GEL FORMATION IN ULTRAFILTRATION IN A PLANE CHANNEL WITH ONE PERMEABLE SURFACE

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We investigate the unsteady-state laminar process of ultrafiltration in a plane channel with one permeable surface in the stage of stationary gelatination.

The formation of a relatively impermeable gel layer on a membrane surface as a result of the concentration polarization phenomenon is one of the main reasons for decreasing the throughput of membrane ultrafiltration flow-through apparatuses [1]. In this connection a clear physical picture of processes occurring in such devices makes it possible to solve many problems associated with the development and creation of new, more effective ultrafiltration apparatuses.

In [2], an unsteady laminar ultrafiltration process was investigated in a plane slot channel with one permeable surface on the segment from the channel inlet to the point where the concentration on the membrane surface reached its maximum value, which is called the gel-formation concentration. The filtration velocity on this segment is directly proportional to the applied pressure, and the main resistance to transmembrane mass exchange is exerted by the membrane.

In the present work we will consider the second stage of the process when the concentration on the membrane surface reaches the gel-formation concentration. From this point on the membrane surface a thin film of high-molecular compounds (gel) is formed, which exerts an additional resistance to transmembrane mass exchange. The resulting gel layer will be considered stationary (see a figure in [2]).

We will describe the unsteady laminar regime of ultrafiltration in a plane channel with one permeable surface by an equation of convective diffusion, which for convenience will be presented in dimensionless form:

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

Let ξ_1 be the point beginning with which the formation of a gel layer on the membrane surface occurs. Then for $\xi > \xi_1$ the following boundary conditions will be fulfilled:

$$u|_{\eta=\delta} = 0 ; \quad v|_{\eta=\delta} = - V_\delta ;$$

$$\varphi V_\delta \Theta_g + \frac{1}{Pe} \frac{\partial \Theta}{\partial \eta} \bigg|_{\eta=\delta} = \Theta_g \frac{\partial \delta}{\partial \tau} ; \tag{2}$$

$$\Theta|_{\eta=2h} = 1 ; \quad \Theta|_{\eta=\delta} = \Theta_g ; \quad \delta (\xi, \tau)|_{\xi=\xi_1} = 0 .$$

Moreover, proceeding from boundary-layer theory, it can be assumed that $\Theta|_{\eta=\Delta} = 1$ and $\frac{\partial \Theta}{\partial \eta}\big|_{n=\Delta} = 0$. This means that the concentration increases only within the limits of the diffusion boundary layer. With allowance for this circumstance and boundary conditions (2) we integrate Eq. (1) across the diffusion boundary layer. As a result, we have

$$\frac{\partial}{\partial \tau} \int_\delta^\Delta (\Theta - 1)\, d\eta + (2\Theta_\delta - 1)\frac{\partial\delta}{\partial\tau} + \frac{\partial}{\partial\xi}\int_\delta^\Delta u\,(\Theta - 1)\, d\eta = V_\delta \Gamma_g\,, \tag{3}$$

where $\Gamma_g = 1 - (1 - \varphi)\Theta_g$.

Now we make use of the velocity distribution [2] for the motion of a high-molecular solution of liquids in a thin plane channel with one permeable wall under conditions of gel formation on the membrane surface:

$$\hat{u} = \frac{3}{2h\,(1 - f/2h)^3}\left(2h\bar{u}_0 - \int_0^x V_f\,dx\right)\left(\frac{y - f}{h} - \frac{y^2 - fy}{2h^2}\right), \tag{4}$$

$$\hat{v} = \frac{-V_f + 3\,(1 - f/2h)\left(2\bar{u}_0 h - \int_0^x V_f\,dx\right)\frac{\partial f}{\partial x}}{(1 - f/2h)^3} \times$$

$$\times \left(1 + \frac{3f}{2h} + \frac{y^2\,(2y - 3f)}{8h^3} + \frac{3y\,(2f - y)}{4h^2}\right) + \frac{2\bar{u}_0 h - \int_0^x V_f\,dx}{(1 - f/2h)^3}\left(1 - \frac{y^2}{4h^2} + \frac{y}{h}\right)\frac{3}{2h}\frac{\partial f}{\partial x}. \tag{5}$$

Since $f \ll h$, the quantities f/h and $\partial f/\partial x$ in expressions (4) and (5) can be neglected, because of their smallness in comparison with unity. Correspondingly, after making these expressions dimensionless, we have:

$$u = 3\left(1 - \frac{1}{2}\int_0^\xi V_\delta\,d\xi\right)\left(\eta - \frac{1}{2}\eta^2\right), \tag{6}$$

$$v = -V_\delta\left(1 + \frac{1}{4}\eta^3 - \frac{3}{4}\eta^2\right). \tag{7}$$

To solve the posed problem, we use a semiintegral approach, the essence of which is stated in [2-4]. Since the thickness of the diffusion boundary layer is small, we can restrict ourselves in formulas (6) and (7) to the first terms in η. The nonstationary concentration distribution will be found by solution of the following stationary problem:

$$3\left(1 - \frac{1}{2}V\xi_1 - \frac{1}{2}\int_{\xi_1}^\xi V_\delta\,d\xi\right)(\eta - \delta)\frac{\partial\Theta}{\partial\xi} - V_\delta\frac{\partial\Theta}{\partial\eta} = \frac{1}{Pe}\frac{\partial^2\Theta}{\partial\eta^2}\,, \tag{8}$$

$$\varphi V_\delta \Theta_g + \frac{1}{Pe}\frac{\partial\Theta}{\partial\eta}\bigg|_{\eta=\delta} = 0\,.$$

We relate the drop in permeability to the thickness of the gel layer and present it in the form

$$V_\delta = \frac{V}{1 + k\delta}\,. \tag{9}$$

The first terms of Eq. (8) in the immediate vicinity of the membrane surface will be small in comparison with the remaining ones. Neglecting them, we integrate Eq. (8) twice with allowance for the boundary conditions. In the end we obtain the stationary distribution of the concentration near the membrane

$$\Theta = \Theta_g\,[1 - \varphi\,(1 - \exp\,(-PeV_\delta\,(\eta - \delta)))]\,. \tag{10}$$

Then, proceeding from Eq. (10) and the physical considerations underlying boundary-layer theory, the channel-height distribution of the concentration can be presented as follows:

$$\Theta = \begin{cases} \Theta_g, & 0 \le \eta \le \delta, \\ \Theta_g \left[1 - \varphi \left(1 - \exp\left(- \mathrm{Pe}V_\delta \left(\eta - \delta \right) \right) \right) \right], & \delta \le \eta \le \Delta, \\ 1, & \Delta \le \eta \le 2. \end{cases} \tag{11}$$

Hence at $\eta = \Delta$ we find that

$$\Delta - \delta = \frac{1}{\mathrm{Pe}V_\delta} \ln \frac{\varphi \Theta_g}{\Gamma_g}. \tag{12}$$

On the basis of the hypothesis for a quasistationary state [2], the unsteady process will be presented as a combination of two processes: steady-state and especially unsteady.

Now we consider a stationary regime of gel formation. To do this, we substitute the expressions for u and Θ into Eq. (3) and then, with allowance for Eq. (12), integrate it with respect to ξ. As a result, we have:

$$3 \left(1 - \frac{1}{2} V\xi_1 - \frac{1}{2} \int_{\xi_1}^{\xi} V_\delta \, d\xi \right) \frac{\Sigma_g}{\mathrm{Pe}^2 V_\delta^2} = \int_0^{\xi_1} \Gamma_\omega V d\xi + \int_{\xi_1}^{\xi} \Gamma_g V_\delta d\xi. \tag{13}$$

For brevity, we let

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

Analyzing Eq. (13) at $\xi = \xi_1$, we evaluate the position of the point at which gel formation on the membrane begins

$$\int_0^{\xi_1} \Gamma_\omega V d\xi = \frac{3\Sigma_g (2 - V\xi_1)}{2\mathrm{Pe}^2 V^2}. \tag{14}$$

For ideal selectivity Eq. (14) takes the following form:

$$V\xi_1 = \left(\frac{1}{2} + \frac{\mathrm{Pe}^2 V^2}{3\Sigma} \right)^{-1}.$$

As indicated in [2], the dependence of the solute concentration near the membrane surface on ξ has a nonlinear character. By virtue of this fact, the position of the point of gel-formation onset can be generally evaluated from solutions of Eq. (14) for two limiting cases:

a) the linear dependence $\Theta_\omega = 1 + V\xi(\Theta_g - 1)/V\xi_1$;

b) $\Theta_\omega = \Theta_g$.

Then, from integration it follows that

$$\frac{3\Sigma_g}{(\varphi + \Gamma_g) \mathrm{Pe}^2 V^2} \le \frac{V\xi_1}{2 - V\xi_1} \le \frac{3\Sigma_g}{2\Gamma_g \mathrm{Pe}^2 V^2}. \tag{15}$$

Taking into account condition (14), we solve Eq. (13) for the integral

$$\int_{\xi_1}^{\xi} V_\delta d\xi = \frac{3 (2 - V\xi_1) \Sigma_g (V^2 - V_\delta^2)}{V^2 (3\Sigma_g + 2\Gamma_g \mathrm{Pe}^2 V_\delta^2)}. \tag{16}$$

We differentiate expression (16) with respect to ξ and then integrate it applying the boundary condition $V_{\delta\xi=\xi_1} = V$. As a result, we come to an equation that describes the steady-state ultrafiltration process in a plane channel with one permeable surface with gel formation:

$$\frac{2 - V\xi}{2 - V\xi_1} = \frac{V_\delta}{V}\left(\frac{1 + F}{F + \left(\frac{V_\delta}{V}\right)^2}\right) - \frac{1 + F}{\sqrt{F}}\left(\arctan\frac{1}{\sqrt{F}} - \arctan\frac{V_\delta}{V\sqrt{F}}\right), \tag{17}$$

where $F = 3\Sigma_g/2\Gamma_g\mathrm{Pe}^2V^2$.

For ideal selectivity ($\varphi = 1$) formula (17) takes the form:

$$V\xi = 2\left[1 - \frac{V_\delta}{V}\left(\frac{V\xi_1}{2 - V\xi_1} + \frac{V_\delta^2}{V^2}\right)^{-1} + \sqrt{\left(\frac{2 - V\xi_1}{V\xi_1}\right)}\arctan\sqrt{\left(\frac{2 - V\xi_1}{V\xi_1}\right)}\frac{1 - \frac{V_\delta}{V}}{1 + \frac{V_\delta}{V}\frac{2 - V\xi_1}{V\xi_1}}\right]. \tag{18}$$

We consider the especially nonstationary regime. Let τ_1 be the time of the gel-formation onset on the membrane surface. Then, from Eqs. (3) and (9) we obtain

$$\frac{\partial}{\partial\tau}\int_0^\Delta(\Theta - 1)\,d\eta - \frac{V}{kV_\delta^2}(2\Theta_g - 1)\frac{\partial V_\delta}{\partial\tau} = \Gamma_g V_\delta \quad (\tau \geq \tau_1). \tag{19}$$

Having calculated the integral in the left side of this expression and introducing the notation $\Sigma_\tau = \Theta_g - 1 - \Gamma_g\mathrm{Ln}(\varphi\Theta_g/\Gamma_g)$, we come to the ordinary differential equation

$$-\left(\frac{\Sigma_\tau}{\mathrm{Pe}} + \frac{V(2\Theta_g - 1)}{k}\right)\frac{1}{V_\delta^2}\frac{dV_\delta}{d\tau} = \Gamma_g V_\delta. \tag{20}$$

Its solution is as follows:

$$\frac{V_g}{V} = \left(1 + \frac{2(V\tau - V\tau_1)}{\frac{\Sigma_\tau}{\mathrm{Pe}V\Gamma_g} + \frac{2\Theta_g - 1}{k\Gamma_g}}\right)^{-1/2}. \tag{21}$$

The time of the gel-formation onset will be evaluated in much the same way as for the stationary case. Eventually, we obtain

$$\frac{2\Sigma_\tau}{\mathrm{Pe}V(\varphi + \Gamma_g)} \leq V\tau_1 \leq \frac{\Sigma_\tau}{\mathrm{Pe}V\Gamma_g}. \tag{22}$$

As is seen from Eq. (22), the time of the gel-formation onset for ideal selectivity of the membrane is determined by the formula:

$$V\tau_1 = \frac{\Theta_g - \ln\Theta_g - 1}{\mathrm{Pe}V}. \tag{23}$$

Thus, the theory suggested allows one to describe the regime of laminar ultrafiltration at the stage of gel formation in a plane channel with one permeable surface.

NOTATION

$\xi = x/h$, $\eta = y/h$, dimensionless longitudinal and transverse coordinates; $u = \hat{u}/\bar{u}_0$, $v = \hat{v}/\bar{u}_0$, dimensionless velocity vector components; h, half-height of plane channel; $\bar{u}_0$, mean velocity at channel inlet; $\mathrm{Pe} = (\bar{u}_0 h)/D$, diffusion Peclet number; D, diffusion coefficient; $\Theta = C/C_0$, dimensionless concentration of solute; C_0, concentration of solute at channel inlet; Θ_ω, dimensionless concentration of solute on the membrane; Θ_g, the same,

at the point of gel formation; $V = \hat{V}/\overline{u}_0$, transmembrane velocity; f, magnitude of diffusion boundary layer; $\Delta = f/h$, the same, dimensionless; k, coefficient of hydraulic resistance of gel layer.

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