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MOTION OF A BINARY GAS MIXTURE IN A POROUS MEMBRANE WITH A STRAIGHT CHANNEL

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The diffusional baric effect at a porous barrier with a straight channel is investigated theoretically and experimentally with an arbitrary ratio of the channel and pore diameter to the free path length of molecules of the gas mixture.

Porous membranes with straight channels of diameter considerably differing from the characteristic dimensions of the pores are widely used in various technological processes. The straight channels are used for sharp reduction in the diffusional and hydraulic resistance in drying friable and porous materials, and with heterogeneous catalysis in the cores of the thermal tubes. A finely porous membrane with broad penetrating channels combines high specific surface with large conduction. Expansion of the region of application of such membranes is delayed by the limited study of their properties, especially at intermediate Knudsen numbers.

In the present work, a method of determining the effective kinetic coefficients of motion of binary gas mixtures in such membranes over a wide range of Knudsen membranes is described and experimentally tested.

1. Effective Kinetic Coefficients for a Gas of Any Degree of Rarefaction

Consider the isothermal motion of a binary gas mixture under the action of pressure and concentration differences in a porous cylindrical membrane of length ℓ and radius R, along the axis of which there is a straight cylindrical channel of radius r_0 (Fig. 1). Suppose that the lateral surface of the membrane is impermeable, while the mean pore radius is equal to r. With variation in gas-mixture pressure in the membrane with $r \ll r_0$, the following combinations of flow conditions may be realized: $r \ll r_0 \ll \lambda$, free-molecular conditions in the whole membrane; $r \ll r_0 \sim \lambda$, intermediate conditions in the straight channel and free-molecular conditions in the pores; $r \ll \lambda \ll r_0$, viscous flow conditions in the straight channel and free-molecular conditions in the pores; $r \sim \lambda \ll r_0$, viscous conditions in the straight channel and intermediate conditions in the pores; $\lambda \ll r \ll r_0$, viscous flow conditions in the whole membrane.

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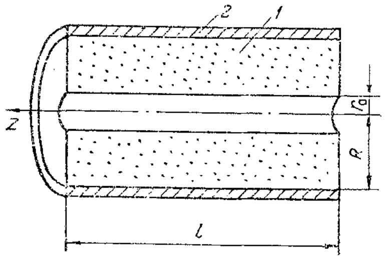

Fig. 1

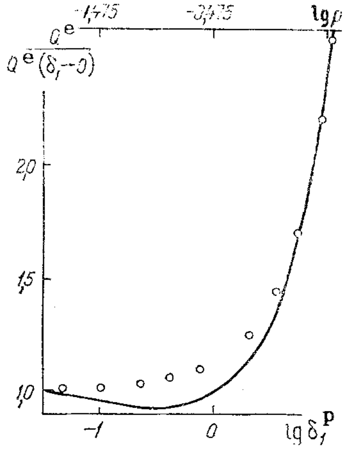

Fig. 2

Fig. 1. Geometry of the membrane: 1) porous section; 2) impermeable shell.

Fig. 2. Dependence of flow rate on the inverse Knudsen number; δ_1^p and $Q^e/Q^e(\delta_1 \to 0)$ are dimensionless.

For such membranes, it is possible to obtain a relatively simple method of calculating the effective kinetic coefficients of isothermal motion of the binary mixture, under the condition that the membrane length ℓ is much larger than its radius R. In this case, the ratio of characteristic times of establishing equilibrium and a steady state along τ_ℓ and across τ_R the membrane is considerably larger than unity (i.e., $\tau_\ell/\tau_R \gg 1$). This allows one to hope that, with steady motion of the mixture in an arbitrary, infinitely small, section of the membrane enclosed between the planes z and z + dz, conditions of local equilibrium – characterized by negligibly small transverse concentration and pressure differences in comparison with the corresponding longitudinal values – are observed. With local equilibrium, the longitudinal pressure and concentration distributions in the straight channel and the porous part of the membrane will coincide. In this case, the longitudinal fluxes of the components in any element dz of the membrane must be considerably larger than the transverse fluxes. Therefore, it may be expected that, for these fluxes, the expressions of nonequilibrium thermodynamics for homogeneous membranes with an impermeable lateral surface will be approximately valid.

In the straight channel, the projections of the mean velocities of motion of the components of the binary gas mixture at the z axis, $\bar{u}_{1z}^{ch}$ and $\bar{u}_{2z}^{ch}$ are related to the pressure $(dp/dz)^{ch}$ and concentration $(dc_1/dz)^{ch}$ gradients producing them through the kinetic coefficients L_{ij}^{ch} [1]

$$\bar{u}_{1z}^{ch}c_1 + \bar{u}_{2z}^{ch}c_2 = -L_{11}^{ch}\left(\frac{dp}{dz}\right)^{ch} - L_{12}^{ch}p\left(\frac{dc_1}{dz}\right)^{ch}, \tag{1}$$

$$\bar{u}_{1z}^{ch} - \bar{u}_{2z}^{ch} = -L_{21}^{ch}\left(\frac{dp}{dz}\right)^{ch} - L_{22}^{ch}p\left(\frac{dc_1}{dz}\right)^{ch}.$$

For the porous part of the membrane, the analogous expressions take the form

$$\bar{u}_{1z}^{p}c_1 + \bar{u}_{2z}^{p}c_2 = -L_{11}^{p}\left(\frac{dp}{dz}\right)^{p} - L_{12}^{p}\left(\frac{dc_1}{dz}\right)^{p}p, \tag{2}$$

$$\bar{u}_{1z}^{p} - \bar{u}_{2z}^{p} = -L_{21}^{p}\left(\frac{dp}{dz}\right)^{p} - L_{22}\left(\frac{dc_1}{dz}\right)^{p}p.$$

If the local-equilibrium condition holds in an element dz of the membrane, the concentration and pressure gradients over the porous medium and over the channel will be

$$\left(\frac{dp}{dz}\right)^{p} = \left(\frac{dp}{dz}\right)^{ch} = \frac{dp}{dz}, \quad \left(\frac{dc_1}{dz}\right)^{p} = \left(\frac{dc_1}{dz}\right)^{ch} = \frac{dc_1}{dz}.$$

In this case, effective kinetic coefficients L_{ij}^M may be introduced for the membrane, and relations of the same form as Eqs. (1) and (2) may be written

$$\bar{u}^M_{1z} c_1 + \bar{u}^M_{2z} c_2 = -L^M_{11} \frac{dp}{dz} - L^M_{12} p \frac{dc_1}{dz},$$

$$\bar{u}^M_{1z} - \bar{u}^M_{2z} = -L^M_{21} \frac{dp}{dz} - L^M_{22} p \frac{dc_1}{dz}.$$

The relation between the effective kinetic coefficients of the membrane L_{ij}^M and the kinetic coefficients L_{ij}^p and L_{ij}^{ch} is determined by the condition that the flux of gas mixture through the membrane is equal to the sum of fluxes through its porous section and through the straight channel. A relation may be found between L_{11}^M, L_{11}^{ch}, and L_{11}^p for the motion of a single-component gas under the action of the pressure difference. The fluxes of gas mixture through the membrane Q^M, the channel Q^{ch}, and the porous medium Q^p are determined by the expressions

$$Q^M = S^M n L^M_{11} \frac{dp}{dz}, \quad Q^{ch} = S^{ch} n L^{ch}_{11} \frac{dp}{dz}, \quad Q^p = S^p n L^p_{11} \frac{dp}{dz}.$$

From the conditions $Q^M = Q^p + Q^{ch}$ and $S^M = S^{ch} + S^p$, it follows that

$$L^M_{11} = \frac{S^{ch} L^{ch}_{11} + S^p L^p_{11}}{S^{ch} + S^p}.$$

Performing analogous calculations for the other coefficients L_{ij}^M, it is simple to obtain a general expression for determining L_{ij}^M in the form

$$L^M_{ij} = \frac{S^{ch} L^{ch}_{ij} + S^p L^p_{ij}}{S^{ch} + S^p}. \tag{3}$$

The effective kinetic coefficients allows the fluxes of gas-mixture components through the membrane and the distribution of concentration and pressure fields in the membrane to be determined, and allow the separation of the mixture on passing through the membrane and the diffusional baric effect arising there to be described. The methods of solving these problems are analogous to those developed for homogeneous membranes [2].

Calculation of L_{ij}^M from Eq. (3) requires a knowledge of the kinetic coefficients of motion of the gas mixture in the straight channel and the porous medium.

2. Kinetic Coefficients of Motion of the Gas Mixture in the Straight Channel

The kinetic coefficients $L_{ij}(\delta_{12})$ characterizing the motion of a binary mixture of gases in a channel of radius r_0 with arbitrary Knudsen numbers Kn were calculated in [3]. The expressions for their calculation are fairly complex, and are not always convenient for practical use. It often proves sufficient to know the simple superpositional formulas giving accurate values of $L_{ij}(\delta_{12})$ within the limits of the viscous and free-molecular flow conditions and values in satisfactory agreement with experiment in the intermediate region [2]. These expressions are given for a long cylindrical channel.

The motion of a gas mixture under the action of a pressure gradient is described by a coefficient $L_{11}(\delta_{12})$ of the form

$$L_{11}(\delta_{12}) = \frac{r_0^2}{8\eta_{12}} + \frac{2r_0}{3p}(c_1 v_{1t} + c_2 v_{2t}) \frac{1 + 2\delta_{12}}{1 + 2.45\delta_{12}}, \quad \text{if} \quad \delta_{12} \leqslant 5; \tag{4}$$

$$L_{11}(\delta_{12}) = \frac{r_0^2}{8\eta_{12}}\left(1 + \frac{4}{\delta_{12}}\right), \quad \text{if} \quad \delta_{12} > 5,$$

$$\delta_{12} = \frac{\delta_1 \delta_2}{c_2\delta_1 + c_1\delta_2}, \quad \delta_i = \frac{p r_0}{\eta_i}\sqrt{\frac{m_i}{2kT}}, \quad v_{it} = \sqrt{\frac{8kT}{\pi m_i}}, \quad i = 1,\ 2. \tag{5}$$

The coefficients $L_{21}(\delta_{12})$ and $L_{12}(\delta_{12})$ are related to the cross phenomena of separation of the gas mixture on passing through the membrane and the appearance of a pressure difference between the volumes in gas mixing through a membrane (diffusional baric effect). In accordance with the Onsager reciprocity rule, $L_{21}(\delta_{12}) = L_{12}(\delta_{12})$. In viscous, free-molecular, and intermediate flow conditions of the gas, the values of these coefficients are

$$L_{12}(\delta_{12}\to\infty)=\frac{\sigma D_{12}}{p},\quad L_{12}(\delta_{12}\to 0)=\frac{2r_0}{3p}(v_{1t}-v_{2t}),$$
$$\frac{1}{L_{12}(\delta_{12}\sim 1)}=\frac{1}{L_{12}(\delta_{12}\to 0)}+\frac{1}{L_{12}(\delta_{12}\to\infty)}. \tag{6}$$

The most rigorous expressions for calculating the diffusional-slip coefficient σ were obtained in [4]. The value of σ may be approximately calculated from the formula

$$\sigma=1.17\frac{m_1-m_2}{m_1+m_2}-0.93\frac{d_1-d_2}{d_1+d_2}.$$

Gas mixing under the action of the concentration difference is described by the kinetic coefficient $L_{22}(\delta_{12})$ calculated from the formula

$$L_{22}(\delta_{12}\to\infty)=\frac{D_{12}}{c_1c_2p},\quad L_{22}(\delta_{12}\to 0)=\frac{2r_0}{3p}\left(\frac{v_{1t}}{c_1}+\frac{v_{2t}}{c_2}\right),$$
$$\frac{1}{L_{22}(\delta_{12}\sim 1)}=\frac{1}{L_{22}(\delta_{12}\to 0)}+\frac{1}{L_{22}(\delta_{12}\to\infty)}. \tag{7}$$

3. Kinetic Coefficients of Motion of a Gas Mixture in a Porous Body

The kinetic coefficients L_{ji}^{p} describing the motion of gas mixture in a porous medium may be determined on the basis of some model of the medium. The most widespread models take the form of a set of parallel capillaries or spherical filling. The first model is preferable for the present problem, since it allows the calculation of L_{ji}^{p} to be reduced to the use of Eqs. (4)-(7) for a straight channel. In the simplest version of identical capillaries, this model contains two free parameters: the radius r_s and the number of channels N per unit area of the porous medium. The use of experimental data on the pressure dependence of the gas flow rate through a sample of a homogeneous porous medium is proposed for their determination. Experimental data in the form of the ratio of the flow rate with arbitrary pressure to the flow rate in free-molecular conditions (reduced flow rate) must be compared to the theoretical dependence of the reduced gas flow rate in the capillary on the rarefaction parameter $\delta_1^{p} = 2r_s/\lambda$ (continuous curve in Fig. 2) [5]. The radius of capillaries of the model r_s may be interpreted as the mean hydraulic radius of the pores. It is selected from the condition of best description of the experimental dependence by the theoretical curve. The specific number of channels N may be determined from the condition of agreement between the free-molecular flow rate according to a model set of capillaries of known radius r_s and the experimental flow rate through the sample $Q^{e}(\delta_1^{p} \to 0)$ of cross-sectional area S^{p} according to the formula

$$N=\frac{Q^{e}(\delta_1^{p}\to 0)\,3l}{2\pi r_s^3 S^{p}}\sqrt{\frac{\pi m}{8kT}}.$$

With known N and r_s, the kinetic coefficients of the porous medium L_{ij}^{p} are determined from the formulas analogous to Eqs. (4)-(7), with the replacement of r_0 by r_s

$$L_{ij}^{p}=NL_{ij}(r_s).$$

This method of finding L_{ij}^{p} with arbitrary Kn^{p} requires the selection of two parameters whose values are independent of the flow conditions of the gas. This favorably distinguishes the present method from that proposed by Deryagin [6], in which it was proposed to determine various sets of empirical coefficients for free-molecular, intermediate, and viscous flow conditions.

4. Diffusional Baric Effect at a Porous Membrane with a Straight Channel

To test the validity of the assumption of local equilibrium in the element of membrane length and the efficiency of the given algorithm for calculating L_{ij}^{M}, experiments are under-

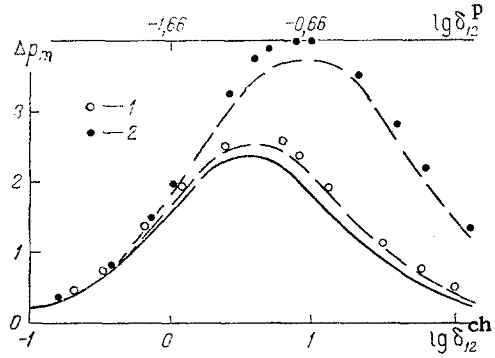

Fig. 3

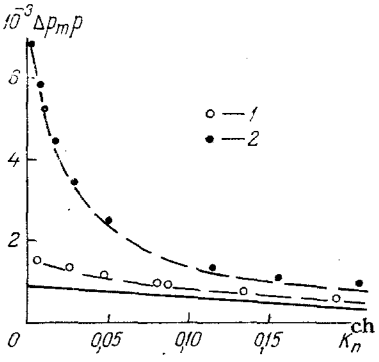

Fig. 4

Fig. 3. Baric effect in a Kr–Xe mixture: 1, 2) membranes with $S^P/S^{ch} = 0.16$ and 2.03, respectively. The dashed curves correspond to calculation from Eqs. (4)-(8) and the continuous curves to calculation for a channel with $r_0 = 0.163$ mm with impermeable walls. Δp_m, Pa; $\delta_{12}{}^{ch}$, $\delta_{12}{}^{P}$ are dimensionless.

Fig. 4. Baric effect for a Kr–Xe mixture in viscous conditions: 1, 2) membranes with $S^P/S^{ch} = 0.16$ and 2.03. The dashed curves correspond to calculation from Eqs. (4)-(8) and the continuous curves to calculation for a channel with $r_0 = 0.163$ mm with impermeable walls. Δp_{mp}, Pa^2; Kn^{ch} is dimensionless.

taken to measure the diffusional baric effect at porous membranes with a straight channel over a wide range of Knudsen numbers. The diffusional baric effect consists in the appearance of a pressure difference between volumes connected by a membrane and initially filled to the same pressure with gas mixtures of different composition. The pressure difference appears in the course of gas mixing on account of the difference in mean rates of penetration of the mixture components through the membrane.

As is known [2], the maximum pressure difference Δp_m and the time for its attainment t_m are determined by all three kinetic coefficients of gas-mixture motion in the membrane

$$\frac{\Delta p_m}{p} = \frac{2p\beta_0 L_{12}^{M}}{\beta_2 - \beta_1}\left(\exp(\beta_1 t_m) - \exp(\beta_2 t_m)\right),$$

$$t_m = \frac{1}{\beta_1 - \beta_2}\ln\frac{\beta_2}{\beta_1},\quad \beta_0 = \frac{S^{M}}{lV}, \tag{8}$$

$$\beta_{1,2} = p\beta_0\left(-L_{11}^{M} - 0.25L_{22}^{M} \pm \sqrt{(L_{12}^{M})^2 + (L_{11}^{M} - 0.25L_{22}^{M})^2}\right).$$

The possible errors in determining any of the coefficients must lead to errors in Δp_m. Therefore, measurement of the diffusional baric effect may be used for the required verification.

Experiments are undertaken on two membranes differing in the cross-sectional area of the porous section. The membranes of required geometry are prepared by sintering nickel powder. A steel wire of diameter 0.3 mm is laid along the sample axis before sintering. After sintering, the membrane pores are filled with rosin, and then the steel core is removed. The radius of the straight channel is $r_0 = 0.163$ mm. Then the resin in the pores adjacent to the channel is dissolved by passing acetone through the channel. The time for solution is determined by the thickness of the porous section of membrane free from rosin. For the two membranes used in the experiment, the cross-sectional area of the porous medium is $S_1{}^P = 0.16S^{ch}$ and $S_2{}^P = 2.03S^{ch}$, where $S^{ch} = \pi r_0{}^2$. The porosity of the membrane and the hydraulic radius of the pores is determined for a control sample without a straight channel obtained under the same sintering conditions, from the same powder. The experimental and theoretical (continuous curve in Fig. 2) dependences, when $\log\delta_1{}^P > 1$, the relation between the gas pressure and the inverse Knudsen number in the pores $\delta_1{}^P$ is established:

$$\lg\delta_1^{P} = \lg p + 0.475.$$

Using Eq. (5), the hydraulic pore radius is determined: $r_s = 3.6$ μm. The porosity of the control sample $\Pi = 0.52$ is determined by a gravimetric method. The number of channels per unit area of the porous medium may conveniently be related to the porosity Π and hydraulic radius r_s as follows:

$$N\pi r_s^2 = AS^p\Pi.$$

The fitting factor $A = 0.2$ is determined from the helium flow rate in free-molecular conditions.

The measurements are performed on an apparatus analogous to that described in [7], consisting of two identical volumes connected by a membrane. The pressure in the inflow system is measured by a mercury or oil U-tube manometer. The pressure difference between the chamber volumes is regulated by a capacitative differential micromanometer [8].

5. Comparison of Theory and Experiment for Diffusional Baric Effect

In Fig. 3, experimental data are shown for the magnitude of the baric effect Δp_m arising in mixing Kr and Xe through a porous membrane with a straight channel. The inverse Knudsen numbers of the straight channel and the porous medium $\delta_{12}{}^{ch}$ and $\delta_{12}{}^{p}$ are plotted along the horizontal axis. The light and dark points show the experimental data obtained for membranes with a ratio of the porous section to the area of the straight channel $S^p/S^{ch} = 0.16$ and $S^p/S^{ch} = 2.03$, respectively. The continuous curves show Δp_m as calculated from Eq. (8) for a straight channel of radius $r_0 = 0.163$ mm with impermeable walls. The dashed curves in Fig. 3 correspond to calculation of Δp_m in complex membranes with the given thicknesses of the porous layer from the effective kinetic coefficient of the membrane $L_{ij}{}^{M}$.

It is evident from a comparison of the theoretical and experimental values that, with any flow conditions and ratio of the cross-sectional areas of the porous section and the straight channel, the deviation of the theoretical predictions from experiment is no more than 10%. The difference is a maximum at intermediate $\delta_{12}{}^{ch}$. The coincidence of the experimental curves of Δp_m for both membranes and the channel with impermeable walls in the region where $\log \delta_{12}{}^{ch} < -0.6$ shows that, in the region of free-molecular flow, the baric effect does not depend on the geometry in all the channels of the membrane.

The greatest deviation in the behavior of Δp_m for membranes of the given type from the analogous quantity for a channel with impermeable walls is observed in the realization of intermediate or viscous flow conditions in the pores. In this region, it is convenient to compare the dependences of $\Delta p_m p$ on Kn^{ch} determined from r_0 (Fig. 4). A channel with impermeable walls gives a linear relation between the quantities. Experiments on a porous membrane with a channel lead to qualitatively different results. As is evident, three characteristic sections may be distinguished in the behavior of $\Delta p_m p$ for complex membranes (points in Fig. 4):

a) $Kn^{ch} > 0.1$, where the behavior of $\Delta p_m p$ for a straight channel and a complex membrane are practically parallel, i.e., the properties of the membrane are determined by the properties of the straight channel;

b) $Kn^{ch} < 0.02$, where a linear dependence of $\Delta p_m p$ on Kn^{ch} is observed with a slope significantly different from that of the straight line calculated for a channel with impermeable walls. On this section, the influence of the porous component on the membrane properties appears more strongly and is more considerable as the cross-sectional area of the porous medium increases;

c) $0.02 < Kn^{ch} < 0.1$, a transitional section corresponding to intermediate flow conditions in the pores, in combination with viscous flow in the straight channel.

The theoretical curves calculated from the effective kinetic coefficients (dashed curves in Fig. 4) give good predictions of the magnitude of the baric effect of the membrane, which confirms the basis for the assumption of local equilibrium in an element of membrane length used in deriving expressions for calculating $L_{ij}{}^{M}$.

NOTATION

ℓ, R, length and radius of membrane; r, characteristic pore size; r_0, radius of straight channel; N, r_s, number of channels of model set of capillaries per unit area of the porous medium and their radius; λ, mean free path length; $\bar{u}_{iz}^{ch}$, $\bar{u}_{iz}^{p}$, $\bar{u}_{iz}^{M}$, projection of mean velocity of motion of molecules of the i-th component in the channel, porous medium, and membrane, respectively, onto the membrane axis; p, T, n, pressure, temperature, and number density of mixture particles; m_i, d_i, mass and diameter of molecules of the i-th component; c_i, concentration of i-th component of mixture; η_{12}, η_i, viscosity of mixture and its i-th component, respectively; D_{12}, σ, mutual diffusion coefficient and diffusional-slip coefficient; k, Boltzmann constant; S^{ch}, S^{p}, S^{M}, cross-sectional area of channel, porous medium, and membrane; Q^{ch}, Q^{p}, Q^{M}, volume flow rate of gas mixture through channel, porous medium, and membrane; Q^{e}, experimental volume flow rate of gas mixture; Kn^{p}, Kn^{ch}, Knudsen number in pores and in channel; δ_{12}^{p}, δ_{12}^{ch}, inverse Knudsen number in pores and in channel; Π, porosity; Δp_m, t_m, maximum magnitude of baric effect and time for its attainment; V, chamber volume.

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HEAT TRANSFER IN VAPOR CONDENSATION ON LAMINAR AND TURBULENT LIQUID JETS, TAKING ACCOUNT OF THE INLET SECTION AND VARIABILITY OF THE FLOW RATE OVER THE JET CROSS SECTION

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The results of numerical modeling of heat transfer in phase transition at jets are outlined.

In [1], condensation at jets was investigated, under the assumption of constant liquid flow rate over the jet cross section. In the present work, this investigation is expanded to the case of variable flow rate.

It is assumed that the liquid jet with initial temperature T_0 and specified (at x = 0) velocity distribution over the cross section of a circular aperture of radius R_0 issues into a space filled with vapor of the same liquid, with saturation temperature T_s; the radical component of the temperature gradient is much larger than the axial component. Correspondingly, the momentum and energy equations for the flow of the liquid jet take the form

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