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Use of Cake Deposition to Improve the Efficiency of Ultra- and Microfiltration Plants

Yu. S. Polyakov
USPolyResearch, Ashland, Pennsylvania 17921, USA
e-mail: ypolyakov@uspolyresearch.com

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Abstract—Fundamental principles and topics of a new approach to designing ultra- and microfiltration plants in which cake deposition and its control are used to improve the membrane filtration efficiency are discussed. A general phenomenological mathematical model for the process of depth membrane filtration, which was suggested by the new approach, is formulated and methods of its numerical and approximate solutions are described. The available particular solutions to the DMF model are analyzed and classified, and recommendations for their practical use are given. The use of the new approach for increasing the efficiency of existing deadend outside-in hollow fiber membrane filters is also discussed. A general mathematical model is formulated for this type of deadend filters and methods of its numerical and approximate solutions are discussed. It is shown that the highest efficiency of outside-in hollow fiber membrane filters can be achieved when the collection efficiency of the membrane surface with respect to suspended particles is highest.

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In separation processes using semipermeable membranes, the components that passed through the pores of the membrane are withdrawn from the feed solution, whereas the retained components are accumulated near the membrane surface, giving rise to concentration polarization. In this case, the retained components can adsorb to the membrane surface and form a gel layer or cake [1]. The increase in osmotic pressure and flow resistance caused by the concentration polarization and cake decreases the driving force of the separation process, transmembrane pressure, which will have a negative impact on the efficiency of membrane operations.

The conventional approach to designing cross-flow ultrafiltration (UF) and microfiltration (MF) plants is based on decreasing the concentration polarization and cake thickness by intensely agitating the liquid near the membrane surface [2–4]. In most cases, the liquid agitation or turbulization is produced by increased tangential flow velocities near the membrane surface, which is accompanied by additional energy and material losses [5].

The conventional approach is based on a considerable number of different mathematical models developed within four major groups: convective diffusion, hydraulic resistances in series, particle trajectory analysis, and turbulent transport in terms of empirical correlations of dimensionless numbers [4, 6, 7]. These models were used to formulate the following recommendations for the design and operation of UF and MF plants:

(1) Tangential flow velocity and local flow instabilities (eddies) near the membrane in the feed-solution channel should be as high as possible to reduce the concentration polarization and particle deposition.

(2) Adsorption of particles to the membrane surface and the interaction between the particles in the boundary layer should be reduced to a minimum.

(3) Specific cake resistance should be as low as possible.

As a result, the membrane technologists directed almost all their efforts toward implementing the above recommendations. Specifically, they suggested use of high tangential velocities in narrow channels, turbulence promoters of different types, corrugated-membrane wafers, curved membrane channels, Dean vortices, ultrasonic agitation, vibration, pulse feed flow, bubbling, low transmembrane pressure, short membrane channels, low-adsorption membranes to particles, and so on [5]. In spite of the fact that all these improvements made it possible to design high-capacity commercial plants, they also resulted in much higher power consumptions and more complicated designs. As a result, ultra- and microfiltration are, as a rule, less profitable than conventional treatment operations such as sedimentation, coagulation, depth filtration, and the like for many water and wastewater treatment applications [6].

One of the alternative approaches to designing UF and MF plants could be the principle of agitation based on the periodic mechanical "sweeping" of cake away from the membrane surface by wipers [8]. It was convincingly shown that a rotating magnetic stirrer equipped with porous plastic wipers touching the membrane surface makes it possible to reduce the modulus of concentration polarization by several times as compared with the same magnetic stirrer without wipers rotating at the same speed. The same laboratory cell was used to show that the wipers almost completely remove the cake layer when concentrated protein solutions are treated. Unfortunately, this approach was not further developed because plastic wipers could damage the thin selective layer of composite polymer membranes. In view of wide implementation of ceramic and other hard membranes in the past ten years, this approach could become a profitable alternative to the conventional techniques.

The law of mass conservation states that the accumulation of particles at one point leads to their depletion, or lack, at another point. This principle has already been successfully used for many years in water treatment, where the particles to be removed from a suspension are caught by particle collectors, such as adsorbents and granular or sand beds, to produce clarified water (filtrate) at the filter outlet [9, 10]. In this case, the particles are deposited at a much higher rate on the inlet layers of collectors and at a much lower rate on the layers of collectors near the filter outlet, providing the decrease in the concentration of particles across the filter; that is, the clarification of the feed suspension. It is obvious that the membrane surface likewise plays the role of a particle collector in UF and MF filters. In contrast to adsorbent and granular beds, the deposition of cake in traditional UF and MF filters has only a negative impact on their efficiency: it reduces the flux of permeate (water permeated through membranes). At the same time, the particle collecting ability of membranes reduces the concentration of particles in the suspension flowing to the filter outlet on the high-pressure membrane side. Consequently, under certain conditions this ability could be used to yield an additional volume of clarified water produced by the flow of water flowing through the high-pressure channel, which would be additional to the flow of water permeated through the walls of semipermeable membranes. In this case, there would be no need to suppress cake deposition because a sufficiently high yield of clarified product could be provided due to the additional flow of filtrate, which is a result of the collection of suspended particles by membranes. This separation process could become a successful alternative to conventional UF and MF operations, which are based on suppressing cake deposition at the cost of additional expenditures.

Traditional granular bed filters are usually characterized by high packing densities of collectors (grains) and low suspension flow velocities across the filter [9, 10]. The corresponding characteristics of outside-in hollow fiber membrane filters are the same. The membrane packing densities in the existing hollow fiber filters are as high as 0.5–0.6, and the liquid tangential velocity around the hollow fibers is about 10^{-4} to -10^{-3} m/s [3]. This implies that a filter made of a bundle of impermeable hollow fibers with a high particle collecting ability could be a rather efficient depth filter. If the hollow fibers in this filter would be semipermeable, it would produce two flows of clarified water during the initial operation period: permeate, produced by the permeation of liquid through the membrane walls, and filtrate, produced by the collection of particles by the shells of hollow fibers at high rates on their inlet layers and at low rates on the layers at the filter outlet (due to the radically decreased concentration of particles in the outlet zone). The filtration process in this filter is called depth membrane filtration (DMF). It is interesting to note that this approach to designing UF and MF filters has much in common with the principles of the cycle of substances in nature, where, as in the above case, several processes beneficially complement each other, providing the optimal balance of substances. In the case of carbon cycle, carbon dioxide is produced by the breathing of living organisms and the decomposition of dead organic matter (remains of herbs and animals) is accomplished by reducers. At the same time, carbon dioxide is absorbed by producers and converted by photosynthesis into organic compounds, which are consumed by consumers. In depth membrane filtration, two beneficially complementing processes are membrane separation, in which the cake deposition reduces the efficiency, and depth filtration, in which the treatment efficiency is achieved by intense particle deposition; that is, cake growth.

The above idea was used to propose two designs of DMF filters: cartridge of wafers (Fig. 1) and radial (Fig. 2) [7, 11–14]. The distinctive feature of DMF filters as compared to conventional types of UF and MF filters is the presence of an additional flow of clarified liquid, filtrate. Deadend UF and MF filters are characterized by a single product flow, permeate. Conventional cross-flow membrane filters produce two flows: permeate and retentate (concentrated suspension), the concentration of particles in the latter is higher than in the feed suspension. Consequently, the DMF filter would have several significant advantages, such as a low-power-consumption operation with a constant flow rate of clarified water at constant pressure, high-water-recovery multistage flowsheet of DMF modules using the filtrate leaving the first stage as the feed for the second stage, and the absence of concentrated retentate, which should be utilized or aftertreated.

Here, it is important to pay attention to the feature of depth membrane filtration that makes it much distinctive from the classical flow diagrams used in chemical engineering. The classical flowsheets are often based on the arrangement of different unit operations in series, where the performance of its separate stages, represented as "black boxes" with inputs and outputs, is optimized stage by stage for the whole flowsheet. In depth membrane filtration, the treatment efficiency can likewise be maximized by optimizing the interaction between these two unit operations, but this should be

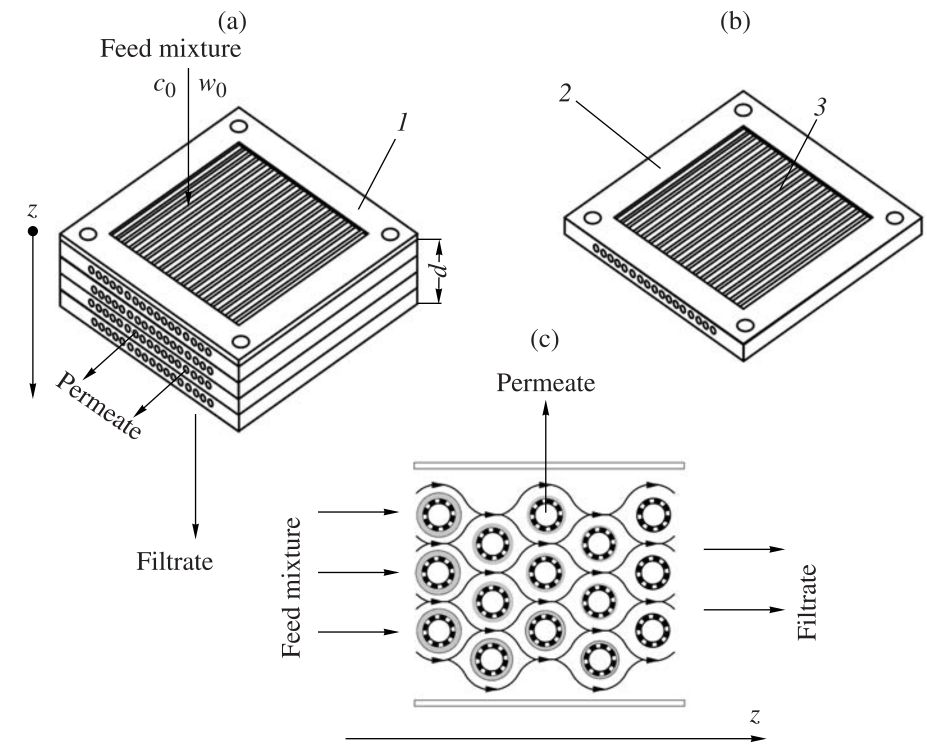

Fig. 1. Rectangular cartridge-type DMF filter: (a) cartridge of hollow fiber wafers (*1*, top plate), (b) separate wafer (*2*, perforated frame; *3*, hollow fiber membrane), (c) flow diagram (gray solid rings—cake layers; porous rings—hollow fibers) [7].

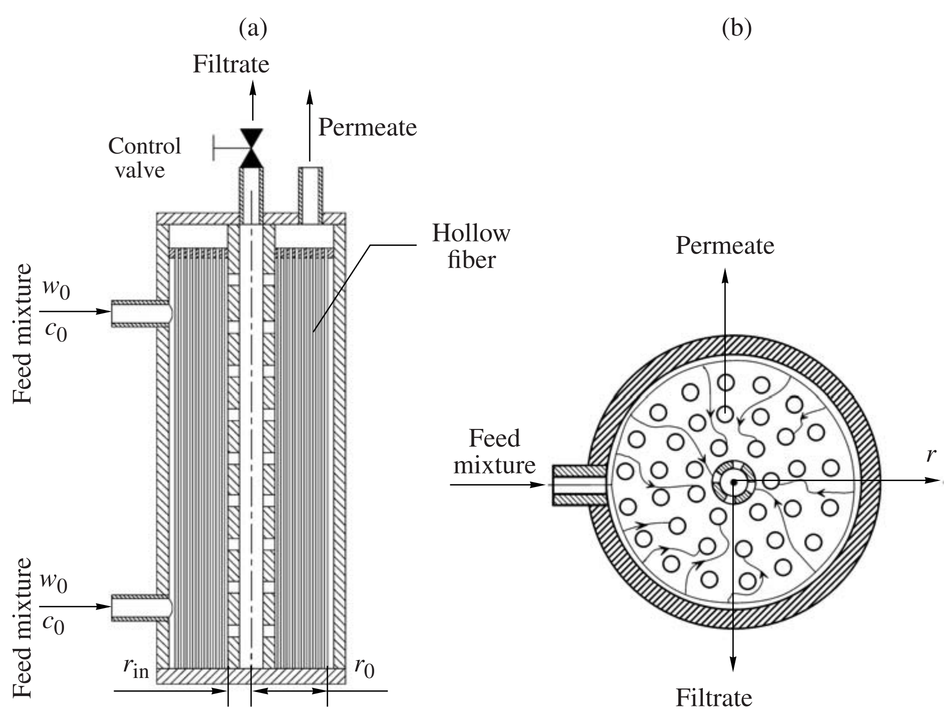

Fig. 2. Radial DMF filter: (a) flow diagram in vertical cross section, (b) flow diagram in horizontal cross section [7].

done with regard to the fact that these operations are concurrent to each other rather than in series and beneficially complement each other. On one hand, this could considerably improve the filter performance. On the other hand, it causes significant difficulties in the mathematical modeling of the process. In this case, the model should consider the coupling of the two processes, which inevitably leads to complex nonlinear integrodifferential equations. In essence, this approach has much in common with synergetics, the distinctive feature of which is the necessity of studying the cooperative action of several processes. As a result, the simulation of depth membrane filtration is associated with a set of difficulties that is typical for synergetic models.

The phenomenological mathematical model [7, 15] developed to describe the operation of DMF filters, in which the pores of the semipermeable membranes retain the particles suspended in the slurry, was formulated using the combination of conventional equations for depth filtration and membrane separation. It includes the law of mass conservation written in differential form:

$$\frac{\partial c}{\partial t} + \frac{\partial (cw)}{\partial z} = -s\frac{\partial \Gamma}{\partial t}, \tag{1}$$

the overall kinetic equation accounting for the growth of cake mass on the membrane surface:

$$\frac{\partial \Gamma}{\partial t} = k_1(\psi_1, \Gamma)c - k_2(\psi_2, \Gamma)\Gamma + k_3 V_p c, \tag{2}$$

the fluid-continuity equation written in integral form:

$$w = w_0 - \int_0^z s V_p dz, \tag{3}$$

the Darcy law accounting for the decline of the permeation flux across the membrane walls:

$$V_p = \frac{P}{\mu(R_m + r_c\Gamma)}. \tag{4}$$

Here, $R_m = P/(\mu V_0)$.

In this case, we use the “clean-filter” initial condition and assume that the suspension concentration at the filter inlet remains constant:

$$c = c_0 \quad \text{when } z = 0, t > 0; \tag{5}$$

$$c = 0, \Gamma = 0 \quad \text{when } t = 0, z > 0. \tag{6}$$

It should be noted that the model given by (1)–(6) can be used both for the cartridge-type filter (Fig. 1) and for the radial filter (Fig. 2). In the latter case, it is necessary to introduce the effective filter-depth coordinate [7, 12]:

$$z = \frac{r_0^2 - r^2}{2r_0}, \tag{7}$$

which reduces the radial-filter problem to the mathematical model given by (1)–(6).

As hollow fiber membranes are not impermeable collectors of particles, continuity equation (3) is written as an integral equation and the overall system of Eqs. (1)–(3) is reduced to a complex nonlinear integrodifferential equation for Γ:

$$\frac{\partial}{\partial t}\left\{\frac{\frac{\partial \Gamma}{\partial t} + k_2(\psi_2, \Gamma)\Gamma}{k_1(\psi_1, \Gamma) + k_3 V_p(\Gamma)}\right\} + \frac{\partial}{\partial z}\left[\frac{\frac{\partial \Gamma}{\partial t} + k_2(\psi_2, \Gamma)\Gamma}{k_1(\psi_1, \Gamma) + k_3 V_p(\Gamma)}\left(w_0 - \int_0^z s V_p(\Gamma) dz\right)\right] = -s\frac{\partial \Gamma}{\partial t} \tag{8}$$

subject to the following initial and boundary conditions:

$$\Gamma = 0, \partial\Gamma/\partial t = 0 \quad \text{when } t = 0, z > 0; \tag{9}$$

$$\Gamma = \Gamma_0 \quad \text{when } z = 0, t > 0. \tag{10}$$

Here, $V_p(\Gamma)$ is given by Eq. (4) and Γ_0 is determined by integrating the equation:

$$\frac{d\Gamma_0}{dt} = [k_1(\psi_1, \Gamma_0) + k_3 V_p(\Gamma_0)]c_0 - k_2(\psi_2, \Gamma_0)\Gamma_0$$

with the initial condition:

$$\Gamma_0 = 0 \quad \text{when } t = 0.$$

The problem given by (8)–(10) does not have an exact analytical solution. It can be solved numerically by passing from Γ to a new function:

$$\nu = \int_0^z V_p(\Gamma) dz.$$

As a result, we come to a nonlinear partial differential equation the solution of which can be found by the generalized Crank–Nicholson method of central finite differences [7]. However, this numerical solution requires a considerable computation time even with modern powerful computers, which makes it difficult to use this method for practical calculations, where we often try to find the optimal relations between the performance and process parameters.

For this reason, a much faster approximate method for designing DMF filters, the central idea of which is closely related to the physics of interaction between membrane separation and depth filtration, was developed [7, 15, 16]. The approximate solution on which the method is based can be used for a wide class of integrodifferential and partial differential equations. Using the idea of averaging the permeate velocity over certain

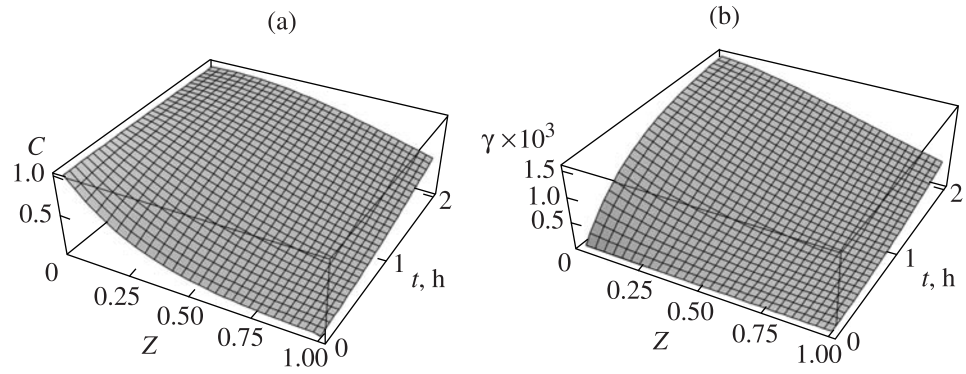

Fig. 3. Profiles of (a) dimensionless concentration of suspended particles C and (b) dimensionless cake deposit γ in a DMF filter: $k_1 = 1.81 \times 10^{-4}$ m/s, $k_2 = 4.20 \times 10^4$ 1/s, $s = 3.88 \times 10^3$ 1/m, $V_0 = 1.39 \times 10^{-5}$ m/s, $\xi = 0.99$, $d = 0.05$ m, $N_\chi = r_c c_0/R_m s = 0.0072$ [14].

space and time intervals, we can divide the original problem into two parts. The first represents the classical model of depth filtration with a corrected coordinate for the filter depth. The second is represented by the Darcy law, which is the main equation for describing the membrane separation.

The simultaneous solution of these two equations by the iterative procedure makes it possible to obtain approximate relations for main process parameters, such as the permeate flux averaged over filter depth, concentration of particles in filtrate, and the concentration of particles in clarified product (filtrate plus permeate), the deviation of which from the corresponding curves found by the numerical solution usually does not exceed 10%. Actually, the approximate method "emulates" how these two concurrent filtration processes interact with each other in a real hollow fiber membrane filter. Mathematically, the approximate problem reduces to the following differential and integral equations:

$$\frac{\partial}{\partial t}\left\{\frac{\dfrac{\partial\Gamma}{\partial t}+k_2(\psi_2,\Gamma)\Gamma}{k_1(\psi_1,\Gamma)+k_3\langle V_p\rangle}\right\}$$

$$+\frac{\partial}{\partial z}\left[\frac{\dfrac{\partial\Gamma}{\partial t}+k_2(\psi_2,\Gamma)\Gamma}{k_1(\psi_1,\Gamma)+k_3\langle V_p\rangle}(w_0-s\langle V_p\rangle z)\right] = -s\frac{\partial\Gamma}{\partial t},$$

$$\langle V_p\rangle = \frac{1}{tz}\int_0^t\int_0^z\frac{P}{\mu(R_m+r_c\Gamma[t_1, z_1, \langle V_p\rangle])}(dz_1)dt_1,$$

which, combined with appropriate initial and boundary conditions, were solved using the effective algorithm developed by Polyakov et al. [16].

The problem given by (8)–(10) was solved for three particular cases of general kinetic equation (2):

(1) k_1 and k_2 are constant parameters, $k_3 = 0$. Mathematically, this corresponds to the linear equation for a reversible reaction of the first order with constant coefficients [7].

(2) k_1 and k_2 are certain functions of V_p, $k_3 = 0$. Mathematically, this corresponds to the linear equation for a reversible reaction of the first order with variable coefficients, the dependence of which on the permeate velocity can be determined using the steady-state Smoluchowski approximation [11, 12].

(3) $k_2 = 0$ with all other things as in Eq. (2). This is the generalization of the phenomenological expression of the theory of depth filtration for the case of permeable collectors, in which the deposition coefficient is an arbitrary function of local cake mass and permeate velocity [15].

The first case is based on linearized equation (2) and is convenient for conducting qualitative studies and determining the optimal relations between the main process parameters [14, 17]. The second case can be used for studying the effect of the variation of permeate velocity on the particle collecting ability of hollow fiber membranes. The third case is the best for processing experimental data because the deposition of particles in real filters is, as a rule, irreversible and all other parameters in Eq. (2) can be chosen rather arbitrarily.

Calculations showed that the profiles of specific cake deposit γ (Fig. 3b) and particle concentration C (Fig. 3a) in depth membrane filtration have a wave-like shape [11, 14], much like those in conventional depth filtration. Initially, the particles intensely deposit on the inlet layers of hollow fibers. Later, they begin to intensely deposit on deeper layers. In this case, the higher is the value of deposition coefficient k_1, the larger is the number of particles deposited at the filter inlet and the larger is the time during which the filtrate (additional flow of clarified water) can be withdrawn from the filter [11, 14]. In addition, this nonuniform

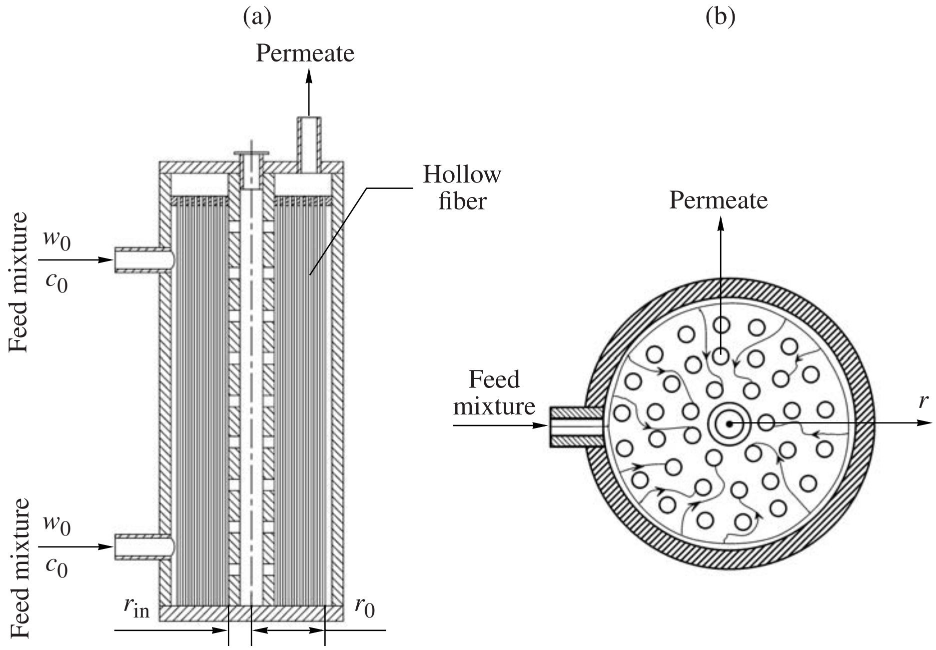

Fig. 4. Radial deadend outside-in hollow fiber membrane filter: (a) vertical cross-section flow diagram and (b) horizontal cross-section flow diagram [19].

deposition of particles along the filter depth leads to a higher value of the permeate flux produced by the whole filter as compared to the filter with a uniform deposit profile along the filter depth. These results contradict the conventional concept that the increased particle collecting ability of UF and MF membranes has a negative impact on the filter performance. In contrast to this concept, they suggest that the efficiency of UF and MF filters can be improved by increasing the rate of cake deposition.

The positive effect that can be achieved using the nonuniform profile of the cake layer along the filter depth makes us reconsider the approach to maximizing the filtration efficiency in deadend outside-in hollow fiber membrane filters (Fig. 4) [18, 19]. The traditional mathematical models for these filters assume a uniform cake profile along the filter depth [4], which is not in agreement with the results of experimental studies for the cake profiles in the filtration using bundles of hollow fibers [20]. The mathematical model for the deadend filter that takes into account the depth nonuniformity of the filtration characteristics at constant transmembrane pressure and ideal (100%) selectivity of membrane pores with respect to particles is analogous to the model given by Eqs. (1)–(6) for a DMF filter except for the continuity equation [18, 19], which in view of expression (7) can be written as

$$w = \int_z^d s V_p dz. \tag{11}$$

In this case, the system of Eqs. (1), (2), (4)–(6), (11) is reduced to the integrodifferential equation:

$$\frac{\partial}{\partial t}\left\{\frac{\dfrac{\partial \Gamma}{\partial t} + k_2(\psi_2, \Gamma)\Gamma}{k_1(\psi_1, \Gamma) + k_3 V_p(\Gamma)}\right\} + \frac{\partial}{\partial z}\left[\frac{\dfrac{\partial \Gamma}{\partial t} + k_2(\psi_2, \Gamma)\Gamma}{k_1(\psi_1, \Gamma) + k_3 V_p(\Gamma)}\int_z^d s V_p dz\right] = -s\frac{\partial \Gamma}{\partial t} \tag{12}$$

subject to the initial and boundary conditions given by expressions (9) and (10).

The problem defined by Eqs. (9), (10), and (12) does not have an exact analytical solution. It can be solved numerically by transferring from Γ to a new function:

$$\nu = \int_z^d V_p(\Gamma) dz.$$

This transfer results in a nonlinear partial differential equation the solution of which can be found by the generalized Crank–Nicholson method of central finite differences [18]. As in the case of a DMF filter, this numerical solution requires high computation times even with modern powerful computers, which makes it difficult to use this method in filter design. At the same time, the approximate method used for a DMF filter makes it possible to obtain a solution to this problem

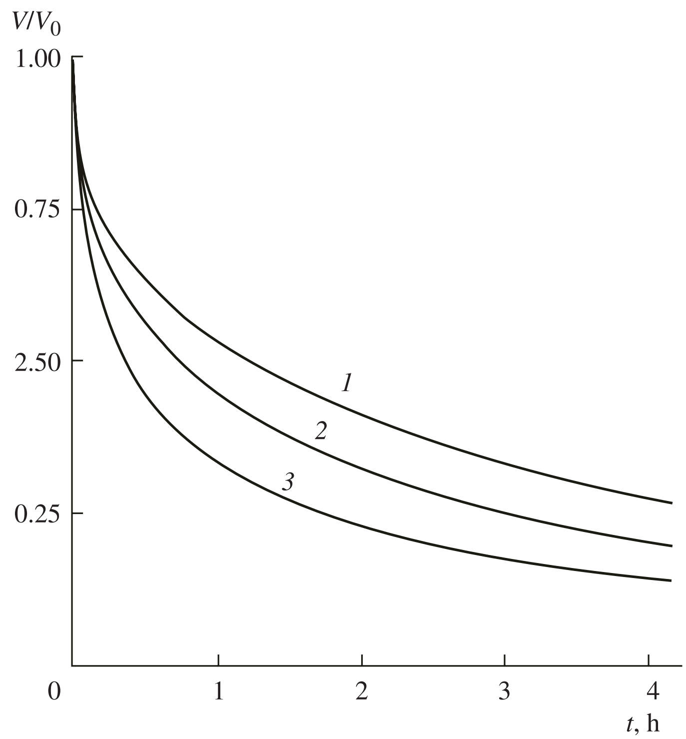

Fig. 5. Dependence of the permeate flux of a deadend outside-in hollow fiber membrane filter on time: $V_0 = 6.94 \times 10^{-5}$ m/s; $k_2 = 4.2 \times 10^{-4}$ 1/s; $N_\chi = r_c c_0/R_m s = 0.0072$, $s = 3.88 \times 10^3$ 1/m; $k_1 = 5.43 \times 10^{-4}$ = (*1*) 5.43×10^{-4}, (*2*) 3.62×10^{-4}, (*3*) 1.81×10^{-4} m/s [17].

with an accuracy acceptable for engineering calculations [19].

The problem given by Eqs. (1), (2), (4)–(6), and (11) was numerically and approximately solved for the case of linearized equation (2); that is, when k_1 and k_2 are constant coefficients, $k_3 = 0$ [19]. The adequacy of this form of Eq. (2) was tested by comparing the calculated results with experimental data [18].

Mathematical modeling was used to establish that the increased particle collecting ability of membranes in deadend outside-in hollow fiber membrane filters leads, as in the case of a DMF filter, to a considerable increase in the filtration efficiency (Fig. 5) [17].

Consequently, the approach to designing UF and MF filters that uses cake deposition for increasing the filtration efficiency can lead to a significantly improved profitability of ultra- and microfiltration and their wider application in water and wastewater treatment. In addition, the key idea of the synergetic cooperation of two unit operations that can maintain the optimal material balance in the system can be used for improving the efficiency of other processes in chemical engineering.

NOTATION

$C = c/c_0$—dimensionless concentration of particles in a suspension;

c—concentration of colloidal particles, kg/m^3;

d—filter depth, m;

k_1—particle deposition coefficient, m/s;

k_2—particle re-entrainment coefficient, accounting for the re-entrainment of particles from the cake, 1/s;

k_3—constant coefficient;

N_χ—ratio of cake and clean-membrane hydraulic resistances;

P—transmembrane pressure, Pa;

R_m—hydraulic resistance of a membrane, m;

r—radial coordinate, m;

r_0—outside radius of the bundle of hollow fibers, m;

r_c—specific hydraulic resistance of a cake, m^3/kg;

r_{in}—inside radius of the bundle of hollow fibers, m;

s—specific surface of a filter, 1/s;

t—time, s;

V—permeate velocity averaged over filter depth, m/s;

V_0—initial permeate velocity, m/s;

V_p—permeate velocity, m/s;

$\langle V_p \rangle$—permeate velocity averaged over time and filter depth, m/s;

w—liquid flow velocity inside a filter, m/s;

w_0—feed flow velocity, m/s;

z—distance from the filter inlet, m;

$Z = z/z_0$—dimensionless distance from the filter inlet;

Γ—specific cake deposit (cake mass per unit membrane shell surface area), kg/m^2;

$\gamma = \Gamma s/c_0$—dimensionless specific cake deposit;

μ—liquid viscosity, Pa s;

ξ—ratio of permeate flow rate to suspension flow rate at filter inlet;

ψ_1, ψ_2——vectors of phenomenological parameters.

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