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COLLISION OF POLYMER PARTICLE WITH RIGID BARRIER

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The article establishes a correlation of the relaxation spectrum with the dissipation of kinetic energy of a polymer particle upon impact.

Impact of polymer particles against a rigid barrier is encountered in many technological processes: in the application of polymer coatings by spraying, in the production of composite materials, in dispersion, etc. For comparatively low velocities, when the dynamic head is smaller than the modulus of elasticity: $\rho U_0^2 \leq G(t_1)$, the principal influence on the characteristics of the impact is exerted by the rheological properties of the polymer. For small bodies: $l \ll ct_i$, $c = \sqrt{G/\rho}$ is the velocity scale of the shear wave, the problem of impact may be dealt with in quasistatic approximation, which is widely used in the theory of elastic bodies [1]. For calculating the deformation of a particle upon impact the results of the solution of contact problems are used. In view of the comparatively small deformations, the rheological properties of the polymer are described by the linear theory of viscoelasticity. The solution of the contact problem for viscoelastic material is constructed with the aid of the principle of correspondence proceeding from the relations for elastic material with analogous geometry [2, 3]. In [4] the impact of a polymer particle was examined for the case of forces of adhesion originating on the contact spot. This made it possible to estimate the force of adhesion required for maintaining the particle on the barrier. The present article examines impact without adhesion. In that case the particle rebounds from the barrier. The decrease of its kinetic energy is determined solely by viscous losses in the polymer. To evaluate the

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Fig. 1. Radius of the region of contact; $a(t) = a(t_1 (t))$ for $t > t_m$. Fig. 2. Restitution coefficient K(β). K, %.

Fig. 3. Dependence of the specific dissipated energy E_d on the speed of impact U_o for PMMA (1) and PS 9.85 (2), 8.09 (3) mm: I, II) empirical formulas [10]; III) $E_d = 0.0831U_o^{2^{\circ}2}$. E_d , J/kg; U_o , m/sec.

relaxational properties of a polymer, it is common practice at impact to measure the restitution coefficient K which is equal to the ratio of the kinetic energies of the particle after and before the collision [5]. However, the formulas used for that are based primarily on relations which were derived under such conditions that stress relaxation in the polymer was regarded as a small correction only (see [5]).

A direct impact of a viscoelastic particle proceeds in two stages. At the first stage $(t \leq t_m)$ the particle becomes deformed, its speed in the direction to the barrier is reduced, the region of contact becomes wider, part of the kinetic energy changes into elastic strain energy. At the second stage $(t > t_m)$ there occurs "unloading" in reverse deformation, the region of contact becomes smaller, part of the elastic energy changes into kinetic energy, and the particle moves away from the barrier. The mathematical formulation of the problem of impact consists of the following equations (see [4]):

first stage (U in the direction toward the barrier)

$$M - \frac{dU}{dt} = -F(t), \quad \frac{dh}{dt} = U; \quad h|_{t=0} = 0, \quad U|_{t=0} = U_0, \tag{1}$$

second stage (U in the direction from the barrier)

$$M - \frac{dU}{dt} = F(t), \quad \frac{dh}{dt} = -U; \quad h|_{t=t_m} = h_m, \quad U|_{t=t_m} = 0.$$
(2)

The values of h_m , t_m are determined from the solution of system (1). The correlation between the force F, the displacement h, and the radius of the contact spot a is given by the formulas [2, 3]:

$$F = \frac{8}{3R(1-\nu)} \int_{0}^{t_{1}(t)} G(t-t') da^{3}(t'), \qquad (3)$$

first stage

$$h = \frac{a^2(t)}{R},\tag{4}$$

second stage

$$h = \frac{a^{2}(t)}{R} - \frac{1}{R} \int_{t_{m}}^{t} J(t-t') d\left[\int_{t_{1}(t')}^{t} G(t'-t'') da^{2}(t'')\right].$$
(5)

The Poisson ratio ν is regarded as constant. The functions of relaxation and creep are inverse to each other:

$$\int_{0}^{t} G(t-t') dJ(t') = 1.$$
 (6)

The function G(t) is correlated with the spectrum $H(\lambda)$ by the dependence [5, 6]:

$$G(t) = \int_{0}^{\infty} \frac{H(\lambda)}{\lambda} \exp\left(1 - \frac{t}{\lambda}\right) d\lambda.$$

At the first stage of the collision $t_1(t) = t$, and at the second stage the time $t_1(t)$ corresponds to the instant preceding t_m when the radius of the region of contact $\alpha(t)$ is equal to its preceding value $\alpha(t_1(t))$ (Fig. 1) [2, 3]. For viscoelastic material, in distinction to elastic material, the dependence $\alpha(t)$ is asymmetric relative to the value of t_m . The difference is that the correlation between the displacement of the center of gravity and the radius of the region of contact is determined at the first stage only by the simple geometric relation (4). It should be noted that in view of the smallness of deformations of bodies in collision at low speed ($\rho U_0^2 \leq G$) the system of Eqs. (1)-(5) also describes the collision of a rigid particle with a polymer barrier (see [3]).

In [7] (see also [4]) a method was worked out for the numerical solution of the problem (1)-(5). The results of computer calculations were presented in [4]. We will carry out an analytical investigation of this problem using a method suggested in [8]. The effect of the section of relaxation spectrum with times longer than t_i will be regarded as elastic, with shorter times as viscous. Then

$$G(t) = \int_{0}^{t} \frac{H(\lambda)}{\lambda} \exp\left(-\frac{t}{\lambda}\right) d\lambda + \int_{t_{i}}^{\infty} \frac{H(\lambda)}{\lambda} \exp\left(-\frac{t}{\lambda}\right) d\lambda \approx \eta_{i} \delta(t) + G_{i}, \quad \delta(t) = \lim_{\lambda \to 0} \frac{1}{\lambda} \exp\left(-\frac{t}{\lambda}\right).$$
(7)

Here, $G_y = \int_{t_i}^{\infty} H(\lambda) d \ln \lambda$, $\eta_i = \int_{0}^{1} H(\lambda) d\lambda$ are the effective values of elasticity and viscosity of a liquid at impact. For (7) the function of creep is written in the form [see (6)]:

$$J = \frac{1}{G} \left[1 - \exp\left(-\frac{t}{\lambda_{i}}\right)^{2} \right], \ \lambda = \frac{\eta_{i}}{G_{i}}.$$

These expressions simplify the relations (3), (5):

$$F = \frac{8}{3R(1-v)} \left(G_{i}a^{3} + 3\eta_{i}a^{2}\frac{da}{dt} \right), \quad t \leq t_{m};$$

$$F = \frac{8}{3R(1-v)} G_{i}a^{3}, \quad t > t_{m};$$

$$h = -\frac{1}{R} \left[a^{2}(t) - \lambda_{i} \int_{t_{m}}^{t} \left[1 - \exp\left(-\frac{t-t'}{\lambda}\right) \right] \frac{d^{2}a^{2}}{dt'^{2}} dt' \right], \quad t > t_{m}.$$

The results of numerical calculations of the collision of a viscoelastic particle presented in [4] confirm the applicability of assumption (7). In that case the system of integrodifferential Eqs. (1)-(5) is equivalent to the system of differential equations which in the dimensionless values $t = t_0\tau$, $U = U_0u$, $h = h_0y$, $a = a_0x$, F = Ff, $G(t) = G_0g(\tau)$ is written in the form:

$$\frac{du}{d\tau} = -g_1 y^{3/2} - \frac{3}{2} g_2 y^{1/2} u, \quad \frac{dy}{d\tau} = u,
y = x^2, \quad u|_{\tau=0} = 1, \quad y|_{\tau=0} = 0;
\frac{du}{d\tau} = g_1 y_e^{3/2}, \quad \frac{dy_e}{d\tau} = -u - g_2 y_e^{3/2},
y = y_e + \frac{g_2}{g_1} u, \quad y_e = x^2,
u|_{\tau=\tau_m} = 0, \quad y_e|_{\tau=\tau_m} = y_m.$$
(8)

(9)

$$g_{1} = \frac{G_{1}}{G_{0}}, \quad g_{2} = \frac{\eta_{1}}{G_{0}t_{0}}, \quad t_{0} = \frac{\omega R}{U_{0}}, \quad h_{0} = \omega R, \quad a_{0} = \omega^{1/2} R,$$

$$F_{0} = \frac{8G_{0}R^{2}\omega^{3/2}}{3(1-\nu)}, \quad \omega = \left[\frac{\pi\kappa(1-\nu)\rho U_{0}^{2}}{2G_{0}}\right]^{2/5},$$



Fig. 4. Dependences: a) elastic medium G = $1.4 \cdot 10^6$ Pa (1); spheres with diameter (cm) 0.87 (2); 2.54 (3); 5.08 (4) [11]; calculation for viscoelastic medium (5); b) spheres with diameter 0.64 (1); 0.87 (2); 1.27 (3); 2.54 (4); 3.81 (5); 5.08 (6) [11]; calculation for viscoelastic medium (7, 8, 9). t_i, msec; $(M^2/RU_0)^{1/5}$, $(kg^2 \cdot sec/m^2)^{1/5}$.

the parameter ω is small. The particle becomes detached from the barrier when at the second stage $y_e = 0$. The dependences of the dimensionless time of impact τ_i and of the restitution coefficient K on the parameters g_1 , g_2 are determined from the expressions:

$$\tau_i(g_1, g_2) = \tau|_{y_e=0}, K(g_1, g_2) = u^2|_{y_e=0}.$$

Transformation of the system (8), (9):

$$u, \tau, y, y_e \rightarrow u', g_1^{-2/5}\tau', g_1^{-2/5}y', g_1^{-2/5}y'_e$$

is equivalent to the transition in it to new coefficients:

$$g_1, g_2 \rightarrow 1, g_2/g_1^{3/5}$$

In particular, these relations show that the duration of impact and the restitution coefficient

$$t_{i} = \left[\frac{\pi \varkappa (1-\nu)\rho U_{0}}{2G_{i}}\right]^{2/5} \frac{R}{U_{0}} \tau_{i} \left(1, \frac{g_{2}}{g_{1}^{3/5}}\right), \quad K = K \left(1, \frac{g_{2}}{g_{1}^{3/5}}\right)$$

are determined by only one parameter $g_2/g_1^{3/5}$. Thus the restitution coefficient is correlated with the duration of the collision by the universal relation K = K(β), $\beta = n_i/G_it_i$, which does not depend on the mass of the particle, its speed, or the radius of curvature at the point of contact. In parametric form it is described by the expression:

$$K = u^{2} \left(1, \frac{g_{2}}{g_{1}^{3/5}} \right) \Big|_{y_{e}=0}, \beta = \frac{g_{2}}{g_{1}^{3/5} \tau_{1} \left(1, \frac{g_{2}}{g_{1}^{3/5}} \right)}$$

This result is the consequence of the approximation (7) in which the relaxation spectrum is characterized all in all by the two integral parameters $n_i(t_i)$ and $G_i(t_i)$.

Having determined the dependence $\beta = \beta(t_i)$ from measurements of the restitution coefficient and of the duration of the impact, we can calculate the relaxation characteristics of the polymer. Since

$$-t_{\mathbf{i}} \frac{dG_{\mathbf{i}}}{dt_{\mathbf{i}}} = H(t_{\mathbf{i}}) = \frac{d\eta_{\mathbf{i}}}{dt_{\mathbf{i}}},$$

we obtain after differentiating the relation $\eta_i = G_i t_i \beta$:

$$\frac{d\ln G_{\mathbf{i}}}{dt_{\mathbf{i}}} = -\left(1 + \frac{d\ln\beta}{d\ln t_{\mathbf{i}}}\right) \frac{\beta}{(1+\beta)t_{\mathbf{i}}}$$

If we approximate the function of β by the power dependence $\beta = (T/t_1)^q$, we have

$$G_{\mathbf{i}} = C[1 + (T/t_{\mathbf{i}})^{q}]^{\frac{1-q}{q}}, \quad \eta_{\mathbf{i}} = CT[1 + (t_{\mathbf{i}}/T)^{q}]^{\frac{1-q}{q}},$$

$$H(t_{\mathbf{i}}) = \frac{(1-q)G_{\mathbf{i}}}{1 + (t_{\mathbf{i}}/T)^{q}}.$$
(10)

For calculating the integration constant C in these expressions additional experimental data are required.

Let us find the behavior of the magnitudes τ_i and K when the proportion of the dissipated energy is small ($g_2 << g_1$) and when it is large ($g_2 >> g_1$). When $g_2 << g_1$, the difference between the motions of the viscoelastic and the elastic particles is small. Then $\tau_i z \tau_i(1,0) =$ $(4/5)^{3/5}B(2/5, 1/2)$ [1], and for calculating K it suffices to calculate the work of the viscous forces along the path of motion of the elastic particle. This yields:

$$K \approx 1 - 3 \left(\frac{4}{5}\right)^{2/5} B\left(\frac{3}{2}, \frac{3}{5}\right) \frac{g_2}{g_1^{3/5}} \approx 1 - 3,460 \frac{g_2}{g_1^{3/5}}.$$

We obtain the same result if we seek the solution of Eqs. (8), (9) in the form of a series with powers of g_2 and confine ourselves to the first term. Thus, with small values of β :

$$K = 1 - \beta \frac{120\pi}{11} \operatorname{tg} \frac{\pi}{10} = 1 - 11,136 \ \beta.$$
(11)

To construct the asymptotic formulas for $g_2 >> g_1$, we go over in Eqs. (8), (9) to the values $\tilde{u} = u$, $\tilde{y} = g_2^{2/3}y$, $\tilde{y}_e = g_2^{2/3}y_e$, $\tilde{\tau} = g_2^{2/3}\tau$. The equations in respect to these variables are obtained by replacing the coefficients g_1 , $g_2 \rightarrow \mu = g_1/g_2^{5/3}$, 1. To construct the asymptotic solution of the problem for $\mu \ll 1$ we use the method of singular disturbances [9]. At the first stage of the collision the external and the internal expansion are constructed in the form of series with powers of μ . In the boundary layer the internal variable is $\mu \tilde{\tau}$. The compound asymptotic expansion for large $\tilde{\tau}$, found by a method that was described in detail in [9], is written in the form (here we use the solution of the problem for a viscous liquid with $g_1 = 0$, $g_2 = 1$ [4]):

$$\tilde{y} = \exp\left(-\frac{2}{3}\mu\tilde{\tau}\right) - 2\sqrt[4]{3}\exp\left(-\frac{3}{2}\tilde{\tau} - \frac{\pi\sqrt{3}}{6}\right) + 2\mu\left[\exp\left(-\frac{2}{3}\mu\tilde{\tau}\right) - \frac{4}{9}\exp\left(-\frac{1}{3}\mu\tilde{\tau}\right)\right] + O(\mu^2),$$
$$\tilde{u} = 3\sqrt[4]{3}\exp\left(-\frac{3}{2}\tilde{\tau} - \frac{\pi\sqrt{3}}{6}\right) - \frac{2}{3}\mu\exp\left(-\frac{2}{3}\mu\tilde{\tau}\right) + O(\mu^2).$$

From this we find the instant when the particle stops and the largest displacement of its center of gravity:

$$\widetilde{\tau}_m \approx \frac{6}{9-4\mu} \left(\ln \frac{9\sqrt{3}}{2\mu} - \frac{\pi\sqrt{3}}{6} \right),$$

$$\widetilde{y}_m \approx \left(1 + \frac{14}{9} \mu \right) \exp\left(-\frac{2}{3} \mu \widetilde{\tau}_m \right) - \frac{8}{9} \mu \exp\left(-\frac{1}{3} \mu \widetilde{\tau}_m \right).$$

For approximate calculations we may put $\tilde{y}_m \approx 1 - 2\mu \quad (\tilde{\tau}_m - 1)/3$. At the second stage of the collision the external expansion is constructed in the form of a series with powers of μ , and the internal expansion with powers of $\mu^{1/3}$. The variable of the boundary layer is $\mu^{1/3}(\tilde{\tau} - \tilde{\tau}_m)$. Directly from the equations for the second stage (9) we find:

$$\tilde{u} = \mu (\tilde{y}_m - \tilde{y}_e) + O(\mu^{5/3}), \quad \tilde{y} = \tilde{y}_m + O(\mu^{2/3}).$$

For large values of $(\tilde{\tau} - \tilde{\tau}_m)$ the compound asymptotic expansion for \tilde{y}_e is written in the form:

$$\tilde{y}_{e} = \mu^{2/3} \left\{ \frac{4\pi}{3\sqrt{3}} \tilde{y}_{m}^{2/3} - \mu^{1/3} [(\tilde{\tau} - \tilde{\tau}_{m}) \tilde{y}_{m} + 2\tilde{y}_{m}^{1/2}] + O(\mu^{2/3}) \right\}.$$

The use of these relations yields

$$\widetilde{\tau}_{i}(\mu, 1) \approx \widetilde{\tau}_{m} + \frac{1}{\mu^{1/3} \widetilde{y}_{m}^{1/3}} \left(\frac{4\pi}{3\sqrt{3}} - \frac{2\mu^{1/3}}{\widetilde{y}_{m}^{1/6}} \right), \quad K = \mu^{2} \widetilde{y}_{m}^{2}$$
(12)

When the polymer has high viscosity ($\mu << 1$), a large part of the kinetic energy is dissipated at the first stage of the collision. The shape of the particle is not restored at the second stage. The ratio of the durations of the first and the second stage is of the order $\mu^{1/3}\ln(1/\mu)$. Expressions (12) and the formula $\beta = 1/\mu \tilde{\tau}_1(\mu, 1)$ determine the function K(β) in parametric form when β is large.

For the numerical solution of Eqs. (8), (9) we used the Runge-Kutta method of second order. At the first stage the calculations ended with u = 0, at the second stage with $y_e = 0$. As scale of the shear modulus we took $G_0 = G_1 + \eta_1/t_0$, then $g_1 + g_2 = 1$. According to the results of numerical calculations, the dependence $K(\beta)$ is plotted in Fig. 2. A comparison with the asymptotic formulas shows that with an accuracy of the order of 10%, the approximation of small β is applicable with K \gtrsim 0.7 and of large β with K \lesssim 0.02.

In experimental work concerned with collisions of polymer particles, most authors confined themselves to calculating the proportion of lost energy (1 - K) which was treated as the logarithmic decrement of the attenuation of free vibrations with frequency $\frac{1}{2} t_1$ [5]. More informative experiments are reported in [10, 11]. Okuda and Choi [10] studied the collision of spheres of polymethyl metacrylate (PMMA) and of polystyrene (PS) with a metallic target. The experiments were carried out at room temperature at which both polymers are in a vitreous state. The diameter of the spheres of PMMA was 9.8 mm, of PS 9.85, 8.09 mm, the speed was 35 to 200 m/sec, duration of impact from $2 \cdot 10^{-5}$ to 10^{-4} sec. The static shear modulus and density of PMMA were 1.39.10° Pa, 1.2.10° kg/m°; of PS 1.26.10° Pa, 1.46.10° kg/m°, respectively. For these experiments $\rho U_0^2/2G \leq 0.02$. The experiments showed that the maximal values of force and deformation of the spheres with $U_0 \leq 100$ m/sec are in good agreement with the calculations by Hertz' formula (elastic sphere) using the static shear modulus. For most speeds the calculated values are higher than the experimental ones. Figure 3 shows the dependences of the specific dissipated energy (per unit mass) on the speed. The experimental data of [10] are described by the formulas $E_d = 0.0439 U_0^{2 \cdot 35}$ for PMMA and $E_d = 0.146 U_0^{2 \cdot 18}$ for PS. The analytical solution of the system (8), (9) in the approximation of low viscosity yields

$$E_{d} = 1,444\eta_{i} U_{o}^{2,2} / \rho^{0,4} G_{i}^{0,6} (1-\nu)^{0,4} R.$$
(13)

The dependence on the speed (13) is in good agreement with the data for PS, and in somewhat poorer agreement for PMMA. However, it can be seen from Fig. 3 that the data for PMMA are also fully described by the expression $F_d = 0.0831U_0^{2*2}$ which is in agreement with (13). By using the static shear moduli, we can determine n_i . For PMMA $n_i \ge 1.1 \cdot 10^3$ Pa•sec, for PS $n_i \ge 10^3$ Pa•sec. Then $H(t_i) \sim n_i/t_i \sim 10^8$ Pa•sec; this corresponds to the values of the relaxation spectrum [5] for the β -process of PMMA and PS (small-scale sections of the chain) at room temperature.

Southern and Thomas [11] investigated the collision of steel balls with diameters from 0.625 to 5 cm with rubber at room temperature and at speeds from 0.03 to 1.7 m/sec, duration of the impact from 10^{-3} to $2 \cdot 10^{-2}$ sec. The experiments were carried out with specimens of natural rubber (NR) vulcanized with the aid of 1 and 6 (weight) % dicumy1 and with specimens of butyl rubber (BR) vulcanized by a vulcanizing group with 2 (weight) % sulfur. Vulcanization was carried out at 150°C for 1 h. The tensile moduli of elasticity measured in the course of several minutes are 0.9.10° and 2.9.10° Pa for NR, and 1.4.10° Pa for BR. These experiments reveal the properties of polymers in the transition zone from vitreous to highly elastic where the relaxation processes are determined by the motion of sections of the macromolecule [5, 6]. For experiments [11] $\rho U_0^2/2G \leq 0.03$. The times of collision for NR are in good agreement with the calculations by Hertz' formula using the "minute" moduli of elasticity. For BR the calculated values are higher than the experimental ones (see Fig. 4a with the data of [11]). With increasing duration of the collision the difference becomes smaller. The dependences of the restitution coefficient on the duration of the collision for NR (with denser space network) and BR with different speeds and sizes of the spheres (Fig. 4b) confirm that a universal relation $K(\beta)$ exists. The data for NR are described by the linear Eq. (11). This makes it possible to estimate $n_i/G_i \sim (4.3-4.9) \cdot 10^{-5}$ sec, then $n_i \sim 50$ Pa·sec, $H(t_i) \sim 2.5 \cdot 10^4$ Pa, which in order of magnitude is in agreement with the values for NR [5]. For BR the dependence K(ti) is described by a power law. A comparison of the values of the restitution coefficient with the results of numerical calculations of the dependence $K(\beta)$ shows that for BR $\beta = (T/t_1)^q$ for T = $1.517 \cdot 10^{-3}$ sec, q = 0.362 (Fig. 4b). Using the formula for the collision of a viscoelectric medium, we obtain the relation:

$$\frac{g_1^{3/5}\beta}{g_2} \left[\frac{8G_i}{3(1-\nu)} \right]^{2/5} t_i = \left(\frac{M^2}{RU_0} \right)^{1/5}.$$
(14)

The function $\beta(t_1)$ determines the dependence $G(t_1)$ with an accuracy up to the integration constant in (10). With its value equal to 0.87 G_e (G_e is the "minute" modulus), the calculated values of the duration of the collision correspond to the experimental data for BR (Fig. 4a). In the range from 2·10⁻³ to 2·10⁻² sec the ratio $G(t_1)/G_e$ drops from 2.7 to 1.6, viscosity increases from 2.3·10³ to 5.7·10³ Pa·sec, the negative slope of the relaxation spectrum $H(\lambda)$ in logarithmic coordinates is 0.47. The obtained data for BR are in agreement with the behavior of elastomers in the transition region [5, 6].

NOTATION

K, restitution coefficient; U_o, speed of impact; G(t), relaxation function; J(t), creep function; v, Poisson ratio; ρ , density of the material; t_i, duration of the collision; M, mass of the particle; t_m, instant of maximal deformation of the particle; h, displacement of the center of mass of the particle; F, force acting from the side of the barrier; α , radius of the contact spot; R, radius of curvature of the particle at the point of contact; H(λ), relaxation spectrum; G_o, scale of the shear modulus; $\delta(t)$, Dirac's delta function; B(x, y), beta function; E_d, specific dispersed energy.

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EFFECT OF LONGITUDINAL DIFFUSION ON SEPARATION

OF GAS MIXTURES USING SEMIPERMEABLE MEMBRANES

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Flow of a binary gas mixture in a cylindrical channel is studied considering presence of longitudinal diffusion fluxes of the components, as occurs in separation of gas mixtures using semipermeable membranes.

This study will consider transport of binary gas mixtures in a cylindrical channel with selectively permeable walls with consideration of diffusion motion of the mixture in the longitudinal direction, caused by a change in component concentration along the channel produced by their differing abilities to penetrate the wall. The results are applicable primarily to separation of gas mixtures and extraction of a target product of specified composition using membranes of metal, glass, polymers, etc. [1-3].

As a rule, in studies of mixtures in channels with semipermeable walls the literature assumes idealized flow models, the so-called ideal displacement model, corresponding to the case in which the longitudinal diffusion flux of a component in the channel is much less than the convective flow, or the total mixing model, based on the assumption that because of intense mixing in the channel, component concentrations constant over length are established [3, 4].

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