*Journal of Engineering Physics and Thermophysics, Vol. 78, No. 2, 2005*

# PERMEABILITY OF A MEMBRANE WITH EXTERNAL-DIFFUSIONAL RESISTANCE TAKEN INTO ACCOUNT

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UDC 66.064

*An ordinary, nonlinear, first-order differential equation that describes the permeability of a membrane with the external diffusional resistance taken into account is obtained. Based on this equation, the rate of penetration as a function of the physical properties of a gas mixture and the parameters of the membrane element have been found.*

Recently, membrane gas separation has been widely used in such technological processes as the processing of natural gases, enrichment of air with oxygen, and raising of the concentration of hydrogen in blow-through gases in ammonia synthesis [1]. The advantages of membrane separation are economy, absence of chemical conversions, long term of service of the membrane modulus, and the possibility of switching setups into an autonomous mode of operation [1, 2]. In order to determine the optimal parameters of the straight-through membrane filter, one must have a clear idea of the basic laws governing gas separation.

At the initial stage of development of membrane technologies, attention was mainly concentrated on the study of mass transfer in the membrane. In the problem thus stated, the external diffusional resistance to the process of separation in the delivery and drainage channels was not taken into account. However, creation of asymmetrical membranes that simultaneously are highly selective and highly permeable has changed the situation, because the influence of that resistance has become compatible with the influence of the intramembrane one. Attempts were made to calculate the convective external diffusional resistance of the membrane element and were generalized in [1]. They were based on the analogy between the processes of heat and mass transfer on a permeable porous surface and did not take into account the specifics of precisely membrane gas separation.

Correct description of convective mass transfer of a binary gas mixture in a straight-through membrane element (Fig. 1) is given in [3], the main result of which was derivation of an integral equation for the rate of penetration through the membrane $V(x)$ with account for the external diffusional resistance:

$$V(x) = \frac{\Lambda M u_0}{\varepsilon \delta_{\mathrm{m}}}\left[p_0 - \frac{(\alpha+1)(\alpha+3)}{\mathrm{Re}\,\varepsilon}x + \frac{(\alpha+1)^2(\alpha+3)}{\mathrm{Re}\,\varepsilon}\int_0^x\left(\int_0^x V(z)\,dz\right)dx\right]\times$$

$$\times\left[1 - \frac{1-c_0}{\left(1-(\alpha+1)\int_0^x V(x)\,dx\right)\left(1-\frac{5\alpha+17}{(\alpha+5)(\alpha+7)}\mathrm{Pe}_D\varepsilon V(x)\right)}\right]. \tag{1}$$

Here $\alpha = 0$ corresponds to a plane-frame module and $\alpha = 1$, to hollow-fiber one. From solution of Eq. (1) it is seen that the rate of penetration $V(x)$ is changed over the length of the channel. The aim of this work is the solution of this equation.

We will consider the expression enclosed in the first parentheses on the right-hand side of Eq. (1). In membrane separation of gas mixtures, the rate of penetration $V(x)$ is small, about $10^{-3}$–$10^{-1}$; therefore the third term is negligibly small in comparison with the first two. Indeed, if we carry out estimation from above, having assumed that

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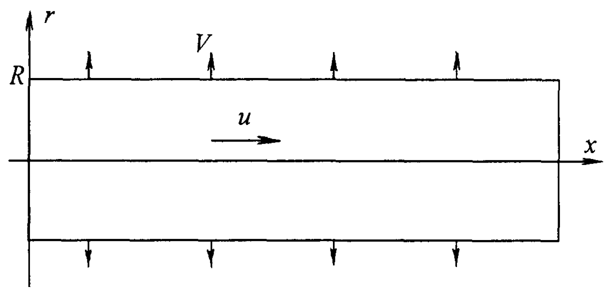

Fig. 1. Schematic of straight-through membrane separation of a gas mixture.

$V(x)$ = const, then the above-mentioned expression will take the form $(\alpha+1)^2(\alpha+3)$ const $x^2/(2\ \mathrm{Re}\ \varepsilon)$. Since $x$ changes from 0 to 1 and const = $10^{-3}$–$10^{-1}$; it is evident that this term can be neglected. Consequently, Eq. (1) can be written in the form

$$V(x)=\frac{\Lambda M u_0}{\varepsilon\delta_{\mathrm{m}}}\left[p_0-\frac{(\alpha+1)(\alpha+3)}{\mathrm{Re}\,\varepsilon}x\right]\left[1-\frac{1-c_0}{\left(1-(\alpha+1)\int\limits_0^x V(x)\,dx\right)\left(1-\frac{5\alpha+17}{(\alpha+5)(\alpha+7)}\mathrm{Pe}_D\varepsilon V(x)\right)}\right]. \tag{2}$$

We put $\frac{\Lambda M u_0}{\varepsilon\delta_{\mathrm{m}}}=A$ and $p_0-\frac{(\alpha+1)(\alpha+3)}{\mathrm{Re}\,\varepsilon}x=p(x)$. We will transform Eq. (2) into a differential one, having introduced the notation $\int\limits_0^x V(x)dx=\varphi(x)$, i.e., $V(x)=\varphi'(x)$. After transition from the variable $V(x)$ to $\varphi(x)$, we obtain

$$\varphi'(x)=Ap(x)\left[1-\frac{1-c_0}{(1-(\alpha+1)\,\varphi(x))\left(1-\frac{5\alpha+17}{(\alpha+5)(\alpha+7)}\mathrm{Pe}_D\varepsilon\varphi'(x)\right)}\right]. \tag{3}$$

We transform relation (3) as

$$(\varphi'(x))^2\frac{(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}{(\alpha+5)(\alpha+7)}-\varphi'(x)\left(1+\frac{Ap(x)\,\mathrm{Pe}_D\varepsilon\,(5\alpha+17)}{(\alpha+5)(\alpha+7)}\right)+Ap(x)\left(1-\frac{1-c_0}{1-(\alpha+1)\,\varphi(x)}\right)=0\,,$$

whence

$$\varphi'(x)=\frac{(\alpha+5)(\alpha+7)}{2(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}+\frac{Ap(x)}{2}-$$

$$-\sqrt{\left(\frac{(\alpha+5)(\alpha+7)}{2(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}+\frac{Ap(x)}{2}\right)^2+\frac{Ap(x)(\alpha+5)(\alpha+7)}{(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}\left(\frac{1-c_0}{1-(\alpha+1)\,\varphi(x)}\right)}\,. \tag{4}$$

In the solution of the quadratic equation, the minus sign was selected to satisfy the boundary condition at $x=0$. We introduce the notation

$$a(x)=\left(\frac{(\alpha+5)(\alpha+7)}{2(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}-\frac{Ap(x)}{2}\right)^2,\quad b(x)=\frac{(\alpha+5)(\alpha+7)}{2(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}+\frac{Ap(x)}{2}\,,$$

$$e(x)=\frac{Ap(x)(1-c_0)(\alpha+5)(\alpha+7)}{(5\alpha+17)\,\mathrm{Pe}_D\varepsilon}\,,$$

TABLE 1. Values of the Setup Parameters Used in the Calculations

| Polymer | $\alpha$ | $\Lambda \cdot 10^{15}$, mole/(m·sec·Pa) | $\delta_m$, μm | $A \cdot 10^8$ | $\mathrm{Pe}_D \varepsilon \cdot 10^3$ |
|---|---|---|---|---|---|
| Cellulose acetate "Gasep" | 0 | 11 | 0.1 | 66 | 46 |
| Polysiloxancarbonate "Karbosil" | 0 | 115 | 0.2 | 345 | 46 |
| Permeable membrane "Dupon" | 1 | 78.6 | 20 | 23.6 | 0.46 |
| Fluoroplastic-42 | 1 | 0.62 | 9 | 0.41 | 0.46 |

Note. The values to calculate the parameters of the setup for membrane gas separation were borrowed from [1]. In all calculation variants, $p_0 = 55.5 \cdot 10^3$ and $u_0 = 3$ m/sec. For the plane-frame module the value $\nu = 1.4 \cdot 10^{-5}$ m$^2$/sec was used (it corresponds to a pressure of 0.1 MPa); for the hollow-fiber module $\nu = 1.4 \cdot 10^{-6}$ m$^2$/sec (it corresponds to a pressure of 1 MPa).

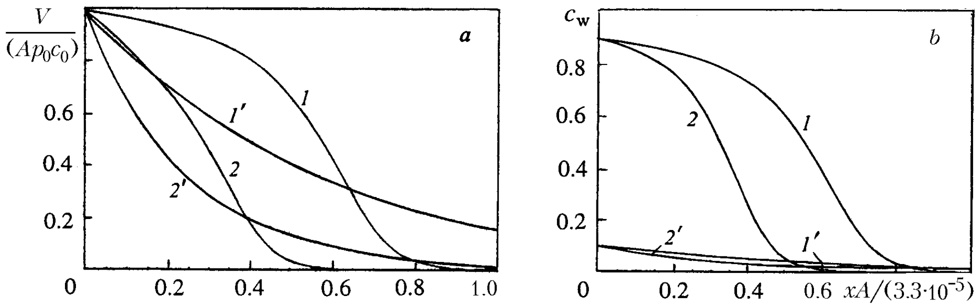

Fig. 2. Permeability (a) and concentration on the wall (b) vs. the distance from the inlet onto the channel: 1, 1′) plane-frame module; 2, 2′) hollow-fiber module [1′, 2′) results for a depleted mixture ($c_0$ = 0.1), 1, 2) for a rich mixture ($c_0$ = 0.9)].

that allows formula (4) to be represented as

$$\varphi'(x) = b(x) - \sqrt{a(x) + \frac{e(x)}{1 - (\alpha + 1)\,\varphi(x)}}\,. \tag{5}$$

The ordinary differential equation (5) is easily solved by the Runge–Kutta method and makes it possible to find the rate of penetration through the membrane with allowance for the external diffusional resistance at the given parameters of the module. The results of calculation carried out for a number of values of the system parameters (see Table 1) are presented in Fig. 2.

We considered two cases: a mixture rich in a separable gas ($c_0$ = 0.9) and a depleted mixture ($c_0$ = 0.1). The concentration of the separated gas $c_w$ on the membrane surface was determined, depending on the longitudinal coordinate, from [3] as

$$c_w(x) = 1 - \frac{1 - c_0}{\left(1 - (\alpha + 1)\int\limits_0^x V(x)\,dx\right)\left(1 - \frac{5\alpha + 17}{(\alpha + 5)(\alpha + 7)}\,\mathrm{Pe}_D \varepsilon V(x)\right)}\,.$$

Using the results of calculations, we selected such self-similar variables in which the form of the graphs depicting the dependence of the rate of penetration $V(x)$ on $A$ did not change. The graphs presented (Fig. 2) allow one to determine the optimal length of the gas-separating membrane element that ensures the highest performance of the module at the given parameters of the system.

## NOTATION

$c$, concentration of a penetrating component; $D$, coefficient of diffusion, $m^2$/sec; $L$, length of the channel, m; $M$, molar mass of the penetrating component, kg/mole; $\overline{p}$, pressure, Pa; $p = \overline{p}(\rho u_0^2)$, dimensionless pressure; $Pe_D = u_0R/D$, diffusion Peclet number; $R$, radius (half-width) of the channel, m; $\overline{r}$, radial coordinate, m; $r = \overline{r}/R$, dimensionless radial coordinate; $Re = u_0R/\nu$, Reynolds number; $\overline{u}$, longitudinal projection of the rate, m/sec; $u = \overline{u}/u_0$, dimensionless longitudinal projection of the rate; $u_0$, mean flow-rate velocity at the inlet to the channel, m/sec; $\overline{V}$, transmembrane velocity, m/sec; $V = \overline{V}L/(u_0R)$, dimensionless transmembrane velocity; $\overline{x}$, longitudinal coordinate, m; $x = \overline{x}/L$, dimensionless longitudinal coordinate; $\delta_m$, effective thickness of the membrane, m; $\varepsilon = R/L$, ratio between two characteristic dimensions of the channel; $\Lambda$, permeability of the membrane, mole·m/(N·sec); $\nu$, kinematic viscosity, $m^2$/sec; $\rho$, density, kg/$m^3$. Subscripts: 0, value at the inlet to the channel; m, membrane; w, channel wall. Superscripts: $'$, derivative with respect to dimensionless longitudinal coordinate.

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