## ULTRAFILTRATION IN A PLANE CHANNEL WITH ONE PERMEABLE SURFACE

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A study is carried out to investigate unsteady-state concentration polarization in the case of laminar ultrafiltration in a plane channel with one permeable surface as a function of the selectivity properties of the membrane.

At present membrane devices of various modifications are more and more widely used for separation of liquid systems [1, 2]. Devices with plane membrane elements located on one side of the channel are one of the modifications. Such devices are manufactured, for example, by the Romicon Company (USA) and others.

The main factor that has a substantial negative effect on the characteristics of the ultrafiltration membrane process is concentration polarization, i.e., gel formation, which results in formation of a poorly permeable layer of high-molecular-weight compounds on the surface of the membrane. A search for optimum designs of membrane apparatuses seems a simplest and most efficient way to decrease the level of gel formation. For this purpose it is necessary to have a clear idea of the main relationships of the process that occurs in these devices.

In [1] numerical methods were used to obtain some versions of the development of concentration polarization in a channel with one permeable wall. However, these results are not informative, since they cannot be used for analysis of and a search for an optimum version of ultrafiltration by broad variation of parameters.

In the present work a semi-integral approach [3-5] was used to find an analytical solution of the problem of unsteady-state ultrafiltration in a channel with one permeable wall, which makes it possible to reveal the physical picture of the process and to find its main relationships.

We will obtain the velocity distribution in a flow of a high-molecular-weight solution of liquids in a thin plane channel with one permeable wall under conditions of gel formation on the surface of the membrane. As a result of escape of some of the liquid through the membrane, the concentration of high-molecular-weight compounds on its surface will increase until it reaches the critical value of gel formation, and the membrane becomes covered with a layer of gel. This layer is a hindrance to penetration of the liquid through the membrane (see Fig. 1).

The system of equations of motion and discontinuity that describes the process is represented in the form

$$-\frac{1}{\rho}P'_{x} + \nu u'_{yy} = 0, \qquad (1)$$

$$P'_{x} = 0$$
, (2)

$$\hat{u}_x + \hat{u}_y = 0 \tag{3}$$

with the boundary conditions

$$\hat{u}|_{y=f} = 0; \ \hat{v}|_{y=f} = -V_f; \ \hat{u}|_{y=2h} = 0; \ \hat{v}|_{y=2h} = 0.$$
(4)

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Fig. 1. Schematic liquid flow in a plane channel with one permeable surface with account for gel formation.

It follows from Eq. (2) that P = P(x). Integrating Eq. (1) with account for boundary conditions (4), we find an expression for the longitudinal component of the velocity vector in the channel:

$$\widehat{\mu} = -\frac{h^2}{\mu} P'_x \left( \frac{y - f}{h} - \frac{y^2 - fy}{2h^2} \right).$$
(5)

The transverse component is determined from discontinuity equation (3), using the expression for the component  $\hat{u}$  for this purpose:

$$\widehat{\nu} = -\frac{2}{3} \frac{h^3}{\mu} \frac{\partial}{\partial x} \left[ P'_x \left( 1 - \frac{3f}{2h} + \frac{y^2 (2y - 3f)}{8h^3} + \frac{3y (2f - y)}{4h^2} \right) \right].$$
(6)

Hence, the flow velocity through the membrane is equal to

$$V_f = \frac{2}{3} \frac{h^3}{\mu} \frac{\partial}{\partial x} \left[ P'_x \left( 1 - \frac{f}{2h} \right)^3 \right].$$
<sup>(7)</sup>

In Eqs. (5) and (6) the unknown quantity  $P'_{x}$  will be eliminated. To do this, the mean flow rate is introduced:

$$\overline{u}_{x} = \frac{1}{2h-f} \int_{f}^{2h} \widehat{u} dy.$$
(8)

Then, at the inlet to the channel at x = 0 it is

$$\overline{u}_0 = -\frac{1}{3} \frac{h^2}{\mu} \frac{\partial P}{\partial x} \bigg|_{x=0}.$$
(9)

We integrate (7) with account for relation (9) and substitute the result into formulas (5) and (6). For the components of the velocity vector we obtain

$$\hat{u} = \frac{3}{2h(1 - f/2h)^3} \left( 2h\overline{u}_0 - \int_0^x V_f \, dx \right) \left( \frac{y - f}{h} - \frac{y^2 - fy}{2h^2} \right),$$
(10)  
$$\hat{v} = \frac{-V_f + 3(1 - f/2h) \left( 2\overline{u}_0 h - \int_0^x V_f \, dx \right) \frac{\partial f}{\partial x}}{(1 - f/2h)^3} \times$$

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$$\times \left(1 + \frac{3f}{2h} + \frac{y^2(2y - 3f)}{8h^3} + \frac{3y(2f - y)}{4h^2}\right) + \frac{2\overline{u}_0 h - \int V_f dx}{(1 - f/2h)^3} \left(1 - \frac{y^2}{4h^2} + \frac{y}{h}\right) \frac{3}{2h} \frac{\partial f}{\partial x}.$$
 (11)

Now, we consider the diffusion problem. For convenience in subsequent manipulations, we work with dimensionless quantities. We consider the first stage of the process of ultrafiltration, in which no gel is formed on the surface of the membrane. Unsteady-state laminar ultrafiltration in a plane channel with one permeable surface is described by the equation of convective diffusion, which is represented in the form

$$\frac{\partial (\Theta - 1)}{\partial \tau} + \frac{\partial u (\Theta - 1)}{\partial \xi} + \frac{\partial v (\Theta - 1)}{\partial \eta} = \frac{1}{Pe} \frac{\partial^2 (\Theta - 1)}{\partial \eta^2}$$
(12)

with the boundary conditions

$$u|_{\eta=0} = 0; \quad v|_{\eta=0} = -V \quad (V = \text{const});$$

$$\varphi V \Theta_{\omega} + \frac{1}{\text{Pe}} \frac{\partial \Theta}{\partial \eta} = 0 \Big|_{\eta=0};$$

$$\Theta|_{\eta=2h} = 1; \quad \Theta|_{\xi=0} = 1; \quad \Theta|_{\tau=0} = 1.$$
(13)

It should be noted that in the search for a solution of unsteady-state diffusion problem (12) and (13), use will be made of the solution of steady-state dynamic problem (10) and (11) found above. This is admissible since Pe >> Re, i.e., the process is quasisteady (at a particular moment a steady-state velocity distribution corresponds to each instantaneous concentration distribution).

Equation (12) is integrated with boundary conditions (13) across the diffusion boundary layer. The requirements  $\Theta = 1$  and  $\partial \Theta / \partial \eta = 0$  at  $\eta = \Delta$  ordinarily used in boundary-layer theory are added to these conditions. As a result

$$\frac{\partial}{\partial \tau} \int_{0}^{\Delta} (\Theta - 1) \, d\eta + \frac{\partial}{\partial \xi} \int_{0}^{\Delta} u \, (\Theta - 1) \, d\eta = V \Gamma \,, \tag{14}$$

where  $\Gamma = 1 - (1 - \varphi)\Theta$ .

The unsteady-state concentration distribution on the membrane is specified proceeding from the solution of the steady-state problem. In the case of ultrafiltration the diffusion Peclet number is very high (Pe  $\approx 10^7$ ), and therefore, at any point of the channel the thickness of the diffusion boundary layer is substantially smaller than its half-height ( $h \gg \Delta$ ), i.e., it lies in the near-membrane region. Then, in the solution of the diffusion problem we can restrict ourselves to the first terms in  $\eta$  in the velocity distributions (10) and (11). Since in the present work pre-gel ultrafiltration is analyzed, in these distributions the terms f/h and  $\partial f/\partial x$  are 0. Thus, for the steady-state case the convective-diffusion equation has the form

$$3\left(1-\frac{1}{2}V\xi\right)\eta\frac{\partial\Theta}{\partial\xi}-V\frac{\partial\Theta}{\partial\eta}=\frac{1}{\operatorname{Pe}}\frac{\partial^{2}\Theta}{\partial\eta^{2}},\qquad(15)$$

and at  $\eta \rightarrow 0$ , i.e., near the membrane, we have

$$\frac{\partial}{\partial \eta} \left( V\Theta + \frac{1}{\text{Pe}} \frac{\partial \Theta}{\partial \eta} \right) \approx 0.$$
 (16)

Under the specified conditions its solution is

$$\Theta = \Theta_{\omega} \left( \xi \right) \left[ 1 - \varphi \left( 1 - \exp \left( - \operatorname{Pe} V \eta \right) \right) \right]. \tag{17}$$

On the basis of the concept of a boundary layer, the concentration distribution in the channel can be represented in the form

$$\Theta = \begin{cases} \Theta_{\omega} \left( \xi \right) \left[ 1 - \varphi \left( 1 - \exp \left( - \operatorname{Pe} V \eta \right) \right) \right], & 0 \le \eta \le \Delta, \\ 1, & \Delta \le \eta \le 2h. \end{cases}$$
(18)

Equating the two expressions for  $\Theta$  in (18) at  $\eta = \Delta$ , we find the unknown thickness of the diffusion boundary layer:

$$\Delta = \frac{1}{\operatorname{Pe}V}\ln\frac{\varphi\Theta_{\omega}}{\Gamma_{\omega}}\,,\tag{19}$$

where  $\Gamma_{\omega} = 1 - (1 - \varphi)\Theta_{\omega}$ .

To determine the unknown concentration on the surface of the membrane  $\Theta_{\omega}(\tau, \xi)$ , the expressions for u,  $\Theta$ , and  $\Delta$  from formulas (10), (17), and (19) are substituted into Eq. (14). After calculation of the integrals we have

$$\frac{1}{\operatorname{Pe}V}\frac{\partial}{\partial\tau}\left(\Theta_{\omega}-\Gamma_{\omega}\ln\frac{\varphi\Theta_{\omega}}{\Gamma_{\omega}}-1\right)+\frac{3}{\operatorname{Pe}^{2}V^{2}}\frac{\partial}{\partial\xi}\left(1-\frac{1}{2}V\xi\right)\times$$
$$\times\left(\Theta_{\omega}-\Gamma_{\omega}\ln\frac{\varphi\Theta_{\omega}}{\Gamma_{\omega}}-\frac{1}{2}\Gamma_{\omega}\left(\ln\frac{\varphi\Theta_{\omega}}{\Gamma_{\omega}}\right)^{2}-1\right)=V\Gamma_{\omega}.$$
(20)

From the physical point of view the unsteady-state process can be considered as two limiting conditions, namely, absolutely unsteady and steady.

We consider the case of steady concentration polarization. Then, it follows from (20) that

$$\frac{\partial}{\partial V\xi} \left( 1 - \frac{1}{2} V\xi \right) \left( \Theta_{\omega} - \Gamma_{\omega} \ln \frac{\varphi \Theta_{\omega}}{\Gamma_{\omega}} - \frac{1}{2} \Gamma_{\omega} \left( \ln \frac{\varphi \Theta_{\omega}}{\Gamma_{\omega}} \right)^2 - 1 \right) = \frac{\operatorname{Pe}^2 V^2 \Gamma_{\omega}}{3}.$$
(21)

It is impossible to integrate this equation in the general form. Therefore, very important particular cases will be analyzed.

Let  $V\xi \ll 1$  and  $\Theta_{\omega} \sim 1$ . These conditions are valid for short distances from the inlet to the channel. The quantity  $\Theta_{\omega}$  is represented in the form  $\Theta_{\omega} = 1 + \varepsilon$ , where  $\varepsilon \ll 1$ . Then  $\ln \varphi \Theta_{\omega} = \ln \varphi (1 + \varepsilon)$ ,  $\Gamma_{\omega} = \ln \varphi [1 - \varepsilon (1 - \varphi)/\varphi]$ . The logarithms are expanded into Taylor series in powers of  $\varepsilon$ , and restricting ourselves to cubic terms of the expansion, we find from formula (21)

$$\frac{d\varepsilon^2}{dV\xi} = 2\varphi^3 \mathrm{Pe}^2 V^2 \left[ 1 - \frac{\varepsilon \left(1 - \varphi\right)}{\varphi} \right].$$
(22)

Ultrafiltration membranes have high selectivity. With this fact in mind, it is possible to assume that  $\epsilon(1-\varphi)/\varphi << 1$ . Then, integrating Eq. (22) with the obvious condition  $\epsilon = 0$  at  $\xi = 0$ , we arrive at a formula for calculation of the concentration on the membrane:

$$\Theta_{\omega} = 1 + \varphi \left( 2Pe^2 V^2 V \xi \right)^{1/3}.$$
 (23)

In the case in which the terms in Eq. (21) containing logarithms of the concentration are small in comparison with the concentration, we have

$$\frac{d}{dV\xi}\left(\left(1-\frac{1}{2}V\xi\right)\left(\Theta_{\omega}-1\right)\right)=\frac{\mathrm{Pe}^{2}V^{2}}{3}\Gamma_{\omega}.$$
(24)

Integration with the boundary condition  $\Theta = 1$  at  $\xi = 0$  gives

$$\Theta_{\omega} = \frac{1 - \frac{3}{2\mathrm{Pe}^2 v^2} - \varphi \left(1 - \frac{1}{2} v\xi\right)^{\frac{2\mathrm{Pe}^2 v^2}{3}(1-\varphi) - 1}}{1 - \varphi - \frac{3}{2\mathrm{Pe}^2 v^2}}.$$
(25)

Analysis of (25) shows that in laminar ultrafiltration in a plane channel with one permeable surface, two limiting cases are possible. The quantity  $(1 - \varphi)$  can be small or large in comparison with the combination  $3/(2\text{Pe}^2 V^2)$ , which characterizes the relation between the convective and diffusion mass transfer.

Let

$$(1-\varphi) << \frac{3}{2\mathrm{Pe}^2 V^2}.$$
(26)

Since in the case of ultrafiltration  $PeV = 10 - 10^3$ , condition (26) implies almost ideal selectivity of the membrane ( $\varphi \approx 1$ ). In view of this fact, we obtain

$$\Theta_{\omega} = 1 + \frac{P e^2 V^2 V \xi}{3 \left(1 - \frac{1}{2} V \xi\right)}.$$
(27)

Thus, for the present case the concentration of the dissolved compound increases continuously on the membrane with distance from the inlet to the channel. For ideal selectivity of the membrane ( $\varphi = 1$ ) Eq. (21) can easily be integrated. Its solution is

$$\Theta_{\omega} - \ln \Theta_{\omega} - \frac{1}{2} \left( \ln \Theta_{\omega} \right)^2 = 1 + \frac{P e^2 V^2 V \xi}{3 \left( 1 - \frac{1}{2} V \xi \right)} .$$
(28)

A comparison of formulas (27) and (28) shows that at  $\Theta_{\omega} >> \ln \Theta_{\omega}$  (this condition was assumed in the derivation of formula (25)) expression (28) becomes (27).

We consider the case in which

$$(1-\varphi) \gg \frac{3}{2\mathrm{Pe}^2 V^2}.$$
(29)

Then, it follows from (25) that

$$\Theta_{\omega} = \frac{1}{1 - \varphi} \left( 1 - \varphi \left( 1 - \frac{1}{2} V \xi \right)^{\frac{2 \operatorname{Pe}^2 V^2}{3} (1 - \varphi)} \right),$$
(30)

i.e., the concentration of the dissolved compound increases rapidly on the membrane with the distance from the inlet to the channel and attains the limit  $\Theta_{\omega} = 1/(1 - \varphi)$ , which remains unchanged over the rest of the length of the channel.

Now, we turn to the study of absolutely unsteady ultrafiltration. We find from (20)

$$\frac{\partial}{\partial V\tau} \left( \Theta_{\omega\tau} - \Gamma_{\omega\tau} \ln \frac{\varphi \Theta_{\omega\tau}}{\Gamma_{\omega\tau}} - 1 \right) = \text{Pe}V\Gamma_{\omega\tau}.$$
(31)

It is impossible to obtain the general solution of this equation, and particular cases will be considered.

For short times of the unsteady-state process, it can be assumed that  $\Theta_{\omega\tau} \sim 1$ . Then, representing  $\Theta_{\omega\tau}$  in the form  $\Theta_{\omega\tau} = 1 + \varepsilon$ , expanding the logarithms into Taylor series in powers of  $\varepsilon$ , and restricting ourselves to the initial terms of the expansion, we arrive at the differential equation

$$\frac{d\varepsilon^2}{dV\tau} = 2\varphi^2 \operatorname{Pe} V \left( 1 - \frac{\varepsilon \left( 1 - \varphi \right)}{\varphi} \right) \,. \tag{32}$$

Assuming  $\varepsilon(1-\varphi)/\varphi \ll 1$  and integrating under the condition  $\varepsilon = 0$  at  $\tau = 0$ , we find

$$\Theta_{\omega\tau} = 1 + \varphi \sqrt{2 \text{Pe}VV\tau} . \tag{33}$$

At  $(1 - \varphi) \ll 3/(2Pe^2V^2)$  (this is equivalent to the condition  $1 - \Gamma_{\omega\tau} \ll 1$ ), we obtain from Eq. (31)

$$\Theta_{\omega\tau} - \ln \Theta_{\omega\tau} = 1 + \text{Pe}VV\tau . \tag{34}$$

For long times of the ultrafiltration process or substantial PeV, when  $\Theta_{\omega\tau} >> \ln \Theta_{\omega\tau}$ , we have

$$\Theta_{\omega\tau} = \mathrm{Pe}VV\tau \;. \tag{35}$$

When  $(1 - \varphi) >> 3/(2 \text{Pe}^2 V^2)$  and  $\Theta_{\omega\tau} >> \ln \Theta_{\omega\tau}$ , Eq. (31) has the form

$$\frac{\partial \Theta_{\omega\tau}}{\partial V\tau} = \operatorname{Pe} V \Gamma_{\omega} . \tag{36}$$

Integrating (36), when  $\Theta_{\omega\tau} = 1$  at  $\tau = 0$ , we find

$$\Theta_{\omega\tau} = \frac{1}{1-\varphi} \left[ 1 - \exp\left( -\left(1-\varphi\right) \operatorname{Pe} V V \tau \right) \right].$$
(37)

In conclusion, we determine the time  $\tau_s$  needed to attain steady-state operation. From the condition  $\Theta_{\omega} = \Theta_{\omega\tau}$  and relations (23) and (33)  $\tau_s$  at  $\Theta_{\omega} \sim 1$  follows:

$$\tau_{\rm s} = \left(\frac{1}{2} \operatorname{Pe} \xi^2\right)^{1/3}.$$
(38)

From formulas (28) and (34), (30) and (37) we obtain  $\tau_s$  for the cases  $(1 - \varphi) \ll 3/(2\text{Pe}^2 V^2)$  and  $(1 - \varphi) \gg 3/(2\text{Pe}^2 V^2)$ , respectively:

$$\tau_{\rm s} = \frac{{\rm Pe}V\xi}{3\left(1-\frac{1}{2}V\xi\right)},\tag{39}$$

$$\tau_{\rm s} = \frac{1}{3} \operatorname{Pe} V \xi \ . \tag{40}$$

Thus, the suggested theory is valid for the description of laminar ultrafiltration up to the stage of gel formation in a plane channel with one permeable surface.

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## 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 components of the velocity bector; h, half-height of the plane channel;  $\bar{u}_0$ , average velocity at the inlet to the channel;  $v, \mu$ , kinematic and dynamic viscosities;  $Pe = (\bar{u}_0 h)/D_0$ , diffusion Peclet number; D, diffusion coefficient;  $\Theta = C/C_0$ , dimensionless concentration of the dissolved compound;  $C_0$ , concentration of the dissolved compound at the inlet to the channel;  $\Theta_{\omega}$ , dimensionless concentration of the dissolved compound on the membrane;  $V = \hat{V}/\bar{u}_0$ , transmembrane velocity; f, thickness of the diffusion boundary layer;  $\Delta = f/h$ , dimensionless thickness of the diffusion boundary layer; P, pressure in the channel.

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