. Fiz., 42, No. 3, 857-866 (1962).
8. A. A. Tarin, "Investigation of the diffusion separation and conductivity of gas mixtures in a capillary tube with their efflux into a vacuum," Author's Abstract of Candidate's Dissertation, Physical-Mathematical Sciences, Sverdlovsk (1978).
9. N. D. Pochuev, V. D. Seleznev, and P. E. Suetin, "Flow of a binary gas mixture with arbitrary accommodation of a tangential impulse," Zh. Prikl. Mekh. Tekh. Fiz., No. 5, 37-41 (1974).
10. V. D. Akin'shin, S. F. Borisov, B. T. Porodnov, and P. E. Suetin, "Experimental study of the flow of low-density gases in a capillary sieve at different temperatures," Zh. Prikl. Mekh. Tekh. Fiz., No. 2, 45-49 (1974).

HYDRODYNAMIC CHARACTERISTICS OF A FLAT-FLAME INJECTOR TORCH WITH A VORTEX INJECTOR

R. K. Narkhodzhaev and E. R. Kasparov

UDC 662.951.23/27:532.5

The optimum twist of an injected flow is established through studies of a function describing the hydrodynamic characteristics of a flat-flame burner.

Existing designs of torches do not meet the requirements of flame decontamination technology. We have developed a new design of flat-flame injector torch with a vortex injector [1, 2]. Presented below are the results of studies of the hydrodynamic characteristics of the proposed torch, which consists of a cylindrical mixing chamber and a slit diffuser. The mixing chamber is equipped with rotary nozzles which allow the twist parameter to be changed from 0 to 2.84.

It is well known that the width of the jet increases more rapidly in the case of swirled flow [3]. Allowing for this, we investigated the velocity distribution in the outlet section of the mixing chamber diffusion nozzle in relation to the twist parameter n for values of n equal to 0, 1, and 2. Velocity curves recorded at these parameter values were compared with the curves obtained with axial location of a nozzle of the same capacity (Fig. 1). Analysis of the curves shows that the velocity of the mixture leaving the diffuser is extremely nonuniform in the case of axial location of the nozzle. Here, the velocity distribution in the outlet section of the mixing chamber is uneven, with the maximum occurring on the torch axis. The existence of this maximum leads to deformation of the flow by the diffuser walls a considerable distance from the outlet section of the mixing chamber and results in a velocity maximum in the axial region of the diffuser outlet section.

The character of the velocity nonuniformity changes little for a slightly twisted flow ($0 < n < 1$), but there is an increase in velocity in the peripheral zones of the slit, with a maximum on the axis. The velocity field smooths out at $n = 1$, and at $n = 2$ the flow may be considered uniform at the slit outlet, although it has a certain axial trough in the mixing chamber. This can be explained by the fact that the rate of development of the cylindrical flow increases with an increase in n. It begins to be deformed by the walls of the diffuser at the outlet of the mixing chamber and gradually degenerates into a plane flow with a uniform velocity distribution. A further increase in the parameter n leads to an increase in the size of the velocity trough in the mixing chamber section, and a nonuniform flow — with the velocity maximum in its peripheral zones — is produced at the slit outlet.

It follows from the results of the experiment that the optimum value of the twist parameter for the model being examined, ensuring uniform flow of the mixture from the slit, should be found within the range $n = 2 \pm 0.1$. The uniformity of the flow in the outlet sec-

---

Mid-Asian Branch of the All-Union Scientific-Research Institute of Gas Utilization in the National Economy and of Underground Storage of Petroleum, Petroleum Products, and Liquefied Gas (VNIIpromgaz). Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 41, No. 4, pp. 708-711, October, 1981. Original article submitted September 16, 1980.

0022-0841/81/4104-1146$07.50