

NOTATION

K, restitution coefficient; U_0 , speed of impact; $G(t)$, relaxation function; $J(t)$, creep function; ν , 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; a , radius of the contact spot; R , radius of curvature of the particle at the point of contact; $H(\lambda)$, relaxation spectrum; G_0 , 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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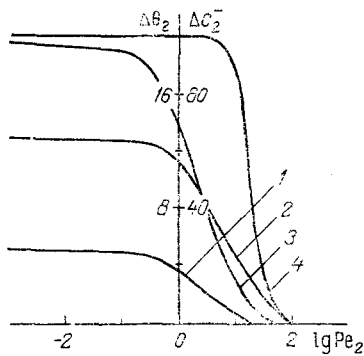


Fig. 1

Fig. 1. Relative deviations of $\Delta\theta_2$ (curves 1, 3) and Δc_2^- (curves 2, 4) versus number Pe_2 : 1) $\theta \sim 0.15$; 2, 3) 0.55; 4) 0.80. $\Delta\theta_2, \Delta c_2^-, \%$.

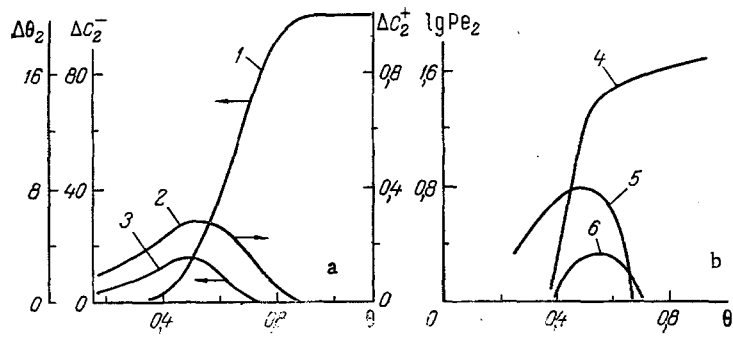


Fig. 2

Fig. 2. Relative deviations for fixed value of Pe_2 (a) and $\log Pe_2$ for fixed relative deviations (b) vs coefficient θ : 1) Δc_2^- ; 2) Δc_2^+ ; 3) $\Delta\theta_2$; 4) $\Delta c_2^- = 10\%$; 5) $\Delta\theta_2 = 5\%$; 6) $\Delta c_2^+ = 1\%$.

Comparative analysis of the total mixing and ideal displacement regimes shows that the process of longitudinal diffusion in the channel leads to reduced separation efficiency [4]. To determine the mixture flow conditions characteristic of ideal displacement, it is necessary to determine the effect of longitudinal diffusion on separation characteristics for various channel and mixture parameters.

The steady-state problem of convective diffusion in a channel with permeable walls was considered in [3, 5, 6]. Depending on the formulation of the problem, both numeric and analytical techniques for its solution have been developed. However, the results of those studies leave unanswered a number of questions, in particular, those of the effect of longitudinal diffusion on separation for given parameters, and determination of parameter values at which change in the separation characteristics due to longitudinal diffusion does not exceed a specified value.

Solution of these problems using the example of mixture flow in an individual channel permits determination of the region of significant effect of longitudinal diffusion on separation characteristics, and development of recommendations for design of equipment consisting of groups of hollow selectively permeable filaments in the interior of which the mixture to be separated is supplied (in the text below we will term such devices membrane elements).

We will consider the isothermal flow of a binary gas mixture in a channel. Assuming that the permeability of each of the mixture components through the walls is independent of their concentrations, the equation describing this process for the more permeable component has the form

$$-d \left(xL - \frac{S_{\perp}}{RT} D_{12} \frac{d(xP_1)}{dl} \right) = \frac{Q_1 2\pi P_1}{\ln(r_1/r_0)} dl(x - \gamma x') \quad (1)$$

in the channel length dL , in accordance with Fick's law. In the future we will assume that pressure losses in the channel produced by the viscous character of the flow, removal of a portion of the mixture through the walls, and change in the molar mass of the mixture are insignificant, and that the pressure at any arbitrary point in the channel is the same, equal to the mixture pressure in the original flow. In this case Eq. (1) can be reduced to the form presented in [3].

It is obvious that penetration of the less permeable component through the wall in a channel length dL is described by an equation analogous to Eq. (1), with replacement of the concentrations x and x' by $1 - x$ and $1 - x'$, respectively. Combining the component transport equations, we obtain for the gas mixture as a whole:

$$-\frac{dL}{dl} = \frac{2\pi P_1}{\ln(r_1/r_0)} [Q_1(x - \gamma x') + Q_2(1 - x - \gamma(1 - x'))] \quad (2)$$

Introducing the dimensionless variables $q = L/G$, $z = l/L_M$, after transformations the system of Eqs. (1), (2) reduces to the form

$$\begin{cases} \frac{dq}{dz} = -\frac{1}{B} [x - \gamma x' + \alpha^* (1 - x - \gamma(1 - x'))], \\ \frac{d^2x}{dz^2} = \frac{Pe_1}{B} (x - \gamma x') + Pe_1 \frac{d(xq)}{dz}, \end{cases} \quad (3)$$

where

$$Pe = GL_M / (S_{\perp} P_1 D_{12} / RT); \quad B = G \ln(r_1 / r_0) / (Q_1 2\pi P_1); \quad \alpha^* = Q_2 / Q_1. \quad (4)$$

Solution of the system of differential equations (3) allows determination of the unknown quantities $\theta = 1 - q|_{z=1}$, $c^- = x|_{z=1}$, $c^+ = (1/\theta)(c - (1 - \theta)c^-)$ for specified Pe_1 , B , α^* , and γ .

Since the process of numerical integration of Eq. (3) has the stability required only in the direction from the point of exit of the flow which has not penetrated the wall to the point of entrance of the original flow, boundary conditions in the form

$$q|_{z=1} = 1 - \theta, \quad x|_{z=1} = c^-, \quad \left. \frac{dx}{dz} \right|_{z=1} = 0 \quad (5)$$

must be used. Then the unknown quantities θ and c^- must satisfy boundary conditions

$$\begin{aligned} 1 - q|_{z=0} &= 0, \\ c - x|_{z=0} + \frac{1}{Pe_1} \left. \frac{dx}{dz} \right|_{z=0} &= 0. \end{aligned} \quad (6)$$

The system of nonlinear equations (6) was solved for θ and c^- by Newton's method [7], while Eq. (3) was integrated by the fourth-order Runge-Kutta method [8].

The right side of Eq. (3) contains the concentration of the easily penetrating component in the low-pressure cavity (LPC) beyond the channel wall x' . The explicit form of the dependence of x' on x is determined by the organization of the gas mixture flow in the LPC (we will assume that here a perpendicular efflux regime is realized). In this case the concentration x' is defined from the expression [4]:

$$x' = \frac{x - \gamma x'}{x - \gamma x' + \alpha^* (1 - x - \gamma(1 - x'))}. \quad (7)$$

We will analyze the effect of longitudinal diffusion on separation characteristics of the membrane element as a function of the problem parameters using the example of separation of an H_2 - N_2 mixture using polypropylene filaments. The H_2 (more permeable component) concentration in the original flow is $c = 0.6$, the pressure ratio $\gamma = 0.02$, and the ratio of the permeabilities of H_2 and N_2 through the polypropylene membrane $\alpha^* = 0.031$ [9]. The value of γ selected was chosen so that the effect of the gas flow regime in the LPC on the separation process would be minimal.

The degree of influence of longitudinal diffusion on separation characteristics of the element is determined by the Peclet number Pe . As the mixture moves along the separation channel a portion of the original flow penetrates into the LPC, so that local values of $Pe = LZ_M / (S_{\perp} P_1 D_{12} / RT)$ decrease, with approach toward the point of exit of the mixture which has not penetrated, to a value $Pe_2 = (1 - \theta)Pe_1$.

From the practical viewpoint, in studying the effect of longitudinal diffusion on separation characteristics of the element it is convenient to be able to use either of the numbers Pe_1 or Pe_2 . For subsequent analysis the number Pe_2 , the value of which is minimal for a given flow regime was chosen.

With increase in Pe_2 the separation characteristics of the element become close to those of an element with ideal displacement, while with decrease in Pe_2 its separation characteristics approach those obtained with the complete mixing model.

As quantities describing the effect of longitudinal diffusion we will use the relative deviations of the element characteristics from values obtained using the complete mixing model (subscript 1) or the ideal displacement model (subscript 2):

$$\Delta a_{1,2} = \left| \frac{a(Pe_2) - a_{1,2}}{a(Pe_2)} \right| \cdot 100\%, \quad a = \{\theta, c^-, c^+\}.$$

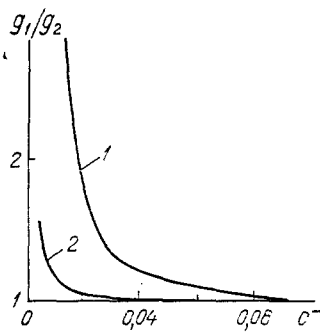


Fig. 3

Fig. 3. Nonpenetrating flow productivity vs oxygen concentration c_2^- .

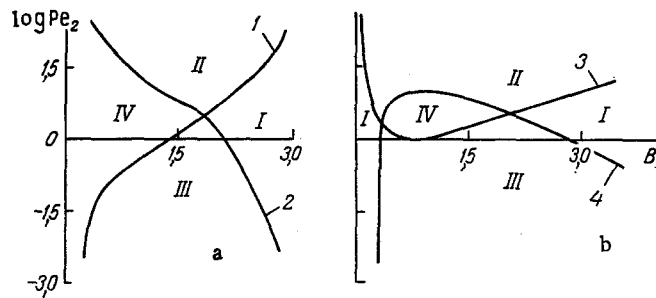


Fig. 4

Fig. 4. Lines of constant $\Delta c_{1,2}^- = 3\%$ (a) and $\Delta \theta_{1,2} = 5\%$ (b): 1) Δc_1^- ; 2) Δc_2^- ; 3) $\Delta \theta_1$; 4) $\Delta \theta_2$.

From the curves shown in Fig. 1 it is evident that with reduction in Pe_2 to the value 0.1 the changes $\Delta \theta_2$ and Δc_2^- reach values corresponding to those of element separation characteristics calculated with the ideal displacement and complete mixing models, and do not change for further decrease in Pe_2 . Thus, it can be assumed that for numbers $Pe_2 \leq 0.1$ the total mixing regime is realized in the element.

As is evident from Eq. (1) the diffusion flow of a component in the high pressure volume depends not only on the parameters appearing in Pe_2 , but also on the concentration gradient. For a fixed membrane selectivity α^* and original flow composition the component concentration gradient in the high pressure volume is defined by the value of θ , with increase of which the concentration of the more permeable component in the nonpenetrating flow decreases, while its mean concentration gradient in the high pressure volume increases [10]. This leads to an increase in the diffusion flow of the component. In turn, at fixed values of G and element geometric parameters, increase in θ leads to a reduction of the convective flow of the component in any section of the element. The combined effect of these factors leads to intensification of the relative contribution of the diffusion mechanism to the net transport of the component, longitudinal mixing in the element, and the degree of influence of longitudinal diffusion on the separation process, as is confirmed by the results shown in Fig. 1.

The results shown in Fig. 2 illustrate the effect of longitudinal diffusion on various separation characteristics of the element. With increase in θ the concentration of the more permeable element in the nonpenetrating flow tends to zero [10], the diffusion flow in the longitudinal direction increases, which leads to a corresponding growth in c^- and Δc_2^- . Longitudinal diffusion then affects not only the concentration of the easily penetrating component in the residual flow, but also its value at every section of the element. In particular, the concentration of the easily penetrating component in the input section of the element proves to be less than in the original flow.

The effect of longitudinal diffusion on c^+ and θ is insignificant, especially in the region of low and near-unity values of the flow separation coefficient. For small values of θ the concentrations of the easily penetrating component in the nonpenetrating and original flows differ little, so that the relative contribution of the diffusion mechanism to total component transport is insignificant and the values of Δc_2^+ and $\Delta \theta_2$ are small. To determine the degree of influence of longitudinal diffusion on c^+ and θ in the high θ range we will make use of the component material balance relationship in the element:

$$\theta c^+ + (1 - \theta) c^- = c.$$

With increase in θ the quantities c^- and $(1 - \theta)c^-$ decrease, whence it follows that for a fixed value of component concentration in the original flow $\theta c^+ \approx \text{const}$. Since longitudinal diffusion leads to decrease in θ and c^+ , the relative changes $\Delta \theta_2$ and Δc_2^+ will decrease with further increase in the flow separation coefficient. This, in particular, explains the fact that values of c^+ and θ calculated with the ideal displacement and total mixing models practically coincide for flow separation coefficients close to unity. In addition, we must note that longitudinal diffusion can have a significant effect on the value of the nonpenetrating flow $(1 - \theta)G$ and in regions of large θ the relative deviation $\Delta(1 - \theta)_2$ will exceed $\Delta \theta_2$. This fact can prove important in cases where the element is intended for production of a product in the residual flow and is used in the high θ range.

On the basis of the above results, we may conclude that the degree of influence of longitudinal diffusion on the separation process is characterized by Pe_2 and θ . In connection with this it will be desirable to introduce a classification of elements with respect to the flow separation coefficient.

We can arbitrarily divide the majority of membrane elements into three categories with respect to separation characteristics. The first category includes membrane elements with a flow separation coefficient $\theta \leq 0.2$, used to obtain mixtures with a relatively high content of the easily penetrating component in the penetrating flow. The second category consists of elements with coefficients $0.2 < \theta < 0.8$, used in multistage equipment such as counterflow stages or columns, used to obtain high separation coefficients while maintaining economy in raw materials [11]. Finally, the third class consists of membrane elements with separation coefficients close to unity ($\theta \geq 0.8$). These are used to obtain mixtures with a low content of the easily penetrating component in the nonpenetrating flow.

Using the above classification, we will evaluate the characteristic values of Pe_2 at which deviations of separation characteristics from the values calculated with the ideal displacement model do not exceed a specified value.

For elements in the first category the effect of longitudinal diffusion is insignificant (Fig. 2a) and $Pe_2 \approx 1$. Values of $\Delta\theta_2$, Δc_2^- and Δc_2^+ in elements of the second type are low at $Pe_2 \geq 10-50$ (Fig. 2b). In addition, because of the series connection involved, the effect of longitudinal diffusion on separation characteristics of a multistage device increases as compared to the effect of diffusion on the separation process in an individual stage. Thus, for example, it was shown in [12] that for other conditions equal the net membrane area in a chain of elements with total mixing is 20% greater than in a chain of ideal displacement elements.

For elements of the third type the corresponding Pe_2 value must be chosen such that the quantities Δc_2^- and $\Delta(1-\theta)_2$ are sufficiently small (in Fig. 2b $Pe_2 \geq 100$).

It must be stressed again that the classification presented above is quite arbitrary and the limits of one or the other category can be shifted toward either increased or decreased flow separation coefficient. Moreover, the estimates of the numbers Pe_2 were performed for fixed values of the deviations $\Delta\theta_2$, Δc_2^- and Δc_2^+ . It is obvious that with increase in the range of admissible $\Delta\theta_2$, Δc_2^- , Δc_2^+ values the corresponding value of Pe_2 will decrease.

To evaluate the numbers Pe_1 and Pe_2 together with the degree of deviation of operation of a real membrane apparatus from the ideal displacement regime due to longitudinal diffusion we will consider the example of separation of the binary mixture O_2-N_2 ($c_{O_2} = 0.21$, $c_{N_2} = 0.79$) in filaments of poly-4-methylpentane-1 (selectivity $\alpha_{N_2/O_2}^* = 0.303$ [13], $r_0 = 1.26 \cdot 10^{-4}$ m, $r_1 = 1.5 \cdot 10^{-4}$ m, $l_M = 0.35$ m) at a pressure ratio $\gamma = 0.17$.

Figure 3 (curve 1) shows the dependence of the ratio of productivities for the nonpenetrating flow for the ideal displacement regime g_1 and with consideration of longitudinal diffusion g_2 , where $g_{1,2} = (1 - \theta_{1,2})G_{1,2}$, on oxygen concentration in the nonpenetrating flow. If the element operates in the range of low flow separation coefficients ($\theta = 0.2$, $c_{O_2}^- = 0.16$) then the numbers Pe_1 and Pe_2 are quite high (220.0 and 174.2, respectively) and longitudinal diffusion has practically no effect on element output. If the element is of the third type ($\theta = 6.88$, $c_{O_2}^- = 0.02$), then the values of Pe_1 and Pe_2 decrease markedly (to 44.0 and 5.5), and longitudinal diffusion leads to a reduction in output in the nonpenetrating flow by 80% as compared to the ideal displacement regime. It is possible to decrease the influence of longitudinal diffusion by increasing the values of Pe_1 and Pe_2 and simultaneously decreasing the coefficient θ at which it is possible to achieve the required O_2 concentration in the nonpenetrating flow. Curve 2 of Fig. 3 corresponds to an element in which a filament with smaller cross section is used ($r_0 = 1.26 \cdot 10^{-5}$ m, $r_1 = 3.75 \cdot 10^{-5}$ m). In this case at $c_{O_2}^- = 0.02$ ($\theta = 0.85$, $Pe_1 = 900.0$, $Pe_2 = 135.2$) the reduction in productivity due to longitudinal diffusion is only 5%. Pressure losses in motion of the mixture being separated along the high pressure channel caused by viscous character of flow, efflux of mixture through the lateral surface, and change in molar mass of the mixture do not exceed 1%. Further reduction in internal radius of the filament leads to a significant increase in pressure losses over the element length and degradation of the element separation characteristics due to attenuation of the driving force behind the separation process [14].

The example presented illustrates the possibility that geometric parameters of the element may have a significant effect on the efficiency of its operation, which should be con-

sidered in designing membrane equipment. To determine the degree of influence of longitudinal diffusion on element separation characteristics and find optimum methods for decreasing this effect, one can use curves such as those presented in Fig. 4. As a parameter here we use the quantity B from Eq. (4), which is defined by the equipment geometry and original flow parameters. Figure 4 does not show the functions $\log Pe_2(B)$ at $\Delta c_{1,2}^+ = \text{const}$, since as follows from Fig. 2, these curves have the same form as at $\Delta \theta_{1,2} = \text{const}$.

Using curves 1 and 3 of Fig. 4, values of Pe_2 can be determined below which for a given B the element characteristics will be close to the case of total mixing. With decrease in B these values increase, and in the region of low B the relative deviation $\Delta \theta_2$ will also be low for a large Pe_2 interval. If lines of constant $\Delta(1 - \theta)_{1,2}$ were drawn in Fig. 4, they would appear similar to the lines of constant $\Delta c_{1,2}^-$.

The character of the dependence of Pe_2 on the parameter B permits distinguishing certain regions in the plane of the function $\log Pe_2(B)$: region I, values of B and Pe_2 at which element separation characteristics obtained in the total mixing or ideal displacement approximation are similar to each other; II, in which element characteristics correspond to an element with ideal displacement; III, in which the separation process occurs in a regime close to total mixing; IV, in which the effect of longitudinal diffusion must be considered in calculating element separation characteristics (Fig. 4).

By using the expressions for B and Pe_2 an optimal method of reducing the effect of longitudinal diffusion on the separation process can be chosen. Thus, for example, if the geometric dimensions of the apparatus and mixture and original flow parameters correspond to regions III or IV, then proportional increase in apparatus length and membrane thickness can force the separation process to approach the ideal displacement regime (region II). If it is not possible to produce a flow in the element close to the ideal displacement regime, the organization of the flow in the low pressure volume has practically no effect on the separation process and use of a counterflow configuration leads to no noticeable improvement in element separation characteristics.

The majority of the results presented herein were obtained for highly selective membranes ($\alpha^* = 0.031$). For the majority of polymers and mixtures lower selectivity values are characteristic [13], as a result of which the flow which does not penetrate the membrane will be less depleted of the more permeable component and the concentration gradient will be significantly less than in the case considered herein. In connection with this fact the quantitative results obtained herein can be used as estimates of the upper limit of the effect of longitudinal diffusion on separation characteristics of real membrane apparatus.

In conclusion, we note that study of the degree of influence of longitudinal diffusion in the low pressure volume on element separation characteristics is of independent interest. Study of that problem will permit an answer to the question of the practical possibility of realizing an ideal counterflow element.

NOTATION

c, c^+, c^- , relative molar concentrations of easily diffusing component in original, penetrating, and nonpenetrating flows; D_{12} , mutual diffusion coefficient of mixture components, m^2/sec ; G , molar flow rate of initial flow, mol/sec ; L , convective flow of mixtures in channel section with longitudinal coordinate ℓ , mol/sec ; ℓ_M , separation channel length, m ; P_1 , gas mixture pressure in original flow; Q_1, Q_2 , permeabilities of mixture components through wall material, $\text{mol}\cdot m/(\text{m}\cdot\text{sec}\cdot\text{Pa})$; R , universal gas constant, $8.314 \text{ J}/(\text{mol}\cdot\text{deg})$; r_1, r_0 , internal and external channel radii, m ; S_M , total area of side walls, m^2 ; S_\perp , channel cross section area, m^2 ; T , mixture temperature; x, x' , easily penetrating component concentration in cross section ℓ and beyond channel wall, respectively; γ , pressure ratio in penetrating and original flows, $\gamma < 1$; θ , flow separation coefficient, equal to ratio of penetrating and original flow rates.

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HEAT AND MASS TRANSFER IN THE STEADY WALL COMBUSTION
OF A FILTRATED GAS MIXTURE

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UDC 536.46

The influence of external and internal heat and mass transfer on the temperature distribution in the solid and gas phases and on the stability of the combustion zone in "standing-wave" conditions is investigated.

Surface combustion of filtrated fuel, which occurs in porous radiators, refractory preheaters, and chemical-technology processes and apparatus, is a complex phenomenon whose analysis entails taking account of heat and mass transfer of interacting media and the chemical kinetics of homogeneous reaction of the components. In [1, 2], where surface combustion was investigated for a porous radiator, it was assumed that the temperatures of the body skeleton $t_T = (T_T - T_\infty)/T_\infty$ and the filtrated fuel $t_I = (T_I - T_\infty)/T$ are the same.

In the present work, the results of theoretical investigation of the surface combustion of filtrated fuel are given, together with an analysis of the conditions corresponding to a "standing wave," taking account of the difference in t_T and t_I . The results of analytical investigations are compared with experiment.

The problem is formulated as follows. A stoichiometric gas mixture is filtered through a semiinfinite body (Fig. 1), consisting of a chemically inert porous medium ($-\infty \leq x \leq 0$) with impermeable heat-insulated side walls, at a velocity $v = (j/\rho)_I$, where j_I is the total density of a longitudinal flux of injectant consisting of fuel j_f and oxidant j_o ($j_I = j_f + j_o$); ρ_I is the gravimetric density of the gas mixture. It is assumed that the chemical reaction of the porous medium with the injected gas does not occur; the thermophysical properties of the interacting media and the porosity of the medium do not change in the course of the process; the rate of chemical combustion is infinitely large in comparison with the diffusion rate, i.e., the flame-front model is valid; filtrational effects may be neglected. The latter assumption, allowing the filtration equation to be eliminated from consideration, does not lead to significant calculation errors, since the present work is an investigation of conditions of low injection rate of injectant relative to a porous radiator with surface combustion, for which $v_\infty = 0.37-0.172$ m/sec [1, 2], at large filtration coefficients and porosity of the body skeleton $\Pi = 80\%$.

The following physical picture is considered. Combustion of the injectant is initiated by a source (acting only at the initial moment of time) at the end surface of the body ($x = 0$).

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