# ULTRAFILTRATION IN A PIPE FILTER WITH GELATION

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*We investigate the process of unsteady-state ultrafiltration with gelation under laminar flow conditions in a pipe filter with nonideal selectivity of its membrane.*

As a promising method for cleaning, separation, and concentration of dissolved or disperse particles, ultrafiltration has found wide application in the food, pharmaceutical, textile, metal-working, and electronic industries and in biology and medicine [1]. Hollow-fiber and pipe membrane apparatuses have come into widespread use.

The process of ultrafiltration is accompanied by the phenomenon of concentration polarization, which occurs in a pre-gel regime in hollow-fiber filters and in a gel regime in pipe filters [2].

To describe concentration polarization, the literature usually resorts to an integral method [3, 4], whose drawbacks are discussed in [5]. In [6], using a semi-integral method [5], a description is given for concentration polarization in axisymmetric membrane elements (hollow fibers and pipes) in a pre-gel regime.

Below we consider the gel regime of polarization in continuous-flow laminar ultrafiltration with nonideal selectivity of the membrane in a pipe filter.

With allowance for gelation, we obtain the velocity distribution in a cylindrical channel. We assume that the flow at the channel inlet is fully developed. The flow rate of the fluid through the channel cross section considerably exceeds the flow leaving through the membrane, and the thickness of the gel layer is much smaller than the channel radius. Then the equations of motion and continuity take the form

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

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

$$\hat{v}'_r + \frac{\hat{v}_r}{r} = -\hat{u}'_z. \tag{3}$$

under the following boundary conditions:

$$\hat{u} = 0, \quad \hat{v} = V_\delta \quad (r = R - \delta); \tag{4}$$

$$\hat{v} = 0, \quad \hat{u}'_r = 0 \quad (r = 0). \tag{5}$$

From Eq. (2) it follows that $p = p(z)$.

Integrating Eq. (1) with allowance for the first boundary condition (4), we obtain

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$$\hat{u} = -\frac{R^2}{4\mu} p_z' \left( \frac{(R-\delta)^2}{R^2} - \frac{r^2}{R^2} \right) . \tag{6}$$

The equation of continuity yields

$$\hat{v} = -\frac{R^3}{16\mu} \frac{\partial}{\partial z} p_z' \left( \frac{r^3}{R^2} - \frac{2(R-\delta)^2 r}{R^2} \right) . \tag{7}$$

Hence

$$\hat{V}_\delta = \frac{R^3}{16\mu} \frac{\partial}{\partial z} p_z' \left( 1 - \frac{\delta}{R} \right)^3 . \tag{8}$$

The mean flow rate at the channel inlet with $z = 0$ is given by the expression

$$\bar{u}_0 = \frac{R^2}{8\mu} \left( -\frac{\partial p}{\partial z} \right) . \tag{9}$$

Integrating Eq. (8) with account for relation (9) and substituting the result into Eq. (6), we obtain an expression for the longitudinal velocity component:

$$\hat{u} = \frac{2}{R\left(1 - \frac{\delta}{R}\right)^3} \left( R\,\bar{u}_0 - 2\int_0^z V_\delta \, dz \right) \left( \frac{(R-\delta)^2}{R^2} - \frac{r^2}{R^2} \right) . \tag{10}$$

Now, we consider the equation of convective diffusion. It is taken into account that in ultrafiltration the thickness of the diffusive boundary layer $\Delta$ is much smaller than the pipe radius $R$. Introducing the new variable $y = R - r$ and retaining the principal terms, we obtain an equation of convective diffusion, which is written in a conservative form (here and below we will operate with the dimensionless quantities $u$, $v$, $V_\delta$, $\eta$, $\xi$):

$$\frac{\partial(\Theta - 1)}{\partial \tau} + \frac{\partial u(\Theta - 1)}{\partial \xi} - \frac{\partial v(\Theta - 1)}{\partial \eta} = \frac{1}{\mathrm{Pe}} \frac{\partial^2 \Theta}{\partial \eta^2} \tag{11}$$

Equation (11) describes the pre-gel and gel regimes of polarization but under different boundary conditions. The first regime exists from the channel inlet up to a certain point $\xi_1$, which is called the gelation point, where the concentration on the membrane attains the concentration of gelation $\Theta_g$. After the gelation point downstream the second regime of polarization is realized. As noted above, the pre-gel regime was analyzed in [6].

Now, we consider the gel regime.

The boundary conditions after the gelation point, $\xi \geq \xi_1$, take the form

$$u|_{\eta=\delta} = 0 , \quad v|_{\eta=\delta} = V_\delta , \tag{12}$$

$$\varphi V_\delta \Theta_g + \frac{1}{\mathrm{Pe}} \Theta_\eta' \Big|_{\eta=\delta} = \Theta_g \delta_\tau' , \tag{13}$$

$$\Theta|_{\eta=\Delta} = 1 , \quad \Theta|_{\eta=\delta} = \Theta_g . \tag{14}$$

We relate the decrease in the permeability of the membrane to the thickness of the gel layer:

$$V_\delta = \frac{V}{1 + k\delta} . \tag{15}$$

Now we integrate the equation of convective diffusion (11) across the diffusive boundary layer using conditions (12)-(14) and the requirement that $\Theta_\eta' = 0$ at $\eta = \Delta$. Then for $\xi > \xi_1$ we have

$$\frac{\partial}{\partial \tau} \int_\delta^\Delta (\Theta - 1)\, d\eta + (2\Theta_g - 1) \frac{\partial \delta}{\partial \tau} + \frac{\partial}{\partial \xi} \int_\delta^\Delta u\, (\Theta - 1)\, d\eta = V_\delta \Gamma_g\,, \tag{16}$$

where $\Gamma_g = 1 - (1 - \varphi)\Theta_g$. At the gelation point $\xi = \xi_1$ itself the condition $\Theta = \Theta_g$ is satisfied.

We consider Eq. (11) under two limiting conditions: in a highly nonstationary regime and in a stationary regime. For the stationary case, with allowance for the distribution of velocities (7), (8), (10) (since the diffusive-layer thickness is small, we can restrict ourselves to the first terms in the distribution of velocities (7), (10)) we obtain for $\xi > \xi_1$ the relation

$$4 \left(1 - 2V\xi_1 - 2 \int_{\xi_1}^{\xi} V_\delta\, d\xi\right) (\eta - \delta) \frac{\partial \Theta}{\partial \xi} = V_\delta \frac{\partial \Theta}{\partial \eta} + \frac{1}{\mathrm{Pe}} \frac{\partial^2 \Theta}{\partial^2 \eta}\,. \tag{17}$$

In the immediate vicinity of the membrane, i.e., for $\eta \to \delta$, the following relation is valid:

$$V_\delta \frac{\partial \Theta}{\partial \eta} + \frac{1}{\mathrm{Pe}} \frac{\partial^2 \Theta}{\partial^2 \eta} \approx 0\,. \tag{18}$$

Double integration of this relation and application of boundary condition (13) allow us to find that

$$\Theta = \Theta_g \left[1 - \varphi\, (1 - \exp(-\,\mathrm{Pe}\, V_\delta\, (\eta - \delta)))\right].$$

From the physical considerations underlying boundary-layer theory and Eq. (18), the distribution of concentrations for $\xi \geq \xi_1$ can be represented in the form

$$\Theta = \begin{cases} \Theta_g\,, & 0 < \eta \leq \delta\,, \\ \Theta_g \left[1 - \varphi\, (1 - \exp(-\,\mathrm{Pe}\, V_\delta\, (\eta - \delta)))\right], & \delta \leq \eta \leq \Delta\,, \\ 1\,, & \Delta \leq \eta \leq 1\,, \end{cases} \tag{19}$$

where $V_\delta(\xi)$ is a function as yet unknown. To calculate this function, we use integral condition (16). It should be noted preliminarily that Eq. (19) yields

$$\Delta - \delta = \frac{1}{\mathrm{Pe} V_\delta} \ln \frac{\varphi \Theta_g}{1 - (1 - \varphi)\, \Theta_g}\,. \tag{20}$$

Then, substituting the distribution of velocities (17) and concentrations (19) into Eq. (16) and performing single integration over $\xi$, we find for $\xi \geq \xi_1$

$$4 \left(1 - 2V\xi_1 - 2 \int_{\xi_1}^{\xi} V_\delta d\xi\right) \Sigma_g/(\mathrm{Pe} V_\delta)^2 = \int_{\xi_1}^{\xi} \Gamma_g\, V_\delta d\xi + \int_0^{\xi_1} \Gamma_w V d\xi_1\,, \tag{21}$$

where

$$\Sigma_g = \Theta_g - \Gamma_g \ln \frac{\varphi \Theta_g}{\Gamma_g} - \frac{1}{2} \Gamma_g \left(\ln \frac{\varphi \Theta_g}{\Gamma_g}\right)^2 - 1\,. \tag{22}$$

Then we determine the position of the gelation point on the membrane $\xi_1$:

$$\int_0^{\xi_1} \Gamma_w V d\xi = \frac{4\,\Sigma_g\,(1 - 2V\xi_1)}{(\mathrm{Pe}V)^2}. \tag{23}$$

Substituting two limiting cases into the left-hand side of Eq. (23) instead of $\Theta_w$: 1) $\Theta_w = \Theta_g$; 2) linear behavior $\Theta_w = 1 + V\xi(\Theta_g - 1)/V\xi_1$, and performing integration, we obtain the estimate

$$\frac{4\,\Sigma_g}{(\mathrm{Pe}V)^2\,\Gamma_g} \geq \frac{V\xi_1}{1 - 2V\xi_1} \geq \frac{8\,\Sigma_g}{(\varphi + \Gamma_g)\,(\mathrm{Pe}V)^2}. \tag{24}$$

We resolve Eq. (21) for the integral and then perform the differentiation with respect to $\xi$ with subsequent integration with the boundary condition $V_\delta = V$ at $\xi = \xi_1$. This yields an equation that describes the regime of gel polarization in a cylindrical channel:

$$\frac{1 - 2V\xi}{1 - 2V\xi_1} = \frac{V_\delta}{V}(1 + F)\Big/\left(\frac{V_\delta^2}{V^2} + F\right) - \frac{(1 + F)}{F^{1/2}}\left[\arctan\frac{V_\delta}{VF^{1/2}} - \arctan\frac{1}{F^{1/2}}\right], \tag{25}$$

where $F = 4\Sigma_g/\Gamma_g(\mathrm{Pe}V)^2$. When $V\xi_1 \to 0$, we can obtain a simpler solution:

$$\frac{V_\delta}{V} = \left(1 + \frac{3}{8}(V\xi - V\xi_1)\frac{\Gamma_g\,(\mathrm{Pe}V)^2}{\Sigma_g}\right)^{-1/3}, \tag{26}$$

which for $V\xi/V\xi_1 >> 1$ can be transformed to yield

$$V_\delta \approx \left(\frac{8}{3}\,\frac{\Sigma_g}{\Gamma_g\,\mathrm{Pe}^2\xi}\right)^{1/3}. \tag{27}$$

The velocity $V$ (the resistance of the membrane) does not enter into formula (27). This means that the performance of the filter ceases to depend on the pressure (the resistance of the gel layer considerably exceeds the resistance of the membrane, and an increase in pressure is compensated by an increase in the resistance of the gel layer).

Thus, the pattern of laminar continuous-flow ultrafiltration in a cylindrical channel can be divided into three regions. In the first region, which extends from the channel inlet to the gelation point (determined by Eq. (23)), the main resistance to transmembrane flow is offered by the membrane, and the filtration velocity $V$ is directly proportional to the pressure applied [6]. In the second region, which extends from the gelation point and farther downstream, the hydraulic resistances of the membrane and the gel layer will be of the same order. Here the filtration velocity $V_\delta$ depends nonlinearly on the pressure (Eq. (25)) (the pressure is associated with $V$). In the third region, the hydraulic resistance of the gel layer considerably exceeds the resistance of the membrane, the filtration velocitty ceases to depend on the initial pressure, and the entire velocity distribution of filtration from the above two regions is reduced to a single dependence (Eq. (27)).

We consider the unsteady-state regime of ultrafiltration. For this case we have the integral condition

$$\frac{\partial}{\partial\tau}\int_\delta^\Delta (\Theta - 1)\,d\eta + (2\Theta_g - 1)\frac{\partial\delta}{\partial\tau} = \Gamma_g V_\delta, \quad \tau \geq \tau_1, \tag{28}$$

where $\tau_1$ is the time of onset of gelation.

We relate the decrease in the permeability of the membrane to the gel-layer thickness by relation (15).

We prescribe the nonstationary distribution of concentration from the solution of the stationary problem. Then, substituting formula (19) into Eq. (28) with allowance for the fact that $V_\delta$ and $\delta$ depend on the time and there is the obvious relationship

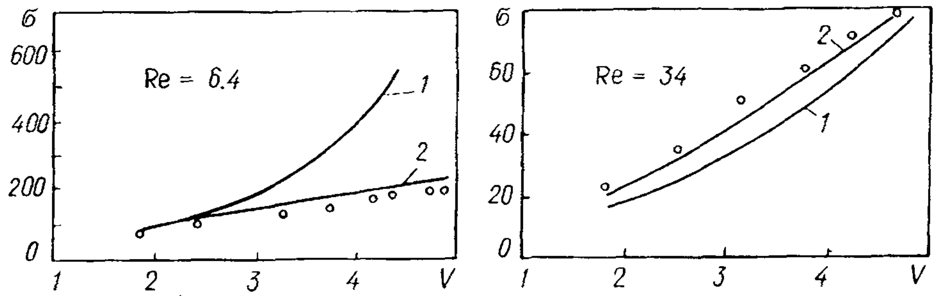


Fig. 1. Comparison between dependences of experimental values of $\sigma$ (mg/m$^2$) (the amount of protein in a polarized layer) and ones calculated by integral and semi-integral methods on the filtration velocity $V$ ($\mu$m/sec) for various values of the Re number: points, experiment; 1, integral method; 2, semi-integral method.

$$\frac{\partial \delta}{\partial \tau} = - \frac{V}{k V_\delta^2} \frac{\partial V_\delta}{\partial \tau}, \tag{29}$$

we obtain

$$\frac{V_\delta}{V} = \left[ 1 + \frac{2 (V\tau - V\tau_1)}{\dfrac{\Sigma_\tau}{\mathrm{Pe} V \Gamma_g} + \dfrac{(2\Theta_g - 1)}{k}} \right]^{-1/3}, \quad \tau \geq \tau_1, \tag{30}$$

where

$$\Theta_g - \Gamma_g \ln \frac{\varphi \Theta_g}{\Gamma_g} - 1 = \Sigma_\tau .$$

Now, we estimate the time $\tau_s$ that is required for attaining steady-state ultrafiltration in a cylindrical channel with gelation. Assuming that the quantities $V\xi_1$ are small, we equate the right-hand sides of Eqs. (30) and (26):

$$V\tau_s = V\tau_1 + \frac{1}{2} \left[ \left( 1 + \frac{3}{8} (V\xi - V\xi_1) \frac{\Gamma_g (\mathrm{Pe} V)^2}{\Sigma_g} \right)^{2/3} - 1 \right] \left( \frac{2\Theta_g - 1}{k} + \frac{\Sigma_g}{\mathrm{Pe} V \Gamma_g} \right). \tag{31}$$

For distances from the channel inlet for which $V\xi / V\xi_1 >> 1$, we obtain

$$V\tau_s = \frac{1}{2} \left( \frac{3}{8} \frac{V\xi\, \Gamma_g (\mathrm{Pe} V)^2}{\Sigma_g} \right)^{2/3} \left( \frac{2\Theta_g - 1}{k} + \frac{\Sigma_\tau}{\mathrm{Pe} V \Gamma_g} \right). \tag{32}$$

We compare the theoretical results obtained, which describe ultrafiltration in the pre-gel and gel regimes in an axisymmetric filter, with experimental data. Figure 1 illustrates experimental data [7] on determination of the amount of protein $\sigma$ in a polarization layer for the pre-gel regime of ultrafiltration in a hollow-fiber filter. From this figure it follows that the results obtained by a semi-integral method [6] describe the experimental data much better than the relations determined by an integral method [7].

And, finally, we compare experimental values of the limiting velocity of ultrafiltration $V_\delta$ for the gel regime of polarization with ones predicted by a semi-integral method (Eq. (27)). The authors failed to discover experimental data on ultrafiltration in a pipe filter in a gel polarization regime. There are results of experiments [8] on ultrafiltration in a gel regime in a plane channel. A comparison of these data with results of theoretical investigations carried out on the basis of the semi-integral method in [9] for a plane channel is presented in Table

TABLE 1. Comparison of Experimental Values of the Limiting Filtration Velocity $V_\delta$ with Ones Calculated by a Semi-Integral Method

| Volumetric concentration $c_0$, g/m$^3$ | Flow velocity $\bar{u}_0$, cm/sec | Limiting velocitty of filtration $V_\delta$, cm/min | |
|---|---|---|---|
| | | experiment | theory |
| 1.87 | 5.8 | 0.018 | 0.020 |
| 1.80 | 11.5 | 0.022 | 0.025 |
| 1.78 | 17.3 | 0.025 | 0.030 |
| 1.76 | 23.0 | 0.028 | 0.033 |
| 1.74 | 34.5 | 0.039 | 0.038 |

1; good agreement between them is evident. This allows one to be sure that the formulas obtained above for the gel regime of polarization in a pipe filter will give an adequate description of experimental results.

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## NOTATION

$\xi = x/R$; $\eta = y/R$, dimensionless longitudinal and transverse coordinates; $u = \hat{u}/\bar{u}_0$, $v = \hat{v}/\bar{u}_0$, dimensionless components of the velocity vector; $R$, pipe radius; $u_0$, average velocity at the channel inlet; $\mathrm{Re} = \bar{u}_0 R/\nu$, Reynolds number; $\nu$, kinematic-viscosity coefficient; $\mathrm{Pe} = \bar{u}_0 R/D$, diffusion Peclet number; $D$, diffusion coefficient; $\Theta = c/c_0$, dimensionless concentration of the substance; $c_0$, concentration of the dissolved substance at the channel inlet; $\Theta_g$, dimensionless concentration of the dissolved substance in the gel layer; $\Theta_w$, dimensionless concentration of the dissolved substance on the membrane; $V = \hat{V}/\bar{u}_0$, transmembrane velocity; $V_\delta$, transmembrane velocity with gelation; $\Delta$, dimensionless width of the diffusive boundary layer; $\delta$, dimensionless width of the gel layer; $\varphi$, selectivity of the membrane; $\mu$, dynamic-viscosity coefficient.

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