

EFFECT OF GEL FORMATION ON THE PROCESS OF LAMINAR CONTINUOUS-FLOW ULTRAFILTRATION

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We investigate the process of ultrafiltration with gel formation in laminar flow in a plane channel in the case of nonideal selectivity of the membrane.

The technology and facilities of membrane processes are important areas of scientific research such as the production of new membranes with improved characteristics.

Extensive use is made of plane-frame membrane apparatuses based on a slotted channel with membranes placed (laid out) on its walls. In view of the large specific surface of the membranes laid out (a small height of the intermembrane channel), ultrafiltration is usually carried out in a laminar flow regime and is characterized by formation of a gel layer on the surface of the membranes. To find a means for effective separation of solutions, it is necessary to conduct a careful investigation of two factors: the selectivity and productivity of the given membrane. They can be studied only on the basis of ideas about the real mechanism of mass transfer through a membrane and the processes occurring on its surface. The theoretical description of the latter in the case of laminar continuous-flow ultrafiltration in plane slotted channels cannot be considered satisfactory [1], and the present work is intended to fill this gap to some extent.

We formulate a transfer equation for the process of laminar ultrafiltration. First, we examine the kinetic equation of gel formation, which is often written in incorrect form [2].

We set up the balance of the mass supplied to and removed from the membrane under the condition of ideal selectivity of it:

$$C_g n_t' = - C_g V n_0 + DC_n' |_{f_t}. \quad (1)$$

The equation of the gel surface is specified in explicit form (see Fig. 1):

$$y = f(x, t),$$

then for a unit normal to the surface

$$n_0 = (-f_x' i + j) / \sqrt{1 + (f_x')^2}.$$

We note that

$$dy = f_x' dx + f_t' dt, \quad \delta n = \delta n n_0 = f_t' dt / \sqrt{1 + (f_x')^2}.$$

We take into account that

$$V = \hat{u}i - \hat{v}j,$$

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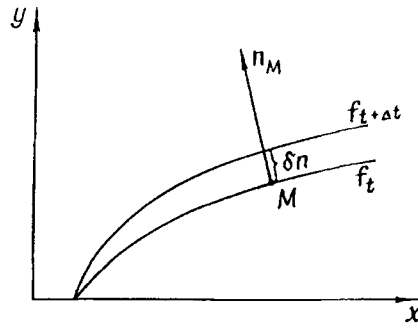


Fig. 1. Formation of a gel layer.

$$C'_n|_f = n_0 (C'_x i + C'_y j)$$

and from the balance equation we obtain the kinetic condition for formation of a gel layer on the membrane surface in the plane case:

$$C_g f'_t = C_g (\hat{u} f'_x + \hat{v})_f + D (-f'_x C'_x + C'_y)_f.$$

Below, we will assume the gel layer on the membrane to be immovable, forming, as it were, a continuation of the membrane; as a result, we have

$$\hat{u}|_f f'_x = 0, \quad f'_x C'_x|_f = 0$$

by virtue of the fact that $C|_f = C_g = \text{const}$. Thus, the kinetic equation for the gel layer takes the form

$$C_g f'_t = C_g \hat{v}_f + D C'_y|_f. \quad (2)$$

In the case of nonideal selectivity of the membrane, the mass balance should take into account withdrawal of solute with the permeate, i.e.,

$$C_g f'_t = C_g (1 - C_{fi}/C_g) \hat{v}_f + D C'_y|_f. \quad (3)$$

Here C_{fi} is the concentration of solute in the permeate.

With allowance for gel formation we obtain the velocity distribution in a plane slotted channel for a laminar regime of flow. We will assume that the flow at the channel inlet is fully developed (since the length of the channel greatly exceeds its height, the inlet hydrodynamic section can be eliminated from consideration). The flow rate of the liquid through the cross section of the channel considerably exceeds the flow passing through the membrane, while the thickness of the gel layer f is much smaller than the half-height of the channel h . In this case the equations of motion and continuity acquire the form

$$-P'_x + \mu \hat{u}''_{yy} = 0, \quad (4)$$

$$P'_y = 0, \quad (5)$$

$$\hat{u}'_x + \hat{v}'_y = 0. \quad (6)$$

The boundary conditions are

$$\hat{u} = 0, \quad \hat{v} = -\hat{V}_f \quad (y = f);$$

$$\hat{v} = 0, \quad \hat{u}'_y = 0 \quad (y = h). \quad (7)$$

From Eq. (5) it follows that $P = P(x)$. Integrating Eq. (4), taking account of conditions (7), we find

$$\hat{u} = - (h^2/\mu) P'_x \left(\frac{y-f}{h} - \frac{y^2-f^2}{2h^2} \right). \quad (8)$$

The continuity equation yields

$$\hat{v} = - \frac{h^3}{3\mu} \frac{\partial}{\partial x} \left[P'_x \left(1 - \frac{3f}{h} - \frac{3(y-f)(y+f)}{2h^2} + \frac{y(y-f)(y+f)}{2h^3} - yf \frac{(f/h-3)}{h^2} \right) \right]. \quad (9)$$

From this it follows that

$$\hat{V}_f = \frac{h^3}{3\mu} \frac{\partial}{\partial x} \left[P'_x \left(1 - \frac{f}{h} \right)^3 \right]. \quad (10)$$

We determine the mean flow rate

$$\bar{u}_x = \frac{1}{h-f} \int_f^h \hat{u} dy = - \frac{h^2}{3\mu} \frac{\partial P}{\partial x} \left(1 - \frac{f}{h} \right)^2. \quad (11)$$

Then at the channel inlet at $x = 0$

$$\bar{u}_0 = - \frac{h^2}{3\mu} \frac{\partial P}{\partial x}. \quad (12)$$

Integrating Eq. (10) with allowance for relation (12) and substituting the result into Eq. (8), we obtain an expression for the longitudinal velocity component:

$$\hat{u} = \frac{3}{h(1-f/h)^3} \left(\bar{u}_0 h - \int_0^x \hat{V}_f dx \right) \left(\frac{y-f}{h} - \frac{y^2-f^2}{2h^2} \right). \quad (13)$$

To describe the phenomenon of concentration polarization, i.e., gel formation, we use an equation of convective diffusion that describes the process of substance accumulation at a given point of space as a function of time. With allowance for the above assumptions it takes the form

$$C'_t + \hat{u}C'_x + \hat{v}C'_y = DC''_{yy}. \quad (14)$$

The process of separation of solutions by the method of ultrafiltration is characterized by large Schmidt numbers, and therefore the characteristic time of the change in the concentration field considerably exceeds the characteristic time of the change in the velocity profile. Consequently, the dynamic problem can be considered to be stationary (this was done above), i.e., each instantaneous distribution of concentration corresponds to its own stationary distribution of velocities at the given moment. The boundary conditions before the point of gel formation on the membrane and after it will be substantially different:

$$\begin{aligned} \hat{u}|_{y=0} = 0, \quad \hat{v}|_{y=0} = -\hat{V} \quad (\hat{V} = \text{const}), \quad \phi \hat{V} C_w + DC'_y|_{y=0} = 0, \\ C_y|_{y=h} = 0, \quad C|_{x=0} = C_0, \quad C|_{t=0} = C_0 \end{aligned} \quad (15)$$

for $0 \leq x \leq x_1$ and

$$\hat{u}|_{y=f} = 0, \quad \hat{v}|_{y=f} = -\hat{V}_f, \quad \varphi \hat{V}_f C_g + DC'_y|_{y=f} = C_g f'_t, \quad (16)$$

$$C'_y|_{y=h} = 0, \quad f(x, t)|_{x=x_1} = 0, \quad C|_{y=f} = C_g$$

for $x \geq x_1$ (after the point of gel formation). We connect the drop in the permeability of the membrane with the thickness of the gel layer and represent it in the form

$$\hat{V}_f = \frac{\hat{V}}{1 + \hat{k}_f}, \quad (17)$$

where \hat{k} is the characteristic coefficient of the hydraulic resistance of the gel layer. Equations (9), (10), (13)-(17) form a closed system and make it possible to describe the process of laminar ultrafiltration in a plane slotted channel.

A method for solving the equations of convective diffusion in a channel without gel formation is suggested in [3].

We will employ this method and find a solution to problem (14)-(16) with allowance for gel formation. For convenience, the equation of convective diffusion (14) will be written in a conservative form (here and below we will use dimensionless quantities):

$$\frac{\partial (\Theta - 1)}{\partial \tau} + \frac{\partial u (\Theta - 1)}{\partial \xi} + \frac{\partial v (\Theta - 1)}{\partial \eta} = \frac{1}{Pe} \frac{\partial^2 (\Theta - 1)}{\partial \eta^2}.$$

We integrate this equation across the diffusional boundary layer and use boundary conditions (15) or (16) and the requirements usually applied in boundary-layer theory $\Theta = 1$, $\Theta'_\eta = 0$ at $\eta = \Delta$. Then, before the point of gel formation ($0 \leq \xi \leq \xi_1$) we have

$$\frac{\partial}{\partial \tau} \int_0^\Delta (\Theta - 1) d\eta + \frac{\partial}{\partial \xi} \int_0^\Delta u (\Theta - 1) d\eta = V \Gamma_w, \quad (18)$$

and after the point of gel formation ($\xi > \xi_1$) we have

$$\frac{\partial}{\partial \tau} \int_0^\Delta (\Theta - 1) d\eta + (2\Theta_g - 1) \frac{\partial \delta}{\partial \tau} + \frac{\partial}{\partial \xi} \int_\delta^\Delta u (\Theta - 1) d\eta = V_\delta \Gamma_g. \quad (19)$$

Here $\Gamma_g = 1 - (1 - \varphi)\Theta_g$, $\Gamma_w = 1 - (1 - \varphi)\Theta_w$.

At the very point of gel formation ($\xi = \xi_1$) the condition $\Theta = \Theta_g$ is satisfied. Let us analyze the stationary regime of ultrafiltration in a plane slotted channel. Because of the very large Peclet numbers ($Pe \simeq 10^7$), the thicknesses of the gel layer δ and the diffusional boundary layer Δ will be much smaller than the half-height of the channel, i.e., they lie in the region adjacent to the membrane. This fact considerably simplifies the solution of the problem. It can be assumed that the gel layer does not change the inner height of the channel. The presence of the gel exhibits a direct effect only on the total hydraulic resistance of the membrane and the gel. Therefore, in distributions (9), (10), (11), and (13) the terms f/h can be neglected compared to unity. Moreover, in solving the diffusional problem we can restrict ourselves to just the first terms in the transverse coordinate y in determining the velocity profiles (9) and (13). As a result, the equations of convective diffusion take the following form: before the gel formation point ($0 \leq \xi \leq \xi_1$)

$$3(1 - V\xi) \eta \frac{\partial \Theta}{\partial \xi} - V \frac{\partial \Theta}{\partial \eta} = \frac{1}{Pe} \frac{\partial^2 \Theta}{\partial \eta^2},$$

$$\varphi^{\Theta_w} V + \frac{1}{\text{Pe}} \frac{\partial \Theta}{\partial \eta} = 0 \quad (\eta = 0); \quad (20)$$

and after the gel formation point ($\xi \geq \xi_1$)

$$3 \left(1 - V\xi_1 - \int_{\xi_1}^{\xi} V_{\delta} d\xi \right) (\eta - \delta) \frac{\partial \Theta}{\partial \xi} - V_{\delta} \frac{\partial \Theta}{\partial \eta} = \frac{1}{\text{Pe}} \frac{\partial^2 \Theta}{\partial \eta^2},$$

$$\varphi^{\Theta_g} V_{\delta} + \frac{1}{\text{Pe}} \frac{\partial \Theta}{\partial \eta} = 0 \quad (\eta = \delta), \quad \Theta = \Theta_g \quad (\eta = \delta) \quad V_{\delta} = V/(1 + k/\delta). \quad (21)$$

The stationary distribution of concentration near the membrane has the form

$$\Theta = \Theta_w(\xi) [1 - \varphi (1 - \exp(-\text{Pe} V\eta))], \quad \xi \leq \xi_1, \quad (22)$$

$$\Theta = \Theta_g(\xi) [1 - \varphi (1 - \exp(-\text{Pe} V_{\delta}(\eta - \delta)))] , \quad \xi > \xi_1 .$$

From the physical considerations underlying boundary-layer theory and Eqs. (22) the distribution of concentration in a plane slotted channel can be represented for $0 \leq \xi \leq \xi_1$ as

$$\Theta = \begin{cases} \Theta_w(\xi) [1 - \varphi (1 - \exp(-\text{Pe} V\eta))], & 0 \leq \eta \leq \Delta, \\ 1, & \Delta \leq \eta = 1, \end{cases} \quad (23)$$

and for $\xi \geq \xi_1$ as

$$\Theta = \begin{cases} \Theta_g, & 0 \leq \eta \leq \delta, \\ \Theta_g [1 - \varphi (1 - \exp(-\text{Pe} V_{\delta}(\eta - \delta)))] , & \delta \leq \eta \leq \Delta, \\ 1, & \Delta \leq \eta = 1, \end{cases} \quad (24)$$

where $\Theta_w(\xi)$ and $\delta(\xi)$ are functions as yet unknown. For their determination we use integral equations (18) and (19). First, from relations (23) and (24) we find

$$\Delta = \frac{1}{\text{Pe}V} \ln \frac{\varphi^{\Theta_w}}{\Gamma_w}, \quad 0 \leq \xi \leq \xi_1; \quad \Delta - \delta = \frac{1}{\text{Pe}V_{\delta}} \ln \frac{\varphi^{\Theta_g}}{\Gamma_g}, \quad \xi > \xi_1 .$$

From this it is clear that formation of gel on the membrane is possible when $\Theta_g < 1/(1 - \varphi)$.

Substituting the distribution of velocities from formulas (20), (21) and concentrations from formulas (23), (24) into Eqs. (18), (19), integrating once over ξ , and performing simple calculations, we obtain for $0 \leq \xi \leq \xi_1$

$$\Theta_w - 1 - \Gamma_w \ln \frac{\varphi^{\Theta_w}}{\Gamma_w} - \frac{\Gamma_w}{2} \left(\ln \frac{\varphi^{\Theta_w}}{\Gamma_w} \right)^2 = \frac{(\text{Pe}V)^2 V}{3(1 - V\xi)} \int_0^{\xi} \Gamma_w d\xi \quad (25)$$

and for $\xi \geq \xi_1$

$$3 \left(1 - V\xi_1 - \int_{\xi_1}^{\xi} V_{\delta} d\xi \right) \Sigma_g / (\text{Pe}V_{\delta})^2 = \int_{\xi_1}^{\xi} \Gamma_g V_{\delta} d\xi + \int_0^{\xi_1} \Gamma_w V d\xi . \quad (26)$$

For writing more concisely, we denote

$$\Sigma_g = \Theta_g - 1 - \Gamma_g \ln \frac{\varphi^{\Theta_g}}{\Gamma_g} - \frac{\Gamma_g}{2} \left(\ln \frac{\varphi^{\Theta_g}}{\Gamma_g} \right)^2 .$$

Now we will estimate the position of the point of gel formation on the membrane. From formula (25) or (26) it follows that

$$\int_0^{\xi_1} \Gamma_w V d\xi = 3 \Sigma_g \frac{1 - V\xi_1}{(\text{Pe}V)^2}. \quad (27)$$

In the case of ideal selectivity formula (26) takes the simpler form

$$V\xi_1 = [1 + (\text{Pe}V)^2/3 (\Theta_g - \ln \Theta_g - (\ln \Theta_g)^2/2 - 1)]^{-1}. \quad (27')$$

Analytical and numerical calculations [3] demonstrate the nonlinear character of the dependence of Θ_w on ξ . In place of Θ_w we substitute into the left-hand side of Eq. (27) two limiting cases: a) $\Theta_w = \Theta_g$ and b) the linear behavior $\Theta_w = 1 + V\xi(\Theta_g - 1)/V\xi_1$.

Integrating Eq. (27), we obtain the estimate

$$\frac{3\Sigma_g}{\Gamma_g (\text{Pe}V)^2} \geq \frac{V\xi_1}{(1 - V\xi_1)} \geq \frac{6\Sigma_g}{(\varphi + \Gamma_g) (\text{Pe}V)^2}.$$

Equations (25)-(27) give a full description of the phenomenon of concentration polarization in ultrafiltration in a plane slotted channel before the gel-formation point.

We now analyze the process of ultrafiltration in a channel after the gel-formation point. Solving Eq. (26) for the integral and then differentiating with respect to ξ with subsequent integration with the boundary condition $V_\delta = V$ at $\xi = \xi_1$, we find a transcendental equation that describes the process of ultrafiltration with gel formation.

We denote $3\Sigma_g/\Gamma_g \text{Pe}^2 V^2 = F$. Then

$$\begin{aligned} \frac{1 - V\xi}{1 - V\xi_1} &= \frac{V_\delta}{V} (1 + F) / \left[\left(\frac{V_\delta}{V} \right)^2 + F \right] - \\ &- (1/F)^{1/2} (1 + F) [\arctan (1/F)^{1/2} - \arctan (V_\delta/VF)^{1/2}]. \end{aligned} \quad (28)$$

For $\varphi = 1$

$$\begin{aligned} V\xi &= 1 - \frac{V_\delta}{V} / \left(\frac{V\xi_1}{1 - V\xi_1} + \frac{V_\delta^2}{V^2} \right) + \\ &+ \left(\frac{1 - V\xi_1}{V\xi_1} \right)^{1/2} \arctan \left(\frac{1 - V\xi_1}{V\xi_1} \right)^{1/2} \left(1 - \frac{V_\delta}{V} \right) / \left(1 + \frac{V_\delta}{V} \frac{1 - V\xi_1}{V\xi_1} \right). \end{aligned} \quad (28')$$

If $V\xi_1 \rightarrow 0$, we can obtain a simpler solution:

$$\frac{V_\delta}{V} = [1 + (V\xi - V\xi_1) \Gamma_g \text{Pe}^2 V^2 / 2\Sigma_g]^{-1/3} \quad (29)$$

or in the case of $\varphi = 1$

$$\frac{V_\delta}{V} \approx \left(\frac{3V\xi}{2V\xi_1} - \frac{1}{2} \right)^{-1/3}. \quad (29')$$

When $V\xi/V\xi_1 \gg 1$, Eq. (29) takes the form

$$V_\delta \approx (2 \Sigma_g / \Gamma_g \text{Pe}_\xi^2)^{1/3}, \quad (30)$$

and for $\varphi = 1$

$$V_\delta \approx \left[2 \left(\Theta_g - \ln \Theta_g - \frac{\ln^2 \Theta_g}{2} - 1 \right) / \text{Pe}_\xi^2 \right]^{1/3}. \quad (30')$$

Since the velocity V does not enter into formula (30), this means that the transmembrane velocity V_δ is independent of the pressure (the resistance of the gel layer considerably exceeds the resistance of the membrane, and an increase in the pressure is compensated for by an increase in the resistance of the gel layer).

Thus, the pattern of laminar continuous-flow ultrafiltration in a plane channel can generally be divided into three regions.

In the first region – from the inlet into the channel and before the gel-formation point (determined from Eq. (27)) – the main resistance to transmembrane flow is offered by the membrane. In this section the filtration velocity V will be directly proportional to the pressure applied (for the resistance of membranes Darcy's law is valid [4], which has proved to be very efficient).

In the second region – from the point of gel formation and farther downstream along the main flow – the hydraulic resistances of the membrane and the gel layer will be of the same order of magnitude. Here the filtration velocity V_δ of (28) depends nonlinearly on the pressure (it is associated with V).

In the third region, when the hydraulic resistance of the gel layer considerably exceeds that of the membrane, the filtration velocity V_δ ceases to depend on the initial pressure, and all the distributions over the filtration velocity from the above two regions reduce to the single dependence (30).

Let us now dwell on nonstationary filtration with gel formation. From physical considerations the nonstationary process can be considered as two limiting cases: highly nonstationary and stationary. For the especially nonstationary regime

$$\begin{aligned} \frac{\partial}{\partial \tau} \int_0^\Delta (\Theta - 1) d\eta &= \Gamma_w V, \quad 0 \leq \tau \leq \tau_1, \\ \frac{\partial}{\partial \tau} \int_\delta^\Delta (\Theta - 1) d\eta + (2\Theta_g - 1) \frac{\partial \delta}{\partial \tau} &= \Gamma_g V_\delta, \quad \tau \geq \tau_1. \end{aligned} \quad (31)$$

Here the time τ_1 is the beginning of gel formation.

To solve the nonstationary problem we use a generally applied approach where the nonstationary distribution of concentration is prescribed from the solution of the stationary problem. Then, from Eq. (31), with account for distributions (23) and (24), on condition that Θ_w and δ depend on the time and with the obvious relationship

$$\frac{\partial \delta}{\partial \tau} = - \frac{V}{k V_\delta^2} \frac{\partial V_\delta}{\partial \tau}$$

we obtain:

$$\begin{aligned} \Theta_w - \Gamma_w \ln \frac{\varphi \Theta_w}{\Gamma_w} - 1 &= \text{Pe} V \int_0^\tau \Gamma_w V d\tau, \quad 0 \leq \tau \leq \tau_1, \\ \frac{V_\delta}{V} &= \left[1 + \frac{2(V\tau - V\tau_1)}{\Sigma_\tau / \Gamma_g \text{Pe} V + (2\Theta_g - 1)/k} \right]^{-1/2}, \quad \tau \geq \tau_1. \end{aligned} \quad (32)$$

Here $\Sigma_\tau = \Theta_g - 1 - \Gamma_g \ln \varphi \Theta_g / \Gamma_g$.

Let us estimate the beginning of the time of gel formation. We proceed similarly to the stationary case.

Then

$$\frac{\Sigma_\tau}{\text{Pe}V\Gamma_g} \geq V\tau_1 \geq \frac{2\Sigma_\tau}{\text{Pe}V(\varphi + \Gamma_g)}. \quad (33)$$

For $\varphi = 1$

$$V\tau_1 = (\Theta_g - \ln \Theta_g - 1)/\text{Pe}V. \quad (33')$$

It is pertinent here to note that formulas (32) and (33) are also valid for describing the process of ultrafiltration in a cell without forced mixing. They make it possible to determine the diffusion coefficient of the solute in the solution from the time dependence of the specific performance recorded experimentally in a cell without mixing [5].

If $V\tau_1 \rightarrow 0$ and the times considered are such that $V\tau/V\tau_1 \gg 1$, then the filtration velocity will decrease in time according to the relation

$$\frac{V_\delta}{V} \sim \left(1 + \frac{2kV\tau}{2\Theta_g - 1}\right)^{-1/2}. \quad (34)$$

In conclusion, we estimate the time τ_s needed to attain the stationary regime of ultrafiltration in a plane channel with gel formation on the membrane. Assuming $V\xi_1$ to be small, we equate the right-hand sides of Eqs. (29) and (32):

$$V\tau_s = V\tau_1 + \frac{1}{2} \left[\left(\frac{(V\xi - V\xi_1)\Gamma_g \text{Pe}^2 V^2}{2\Sigma_g} + 1 \right)^{2/3} - 1 \right] \left(\frac{2\Theta_g - 1}{k} + \frac{\Sigma_\tau}{\Gamma_g \text{Pe}V} \right). \quad (35)$$

For distances from the channel inlet for which $V\xi/V\xi_1 \gg 1$, we obtain a simpler relation:

$$V\tau_s = \frac{1}{2} \left(\frac{V\xi\Gamma_g \text{Pe}^2 V^2}{2\Sigma_g} \right)^{2/3} \left(\frac{2\Theta_g - 1}{k} + \frac{\Sigma_\tau}{\Gamma_g \text{Pe}V} \right), \quad (36)$$

which for $\varphi = 1$ has the form

$$V\tau_s = \left(\frac{3V\xi}{2V\xi_1} \right)^{2/3} \left(\frac{2\Theta_g - 1}{2k} + \frac{V\tau_1}{2} \right). \quad (36')$$

Thus, the theory suggested offers a description of laminar ultrafiltration in a plane channel with gel formation on the membrane and allows one to compare experimental and theoretical data.

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NOTATION

$\xi = x/h$, $\eta = y/h$, dimensionless longitudinal and transverse coordinates; $\tau = tu_0/h$, $u = \hat{u}/u_0$, $v = \hat{v}/u_0$, dimensionless time and velocity vector components; h , half-height of the plane channel; u_0 , mean velocity at the channel inlet; t , time; $\text{Pe} = u_0 h/D$, diffusional Peclet number; D , diffusion coefficient; μ , coefficient of dynamic viscosity; f , thickness of the gel layer; $\delta = f/h$, dimensionless thickness of the gel layer; $\Delta = \Delta/h$, dimensionless thickness of the diffusional boundary layer; $\Theta = C/C_0$, dimensionless concentration of the solute; $\Theta_w = C_w/C_0$,

$\Theta_g = C_g/C_0$, dimensionless concentrations on the wall and of gel formation; $k = \bar{k}h$, dimensionless characteristic coefficient of hydraulic resistance of the gel layer; φ , selectivity coefficient of the membrane; $V = \hat{V}/u_0$, $V_\delta = \hat{V}_f/u_0$, dimensionless transmembrane velocity.

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