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ULTRAFILTRATION IN TUBULAR MEMBRANE ELEMENTS WITH ONE PERMEABLE SURFACE

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We consider the nonstationary laminar regime of ultrafiltration in tubular membrane filters with one permeable surface depending on the selective properties of the membrane.

Membrane technologies, including ultrafiltration, are of great importance in many branches of industry. It is these technologies, together with other conventional methods, that have been linked, first of all, to the prospects of solving a number of pressing environmental problems in recent years. These include sewage treatment, water treatment, and treatment of aqueous solutions. This list can be supplemented with problems arising in medicine and in the chemical, petrochemical, food, and other branches of industry, i.e., where it is essential to separate, purify, and concentrate solutions.

Modern industry produces a large number of ultrafiltration apparatuses of various designs [1, 2], among which the apparatuses with tubular membrane elements have come into wide use. Variants of these filters include tubular membrane elements with one permeable surface. The latter are two tubes placed into one another, one of which is a membrane and the other of which is a drainage skeleton with a microporous structure that simultaneously serves as a device for removal of a permeate and support for the membrane. Between the tubes there is a gap over which the liquid to be separated moves. Depending on the location of the membrane, we distinguish three types of similar filters: filters with a membrane inside the skeleton, filters with a membrane outside the skeleton, and a combined variant. The most widely used are the apparatuses with filters of the first type.

Based on the semiintegral method of [3, 4], in the present work we suggest the physical pattern of processes that occur in the above devices, more specifically, its hydrodynamic and diffusion parts. This problem is of great practical importance. Its solution makes it possible to identify many problems that arise in developing and creating new, more perfect ultrafiltration apparatuses, to determine methods of their elimination, and to improve substantially the search for the most optimum operating conditions of the existing devices. The urgency of the latter problem is due to the variety of separable solutions, for each of which one must select filtration parameters of its own.

Let us consider the nonstationary regime of ultrafiltration in an annular channel formed by two coaxial circular cylinders of radii R_1 and R_2 ($R_1 < R_2$). We will assume that the flow of a liquid in the channel between the cylinders is steady laminar, the internal cylinder is permeable, and the gel layer, formed on the cylinder due to the phenomenon of concentration polarization, is stationary (see Fig. 1). The concentration polarization, i.e., gel formation, adversely affects the performance of the membranes: this is accompanied by a drop in the specific productivity and the capacity for regeneration, and the efficiency of separation as a whole decreases. In order to evaluate the degree of influence of this phenomenon, it is necessary to find the concentration distribution of the solutes on the membrane surface. To solve the posed problem, first we must obtain the velocity distribution in the channel that takes into account a partial nonuniform outflow of the

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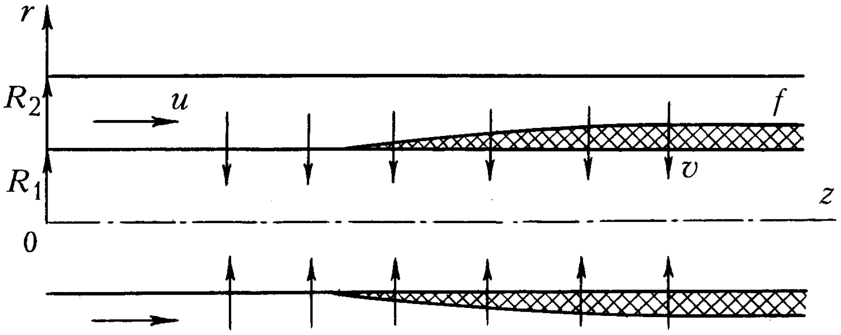

Fig. 1. Schematic representation of separation of liquids in tubular membrane elements with one permeable surface.

liquid through the permeable surface. This outflow decreases along the channel length, since, due to the formation of the gel layer, the permeability of the membrane drops. The legitimacy of this approach is associated with the quasistationary nature of the ultrafiltration process, i.e., the flow field of the solution is virtually independent of the concentration field (the Reynolds and Péclet numbers that determine these fields differ by 3–4 orders of magnitude (Re << Pe)); this means that the latter problem can be considered separately from the basic one. The system of equations of motion and continuity describing the flow will have the form

$$\hat{u}''_{rr} + \frac{1}{r}\hat{u}'_r = \frac{1}{\mu}P'_z\,, \tag{1}$$

$$P'_r = 0\,, \tag{2}$$

$$\hat{v}'_r + \frac{1}{r}\hat{v} = 0 \tag{3}$$

with the following boundary conditions:

$$\hat{u}|_{r=R_2} = 0\,;\quad \hat{v}|_{r=R_2} = 0\,;\quad \hat{u}|_{r=R_1+f} = 0\,;\quad \hat{v}|_{r=R_1+f} = V_f\,. \tag{4}$$

It is obvious from Eq. (2) that $P = P(z)$. Applying the first and third boundary conditions of (4), we integrate Eq. (1):

$$\hat{u} = \frac{P'_z}{4\mu}\left(r^2 - R_2^2 + \frac{R_2^2 - (R_1+f)^2}{\ln((R_1+f)/R_2)}\ln\frac{r}{R_2}\right). \tag{5}$$

However, because of the complexity of expression (5), it cannot be used in this form for further calculations (for solution of a diffusion problem). Therefore, we take the following assumptions that occur in actual ultrafiltration apparatuses:

$$h/R << 1\,;\quad f/h << 1\,, \tag{6}$$

where $h = R_2 - R_1$ and $R = R_1$. It should be noted that the second condition (6) was already kept in mind in posing the problem, i.e., in writing the system of equations (1)–(4), we assumed that the gel height is negligibly small and it does not change the cross section of the channel. Introducing a new variable $y = r - R$, we expand the logarithms into a series in powers *h/R*, *f/R*, and *f/h*, respectively, and restrict ourselves to the first two expansion terms. Then, having applied the expansion $\left(1-\frac{h+f}{2R}\right)^{-1} = 1+\frac{h+f}{2R}$, we obtain

$$\hat{u}=\frac{h^2P_z'}{2\mu}\left(\left(\frac{y^2}{h^2}-1\right)A-\left(1+\frac{f}{h}\right)\left(\frac{y}{h}+1\right)B\right),$$

where

$$A=1+\frac{h+f}{2R}+\left(\frac{h+f}{2R}\right)^2\ ;\ \ B=1+\frac{h+f}{2R}\ .$$

From this, with allowance for the assumptions (6) taken for the longitudinal component of the vector of the liquid velocity in the channel, we have

$$\hat{u}=-\frac{h^2P_z'}{2\mu}\frac{y}{h}\left(1-\frac{y}{h}\right). \tag{7}$$

Now we substitute Eq. (7) into the continuity equation (3). Using Eq. (6) and the expansion $1/(R+y) = (1-y/R)/R$, with the second and fourth boundary conditions of (4), we obtain expressions for the radial component of the profile and for the transmembrane velocity, respectively:

$$\hat{v}=-\frac{h^3P_{zz}''}{12\mu}\left(1+2\frac{y^3}{h^3}-3\frac{y^2}{h^2}\right), \tag{8}$$

$$\hat{V}_f=\frac{h^3P_{zz}''}{12\mu}\,. \tag{9}$$

From Eqs. (7) and (8) we eliminate the unknown pressure gradient P_z'. For this, we integrate Eq. (5) over the channel cross section and calculate the mean velocity at the entrance to the channel (z = 0 and f = 0):

$$\bar{u}_0=-\frac{P_z'|_{z=0}}{8\mu}\left(R_2^2+R_1^2-\frac{R_2^2-R_1^2}{\ln\,(R_2/R_1)}\right).$$

Resorting to expansions similar to the above ones, we find

$$\bar{u}_0=-\frac{h^2P_{z_0}'}{16\mu}\,. \tag{10}$$

Then, integrating Eq. (9) over the channel length and using Eq. (10), we express P_z' in terms of $\bar{u}_0$. Substitution of the result obtained into the initial formulas gives

$$\hat{u}=\frac{8}{h}\left(\bar{u}_0h-\frac{3}{4}\int_0^z V_f\,dz\right)\left(\frac{y}{h}-\frac{y^2}{h^2}\right), \tag{11}$$

$$\hat{v} = V_f\left(1 + 2\frac{y^3}{h^3} - 3\frac{y^2}{h^2}\right). \tag{12}$$

Thus, based on comments (6), it can be considered that with the gel formation taken into account, formulas (11) and (12) determine exactly the flow field of a separable liquid in tubular filters with one permeable surface. As has been noted above, because of the quasistationary nature of the ultrafiltration process the solutions obtained are applicable to the description of the phenomenon of concentration polarization. To do this, we use an equation of convective diffusion that characterizes the process of accumulation of a substance at a certain point of space depending on time. In order to simplify further calculations, we pass to dimensionless quantities. Then the formulation of the problem will appear as follows:

$$\frac{\partial(\Theta - 1)}{\partial\tau} + \frac{\partial u(\Theta - 1)}{\partial\xi} - \frac{\partial v(\Theta - 1)}{\partial\eta} = \frac{1}{\mathrm{Pe}}\frac{\partial^2(\Theta - 1)}{\partial\eta^2}. \tag{13}$$

Now we denote the boundary conditions. Let ξ_1 be the point beginning with which the formation of the gel layer on the membrane surface occurs. The conditions before the given point and after it will be different:

for $\xi \le \xi_1$

$$u|_{\eta=0} = 0\,;\quad v|_{\eta=0} = V\ \ (V = \mathrm{const})\,;\quad \left.\varphi V\Theta_\omega + \frac{1}{\mathrm{Pe}}\frac{\partial\Theta}{\partial\eta}\right|_{\eta=0} = 0\,;$$
$$\Theta|_{\eta=1} = 1\,;\quad \Theta|_{\xi=0} = 1\,;\quad \Theta|_{\tau=0} = 1\,; \tag{14}$$

for $\xi \ge \xi_1$

$$u|_{\eta=\delta} = 0\,;\quad v|_{\eta=\delta} = V_\delta\,;\quad \left.\varphi V_\delta\Theta_g + \frac{1}{\mathrm{Pe}}\frac{\partial\Theta}{\partial\eta}\right|_{\eta=\delta} = \Theta_g\frac{\partial\delta}{\partial\tau}\,;\quad \Theta|_{\eta=1} = 1\,;$$
$$\Theta|_{\eta=\delta} = \Theta_g\,;\quad \delta(\xi, \tau)|_{\xi=\xi_1} = 0\,. \tag{15}$$

For the completeness of the concentration-field pattern, it is necessary that the requirements following from the physical concepts of the boundary-layer structure be added to Eqs. (14) and (15), i.e., it can be assumed that the concentration grows only within the limits of the boundary layer. Then $\Theta|_{\eta=\Delta} = 1$ and $\frac{\partial\Theta}{\partial\eta}\Big|_{\eta=\Delta} = 0$. Taking this fact into account, we integrate Eq. (13) across the diffusion boundary layer with boundary conditions (14) and (15). As a result, we have, respectively

$$\frac{\partial}{\partial\tau}\int_0^\Delta(\Theta - 1)\,d\eta + \frac{\partial}{\partial\xi}\int_0^\Delta u(\Theta - 1)\,d\eta = V\Gamma\,,\quad \xi \le \xi_1\,, \tag{16}$$

$$\frac{\partial}{\partial\tau}\int_\delta^\Delta(\Theta - 1)\,d\eta + (2\Theta_g - 1)\frac{\partial\delta}{\partial\tau} + \frac{\partial}{\partial\xi}\int_\delta^\Delta u(\Theta - 1)\,d\eta = V_\delta\Gamma_g\,,\quad \xi \ge \xi_1\,, \tag{17}$$

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

Equation (16) characterizes the pre-gel regime of ultrafiltration, while Eq. (17) characterizes the stage of gel formation. It is obvious that when $\xi = \xi_1$, the equality $\Theta = \Theta_g$ will be satisfied. We use a semiintegral approach according to which the law of distribution of the concentration of a substance in the boundary layer can be determined immediately from Eqs. (16) and (17). Proceeding from the solution of the stationary problem, we assign the nonstationary distribution of the concentration. Since the thickness of the diffusion boundary layer and the thickness of the gel layer are many times smaller than the channel height, in the formulas of velocity distribution (11) and (12) we can restrict ourselves only to the first terms in η. Then for the pre-gel regime it follows that:

$$8\left(1-\frac{3}{4}V\xi\right)\eta\frac{\partial\Theta}{\partial\xi}-V\frac{\partial\Theta}{\partial\eta}=\frac{1}{\text{Pe}}\frac{\partial^2\Theta}{\partial\eta^2}\,,\qquad \varphi V\Theta_\omega+\frac{1}{\text{Pe}}\frac{\partial\Theta}{\partial\eta}\bigg|_{\eta=0}=0\,, \tag{18}$$

and for the gel regime, that

$$8\left(1-\frac{3}{4}V\xi_1-\frac{3}{4}\int_{\xi_1}^{\xi}V_\delta d\xi\right)(\eta-\delta)\frac{\partial\Theta}{\partial\xi}-V_\delta\frac{\partial\Theta}{\partial\eta}=\frac{1}{\text{Pe}}\frac{\partial^2\Theta}{\partial\eta^2}\,,\qquad \varphi V_\delta\Theta_g+\frac{1}{\text{Pe}}\frac{\partial\Theta}{\partial\eta}\bigg|_{\eta=\delta}=0\,. \tag{19}$$

The first terms of Eqs. (18) and (19) in the immediate vicinity of the membrane surface will be small compared to the other terms. Therefore, they can be neglected. Integrating the remaining expressions with the corresponding boundary conditions twice, we obtain the stationary distribution of the concentration near the membrane:

$$\begin{aligned}\Theta&=\Theta_\omega(\xi)\,[1-\varphi\,(1-\exp(-\,\text{Pe}\,V\eta))]\,,\quad \xi\le\xi_1\,;\\ \Theta&=\Theta_g\,[1-\varphi\,(1-\exp(-\,\text{Pe}\,V_\delta\,(\eta-\delta)))]\,,\quad \xi\ge\xi_1\,.\end{aligned} \tag{20}$$

Consequently, with account for the statement, noted in formulating the problem, that the concentration varies only within the limits of the diffusion layer, the above distribution over the entire channel height will be written in general form as follows:

$$\Theta=\begin{cases}\Theta_\omega(\xi)\,[1-\varphi\,(1-\exp(1-\text{Pe}\,V\eta))]\,, & 0\le\eta\le\Delta\,,\\ 1\,, & \Delta\le\eta\le1\end{cases} \tag{21}$$

in the first stage of the process (before the gel formation) and

$$\Theta=\begin{cases}\Theta_g\,, & 0\le\eta\le\delta\,,\\ \Theta_g\,[1-\varphi\,(1-\exp(-\,\text{Pe}\,V_\delta\,(\eta-\delta)))]\,, & \delta\le\eta\le\Delta\,,\\ 1\,, & \Delta\le\eta\le1\end{cases} \tag{22}$$

in the stage of gel formation. From this, for the sizes of the boundary and gel layers it follows that

$$\Delta=\frac{1}{\text{Pe}V}\ln\frac{\varphi\Theta_\omega}{\Gamma_\omega}\,,\quad \xi\le\xi_1\,; \tag{23}$$

$$\Delta-\delta=\frac{1}{\text{Pe}V_\delta}\ln\frac{\varphi\Theta_g}{\Gamma_g}\,,\quad \xi\ge\xi_1\,. \tag{24}$$

Here $\Gamma_\omega = 1 - (1 - \varphi)\Theta_\omega$.

We relate the drop in the permeability to the gel-layer thickness:

$$V_\delta = V/(1 + k\delta)\,. \tag{25}$$

Thus, all the unknowns needed for finding the concentration are determined. We analyze each of Eqs. (16) and (17) individually.

We substitute into Eq. (16) the velocity distribution from formulas (11) and (12) and the concentration distribution from Eq. (21). With Eq. (23) taken into account, upon calculation of the integrals, we obtain

$$\frac{1}{\mathrm{Pe}V}\frac{\partial}{\partial\tau}\left(\Theta_\omega - \Gamma_\omega \ln\frac{\varphi\Theta_\omega}{\Gamma_\omega} - 1\right) + \frac{8}{\mathrm{Pe}^2V^2}\frac{\partial}{\partial\xi}\left(1 - \frac{3}{4}V\xi\right)\left(\Theta_\omega - \Gamma_\omega \ln\frac{\varphi\Theta_\omega}{\Gamma_\omega} - \right.$$

$$\left. - \frac{1}{2}\Gamma_\omega\left(\left(\ln\frac{\varphi\Theta_\omega}{\Gamma_\omega}\right)^2 - 1\right)\right) = V\Gamma_\omega\,, \quad \xi \le \xi_1\,. \tag{26}$$

The nonstationary ultrafiltration process will be considered as a combination of two limiting cases: the stationary regime and the especially nonstationary regime.

For the stationary regime without gel formation, from Eq. (26) we have

$$\frac{\partial}{\partial V\xi}\left(1 - \frac{3}{4}V\xi\right)\left(\Theta_\omega - \Gamma_\omega \ln\frac{\varphi\Theta_\omega}{\Gamma_\omega} - \frac{1}{2}\Gamma_\omega\left(\left(\ln\frac{\varphi\Theta_\omega}{\Gamma_\omega}\right)^2 - 1\right)\right) = \frac{\mathrm{Pe}^2V^2\Gamma_\omega}{8}\,. \tag{27}$$

It is impossible to find a general solution of this equation; therefore, we restrict ourselves to some of the most important particular cases.

1. For small distances from the entrance to the channel the following conditions can be satisfied: $V\xi \ll 1$ and $\Theta_\omega \sim 1$. Then Θ_ω can be represented in the form $\Theta_\omega = 1 + \varepsilon$, where $\varepsilon < 1$. Whence $\ln \varphi\Theta_\omega = \ln \varphi(1 + \varepsilon)$ and $\Gamma_\omega = \varphi(1 - \varepsilon(1 - \varphi)/\varphi)$. Resorting to expansions of the logarithms into a series in powers ε and restricting ourselves to the cubic expansion terms, from Eq. (27) we obtain

$$\frac{d\varepsilon^3}{dV\xi} = \frac{3}{4}\varphi^3\mathrm{Pe}^2V^2\,(1 - \varepsilon\,(1 - \varphi)/\varphi)\,.$$

If the selectivity of the membranes is rather high (otherwise, there is little sense in performing the process), then the term $\varepsilon(1 - \varphi)/\varphi \ll 1$. Neglecting this term, we integrate the latter expression provided that $\varepsilon|_{\xi=0} = 0$. Then adding unity to the result, we find an expression for the concentration on the membrane surface:

$$\Theta_\omega = 1 + \varphi\,(3\mathrm{Pe}^2V^2V\xi/4)^{1/3}\,. \tag{28}$$

2. Suppose that the terms containing the logarithm of concentration are small compared to the concentration (this is possible for large distances from the entrance to the channel). Then with the boundary condition $\Theta|_{\xi=0} = 1$ Eq. (27) can easily be integrated. Finally, we obtain

$$\Theta_\omega = \frac{1 - 6/\mathrm{Pe}^2V^2 - \varphi\,(1 - 3V\xi/4)^{-1+(1-\varphi)\mathrm{Pe}^2V^2/6}}{1 - \varphi - 6/\mathrm{Pe}^2V^2}\,. \tag{29}$$

We should note here two limiting cases for which formula (29) is considerably simplified:

$$2a)\quad 1-\varphi \ll \frac{6}{\mathrm{Pe}^2V^2}\,,\quad \Theta_\omega = 1+\frac{\mathrm{Pe}^2V^2V\xi}{8\,(1-3V\xi/4)}\,, \tag{30}$$

$$2b)\quad 1-\varphi \gg \frac{6}{\mathrm{Pe}^2V^2}\,,\quad \Theta_\omega = \frac{1}{1-\varphi}\,(1-\varphi\,(1-3V\xi/4)^{(1-\varphi)\mathrm{Pe}^2V^2/6})\,. \tag{31}$$

Undoubtedly, in both variants the concentration on the membrane surface grows continuously with increase in the distance from the entrance to the channel, and in case 2b) it tends to the obvious maximum $\Theta_\omega = 1/(1-\varphi)$.

3. For a membrane with ideal selectivity ($\varphi = 1$) there also exists the exact solution of Eq. (27)

$$\Theta_\omega - \ln\Theta_\omega - \frac{1}{2}\,(\ln\Theta_\omega)^2 = 1+\frac{\mathrm{Pe}^2V^2V\xi}{8\,(1-3V\xi/4)}\,. \tag{32}$$

It is easy to note that by assuming $\Theta_\omega \gg \ln\Theta_\omega$ in Eq. (32), we arrive at variant 2a). Consequently, when condition 2a) is satisfied the membrane selectivity is close to unity (nearly ideal selectivity ($\varphi \cong 1$)).

We analyze the especially nonstationary regime of ultrafiltration without gel formation. Then Eq. (26) will have the form

$$\frac{1}{\mathrm{Pe}V}\frac{\partial}{\partial\tau}\left(\Theta_{\omega\tau}-\Gamma_{\omega\tau}\ln\frac{\varphi\Theta_{\omega\tau}}{\Gamma_{\omega\tau}}-1\right)=V\Gamma_{\omega\tau}\,. \tag{33}$$

It is impossible to obtain a general solution of this equation; therefore, we proceed similarly to the stationary case.

1. $\Theta_{\omega\tau} \sim 1$ (this holds for small times of the process when the concentration of the solution to be separated has no time to change substantially). Then $\Theta_{\omega\tau} = 1+\varepsilon$ and $\varepsilon \ll 1$. Expanding the logarithms into a series in powers ε and restricting ourselves to the first expansion terms from Eq. (33), we find

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

Taking into account that $\varepsilon(1-\varphi)/ \ll 1$, we integrate the last expression provided that $\varepsilon|_{\tau=0} = 0$. Finally, for the concentration on the membrane surface we have

$$\Theta_{\omega\tau} = 1+\varphi\sqrt{2\mathrm{Pe}VV\tau}\,. \tag{34}$$

2. For $(1-\varphi)\Theta_{\omega\tau} \ll 1$ (as follows from the analysis of the stationary regime, this is equivalent to the condition of nearly ideal selectivity $1-\varphi \ll 6/\mathrm{Pe}^2V^2$), Eq. (33) has the solution

$$\Theta_{\omega\tau} - \ln\Theta_{\omega\tau} = 1+\mathrm{Pe}V\tau\,. \tag{35}$$

The same result for ideal selectivity is obtained from Eq. (33).

3. For $\Theta_{\omega\tau} \gg \ln\Theta_{\omega\tau}$ and $(1-\varphi)\Theta_{\omega\tau} \gg 1$ (this is equivalent to $1-\varphi \gg 6/\mathrm{Pe}^2V^2$), the concentration is equal to

$$\Theta_{\omega\tau} = (1-\varphi\exp(-(1-\varphi)\,\mathrm{Pe}VV\tau))/(1-\varphi)\,. \tag{36}$$

It is obvious that formula (36) holds for large times or large values of PeV.

Thus, generalizing the above data at this stage of the process, we can say that the dependence of the concentration on the membrane surface on the distance and time is of a complex nature not allowing one to find exact general analytical solutions. However, the most important particular cases that have been considered and implemented in practice are quite sufficient for us to have an idea of the entire pattern of separation at this stage. The analysis of these cases shows that the resistance of the membrane (here it is associated with V) and its selective properties exert the main influence on the process. For high values of the latter parameter (close to 1), its influence can be ignored. Comparing the formulas for the stationary and the especially nonstationary regimes (the condition $\Theta_\omega = \Theta_{\omega\tau}$) that correspond to the identical conditions for performing the ultrafiltration process, it is also possible to determine the time needed to attain the stationary operating regime

$$\tau_s = \frac{1}{2}\left(\frac{3}{4}\,\mathrm{Pe}\,\xi^2\right)^{1/3}, \quad \Theta_{\omega\tau} \sim 1\,;$$

$$\tau_s = \frac{1}{8}\,\mathrm{Pe}V\xi\,, \quad 1-\varphi >> \frac{6}{\mathrm{Pe}^2V^2}\,; \quad \tau_s = \frac{\mathrm{Pe}V\xi}{8\,(1-3V\xi/4)}\,, \quad 1-\varphi << \frac{6}{\mathrm{Pe}^2V^2}\,. \tag{37}$$

Now we return to Eq. (17), which characterizes the second stage of the ultrafiltration process, i.e., gel formation. First we analyze the stationary part. To do this, we substitute the expressions for u from Eq. (11) and for Θ from Eq. (22) into Eq. (17), and then with account for Eq. (24) integrate with respect to ξ. As a result, we obtain

$$8\left(1-\frac{3}{4}V\xi_1-\frac{3}{4}\int_{\xi_1}^{\xi} V_\delta\, d\xi\right)\frac{\Sigma_g}{\mathrm{Pe}^2V_\delta^2} = \int_0^{\xi_1}\Gamma_\omega V d\xi + \int_{\xi_1}^{\xi}\Gamma_g V_\delta d\xi\,. \tag{38}$$

To contract the representation, we denote here

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

When $\xi = \xi_1$, the position of the point of onset of gel formation on the membrane is obtained:

$$\int_0^{\xi_1}\Gamma_\omega V d\xi = \frac{8\Sigma_g\,(1-3V\xi_1/4)}{\mathrm{Pe}^2V^2}\,. \tag{39}$$

We are unable to calculate this integral in general form. However, as follows from the examples considered above, the dependence of the solute concentration near the membrane surface on ξ has a nonlinear nature; therefore, it can be evaluated from solutions (39) 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, upon substitution of these values into Eq. (39) and integration, we obtain the bounds of the region of determination of the given point:

$$\frac{12\Sigma_g}{(\varphi+\Gamma_g)\,\mathrm{Pe}^2V^2} \le \frac{V\xi_1}{4/3 - V\xi_1} \le \frac{6\Sigma_g}{\Gamma_g\mathrm{Pe}^2V^2}\,. \tag{40}$$

For the ideal selectivity the position of the point of onset of gel formation is determined from the formula

$$V\xi_1 = \left(\frac{3}{4} + \frac{\mathrm{Pe}^2 V^2}{8\Sigma_{\mathrm{g}}}\right)^{-1} .$$

Taking condition (39) into account, we solve Eq. (38) for the integral

$$\int\limits_{\xi_1}^{\xi} V_\delta d\xi = \frac{8\,(1 - 3V\xi_1/4)\,\Sigma_{\mathrm{g}}\,(V^2 - V_\delta^2)}{V^2\,(6\Sigma_{\mathrm{g}} + \Gamma_{\mathrm{g}}\mathrm{Pe}^2 V_\delta^2)} . \tag{41}$$

We differentiate expression (41) with respect to ξ and then integrate it applying the boundary condition $V_\delta|_{\xi=\xi_1} = V$:

$$\frac{4/3 - V\xi}{4/3 - V\xi_1} = \frac{V_\delta}{V}\left(\frac{1+F}{F + (V_\delta/V)^2}\right) - \frac{1+F}{\sqrt{F}}\left(\arctan\frac{1}{\sqrt{F}} - \arctan\frac{V_\delta}{V\sqrt{F}}\right), \tag{42}$$

where $F = 6\Sigma_{\mathrm{g}}/\Gamma_{\mathrm{g}}\mathrm{Pe}^2 V^2$.

For the ideal selectivity ($\varphi = 1$) formula (42) takes the form

$$V\xi = \frac{4}{3}\left(1 - \frac{V_\delta}{V}\left(\frac{V\xi_1}{4/3 - V\xi_1} + \frac{V_\delta^2}{V^2}\right)^{-1} + \sqrt{\frac{4/3 - V\xi_1}{V\xi_1}}\ \arctan\sqrt{\frac{4/3 - V\xi_1}{V\xi_1}}\ \frac{1 - \dfrac{V_\delta}{V}}{1 + \dfrac{V_\delta}{V}\,\dfrac{4/3 - V\xi_1}{V\xi_1}}\right).$$

For the values of $V\xi_1$, which are close to zero, solution of Eq. (41) is considerably simplified

$$\frac{V_\delta}{V} = \left(1 + (V\xi - V\xi_1)\,\frac{\Gamma_{\mathrm{g}}\mathrm{Pe}^2 V^2}{4\Sigma_{\mathrm{g}}}\right)^{-1/3} , \tag{43}$$

and for $\varphi = 1$ the solution is of the form

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

In the case where, in addition, the condition $V\xi/V\xi_1 >> 1$ is satisfied, from the latter expression it follows that

$$V_\delta = \left(\frac{4\Sigma_{\mathrm{g}}}{\Gamma_{\mathrm{g}}\mathrm{Pe}^2\xi}\right)^{1/3} . \tag{44}$$

When $\varphi = 1$, formula (44) will be written for clarity in open form:

$$V_\delta = \left(\frac{4}{\mathrm{Pe}^2\xi}\left(\Theta_{\mathrm{g}} - \ln\Theta_{\mathrm{g}} - \frac{1}{2}\ln^2\Theta_{\mathrm{g}} - 1\right)\right)^{1/3} .$$

We comment briefly on the stage conducted. From the general solution (42) it is obvious that in addition to the resistance of the membrane and its selectivity, the resistance of the gel layer also exerts an influence on the process. This resistance increases gradually and becomes predominant (Eq. (43)), and provided that $V\xi/V\xi_1 >> 1$ it increases so much that the resistance of the membrane can be simply neglected (Eq. (44)). In this connection, the regime of ultrafiltration with gel formation can be conventionally subdivided into two intervals: 1) from the point ξ_1 to a certain point $\xi > \xi_1$ where the resistances of the membrane and of the gel layer are quantities of the same order of magnitude; 2) further, i.e., beyond the given interval and to the end of the channel, the latter quantity is of decisive importance during the process.

Now we consider the especially nonstationary regime of ultrafiltration with gel formation. Suppose that τ_1 is the time of onset of gel formation on the membrane surface. Then after differentiation of Eq. (25), from Eq. (17) we obtain

$$\frac{\partial}{\partial\tau}\int_{\delta}^{\Delta}(\Theta - 1)\,d\eta - \frac{V}{kV_{\delta}^{2}}(2\Theta_{\mathrm{g}} - 1)\frac{\partial V_{\delta}}{\partial\tau} = \Gamma_{\mathrm{g}}V_{\delta}\,, \quad \tau \ge \tau_1\,. \tag{45}$$

Substituting Eqs. (22) and (24) into expression (45), we calculate the integral on the left-hand side of expression (45). As a result, upon introducing the notation $\Sigma_\tau = \Theta_{\mathrm{g}} - 1 - \Gamma_{\mathrm{g}} \ln\ (\varphi\Theta_{\mathrm{g}}/\Gamma_{\mathrm{g}})$, we arrive at the ordinary differential equation

$$-\left(\frac{\Sigma_\tau}{\mathrm{Pe}} + \frac{V(2\Theta_{\mathrm{g}} - 1)}{k}\right)\frac{1}{V_{\delta}^{2}}\frac{dV_{\delta}}{d\tau} = \Gamma_{\mathrm{g}}V_{\delta}\,. \tag{46}$$

whose solution is as follows:

$$\frac{V_{\mathrm{g}}}{V} = \left(1 + \frac{2\,(V\tau - V\tau_1)}{\Sigma_\tau/\mathrm{Pe}\,V\Gamma_{\mathrm{g}} + (2\Theta_{\mathrm{g}} - 1)/k\Gamma_{\mathrm{g}}}\right)^{-1/2}\,. \tag{47}$$

The time of onset of gel formation will be evaluated in a way similar to the stationary case. Finally, we obtain

$$\frac{2\Sigma_\tau}{\mathrm{Pe}V\,(\varphi + \Gamma_{\mathrm{g}})} \le V\tau_1 \le \frac{\Sigma_\tau}{\mathrm{Pe}V\Gamma_{\mathrm{g}}}\,. \tag{48}$$

Whence it is obvious that the time of onset of gel formation for the ideal selectivity of the membrane is calculated from the formula

$$V\tau_1 = \frac{\Theta_{\mathrm{g}} - \ln\Theta_{\mathrm{g}} - 1}{\mathrm{Pe}V}\,. \tag{49}$$

By analogy with the pre-gel stage of the process considered above, by equating the corresponding formulas of the stationary and especially nonstationary regimes, it is possible to determine the time when it attains the stationary regime. Then, assuming that $V\xi_1$ is small, from Eqs. (43) and (47) we have

$$V\tau_{\mathrm{s}} = V\tau_1 + \frac{1}{2}\left(\left(\frac{(V\xi - V\xi_1)\,\Gamma_{\mathrm{g}}\mathrm{Pe}^2V^2}{4\Sigma_{\mathrm{g}}} + 1\right)^{2/3} - 1\right)\left(\frac{2\Theta_{\mathrm{g}} - 1}{\Gamma_{\mathrm{g}}k} + \frac{\Sigma_\tau}{\Gamma_{\mathrm{g}}\mathrm{Pe}V}\right). \tag{50}$$

In the case where $V\xi/V\xi_1 >> 1$, the expression has the form

$$V\tau_s = \frac{1}{2}\left(\frac{(V\xi - V\xi_1)\,\Gamma_g \mathrm{Pe}^2 V^2}{4\Sigma_g}\right)^{2/3}\left(\frac{2\Theta_g - 1}{\Gamma_g k} + \frac{\Sigma_\tau}{\Gamma_g \mathrm{Pe} V}\right).$$

For $\varphi = 1$, the latter expression takes the form

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

Thus, the analytical solutions obtained in this work make it possible to form the general pattern of nonstationary concentration polarization – gel formation – with laminar ultrafiltration in tubular filters with one permeable surface. We briefly mention once again the basic laws of the given process. Thus, at the first stage, before the onset of gel formation, the main factor affecting its course is the membrane itself (more exactly, its resistance). At the second stage, i.e., after the point of gel formation, the appearing gel layer introduces fundamental changes into the process: at a certain initial channel length the influence of this layer is comparable with the influence of the membrane, and subsequently it becomes determining. The selective properties of the membrane are mainly of great importance at the first stage. Here the change in the selectivity that at first glance is relatively small may reflect substantially on the entire pattern of separation.

NOTATION

$\xi = x/h$ and $\eta = y/h$, dimensionless longitudinal and transverse coordinates; $u = \hat{u}/\bar{u}_0$ and $v = v/\bar{u}_0$, dimensionless components of the velocity vector; $\bar{u}_0$, mean velocity at the entrance to the channel; $R = R_1$, R_2, and $h = R_2 - R_1$, inner and outer radii and characteristic dimension of the channel; t and $\tau = t\bar{u}_0/h$, time and dimensionless time; τ_s, dimensionless time of attainment of the stationary regime; μ, coefficient of kinematic viscosity; $\mathrm{Pe} = (\bar{u}_0 h)/D$, diffusional Péclet number; D, diffusion coefficient; C, concentration of the solute; $\Theta = C/C_0$, the same, dimensionless; C_0, concentration of the solute at the entrance to the channel; Θ_ω, dimensionless concentration of the solute on the membrane; $\Theta_{\omega\tau}$, the same, in the especially nonstationary regime; Θ_g, nondimensional concentration of gel formation; $V = \hat{V}/\bar{u}_0$ and $V_g = \hat{V}_f/\bar{u}_0$, dimensionless transmembrane velocity; k, dimensionless coefficient of hydraulic resistance of the gel layer; f, size of the gel layer; $\delta = f/h$, the same, dimensionless; Δ, dimensionless size of the diffuse boundary layer.

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