## MODEL OF STEADY-STATE ULTRAFILTRATION OF COLLOIDAL SUSPENSION WITH FORMATION OF NON-NEWTONIAN CONCENTRATION POLARIZATION LAYER AND COMPRESSIBLE DEPOSIT UNDER LAMINAR CROSS-FLOW

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#### ABSTRACT

A model for description of local filtrate flux and local solids concentration distribution in the filter channel at the steady-state of the cross-flow membrane filtration of colloidal suspension is proposed.

The model accounts for rheological and compression-permeability properties of the filtered material:

$$\dot{\gamma} = \dot{\gamma}(c,\tau) \tag{a}$$

where  $\dot{\gamma}$  is the shear rate, c is the solids concentration, and  $\tau$  is the shear stress,

$$\Pi = \Pi(c) \tag{b}$$

where  $\Pi$  is the osmotic pressure or solid pressure, and

$$k = k(c) \tag{C}$$

where k is the permeability.

Analytical solution of a system of mass and pressure balance equations under the assumption of a thin concentration polarization (CP) layer yields the local filtrate flux distribution along the membrane, J(x)

$$J(x)^{-3} = J(0)^{-3} + 3c_0\mu_f^2 x / (2M(c_{sg}, \tau_w))$$
(d)

where J(0) is the filtrate flux across the membrane in the absence of the concentration polarization,  $c_0$  is the solids concentration in the bulk,  $\mu_f$  is the filtrate viscosity, *x* is the distance from the entrance to the filter channel, and  $M(c_{sg}, \tau_w)$  is the integral characteristics of the filtered material, which is defined as

$$M(c_{sg}, \tau_w) = \int_{c_0}^{c_{sg}} (c - c_0) k(c) \frac{\partial \Pi(c)}{\partial c} \left[ \int_{c}^{c_{sg}} \dot{\gamma}(c, \tau_w) k(c) \frac{\partial \Pi(c)}{\partial c} dc \right] dc$$
(e)

(where  $\tau_w$  is the applied wall shear stress, and  $c_{sg}$  is the sol-gel transition concentration, i.e. the maximal solids concentration in the CP layer).

It should be noted that according to Eqs. (d) and (e), filtrate flux is independent of deposit properties (i.e., there is no need to define or measure neither  $\dot{y}$ ,  $\Pi$  nor k at  $c > c_{sg}$  for the local filtrate flux calculation). However, Eqs. (b) & (c) for  $c > c_{sg}$  can be used to predict the local solids concentration in the deposit c(x,z) for the found J(x) with the help of the integral form of Darcy's equation (where z is the normal distance to the membrane surface).

#### **KEYWORDS**

Model, membrane filtration, cross-flow filtration, concentration polarization, deposit, local properties, non-Newtonian, compression-permeability.

### INTRODUCTION

The present paper is devoted to the modelling of the steady-state cross-flow filtration of Brownian suspension that results in formation of concentration polarization (CP) layer, which can exhibit non-Newtonian rheological properties (e.g., shear thinning), that is optionally followed by formation of compressible deposit (a gel having a yield stress that exceeds an applied wall shear stress in the filter channel).

The model development was started in our previous work (Loginov et al., 2021), however, this paper only described filtration under the limiting flux conditions in a short filtration (when the fully gel covers the membrane surface and the transmembrane pressure is constant along the filter channel).

In the present paper we provide a full model, which accounts for the transmembrane pressure distribution along the filter channel, is applicable for sub-critical filtration conditions (before the gel formation), and accounts for the gel appearance (between the critical and limiting filtration conditions). Here, for readers' convenience, we recall the model assumptions and the model development, which are partially published in (Loginov et al., 2021).

### MODEL DEVELOPMENT

Following equations are obtained for the case of cross-flow in a cylindrical filter channel. Fig. 1 presents a scheme used for the model derivation.



Fig. 1. Filtration of a feed suspension with particle volume fraction  $\phi = \phi_0$  through a fully retentive membrane results in formation of filtrate ( $\phi = 0$ ) and CP layer with the thickness  $h_{CP}(x)$  and local concentration  $\phi(x, z) > \phi_0$  followed by formation of a deposit with the thickness  $h_d(x)$  and local concentration  $\phi(x, z) > \phi_{sg}$ , where  $\phi_{sg}$  is a concentration of sol-gel transition. Local particle concentration on the membrane surface is denoted as  $\phi_w(x)$ .

The current model derivation follows general approach and uses some of assumptions proposed by Gaddis (1992) and Bacchin et al. (2002).

The model is obtained for the case, when thicknesses of CP layer and deposit are negligible as compared to the filter channel radius. Consequently, it is assumed that tangential gradients of different parameters in the CP layer are negligible as compared to respective normal gradients.

At a steady state, there is no additional particle accumulation inside the filter channel (neither the particle redistribution between the flowing CP layer and the immobile deposit). The total flux of particles across a transverse section of the filter channel at any distance from the entrance *x* is equal to the total particle flux at the entrance of the filter channel  $Q_0\phi_0$ , where  $Q_0$  is the average tangential flow rate at the entrance to the filter channel (at *x* = 0), and  $\phi_0$  is the particle volume fraction in feed suspension. Filtration results in a gradual concentrating of particles in the CP layer, where  $\phi(x, z) > \phi_0$ . Following the idea of Gaddis (1992) and Bacchin et al. (2002), the total particle flux across a transverse section of the filter channel at any distance from the entrance *x* is presented as a sum of two virtual components: (1) tangential transport of excess particles in the flowing CP layer and (2) transport of suspension having the initial particle volume fraction  $\phi_0$ . Therefore, the total transport of particles through any cross section of the filter channel can be written as

$$Q_0 \phi_0 = 2\pi R \int_{h_d(x)}^{h_{\rm CP}(x)+h_d(x)} u(x,z) (\phi(x,z) - \phi_0) \,\mathrm{d}z + Q(x)\phi_0 \tag{1}$$

where u(x, z) is the local tangential flow rate of liquid and particles, Q(x) is the average tangential flow rate through the entire cross section of the channel (including the CP layer), and *R* is the inner radius of the filter channel. The integration in Eq. (1) is done over the local thickness of CP layer  $h_{CP}(x)$ , and the lower integration limit  $h_d(x)$  is the local deposit thickness (note that  $h_d(x) = 0$ , if the deposit is absent at given *x*).

Using the definition of shear rate  $\dot{Y}$ , the local tangential flow rate u(x, z) can be presented as

$$u(x,z) = \int_{h_d(x)}^{z} \dot{\gamma}(x,z) \,\mathrm{d}z \tag{2}$$

where  $\dot{\gamma}(x,z)$  is the normal component of local shear rate (as it is stated before, its tangential component is neglected for the case of thin CP layer). It is assumed that there is no liquid slip at the CP–solid (membrane or deposit) boundary (at  $z = h_d(x)$ ).

Further description of particle transport in the CP layer requires the knowledge of its local rheological properties (i.e., shear stress dependence of shear rate at a given particle concentration):

$$\dot{\gamma} = \dot{\gamma}(\phi, \tau) \tag{3}$$

As soon as the CP layer thickness is assumed to be negligible as compared to the channel radius R, it can be assumed that the shear stress in the CP layer is practically constant at given x and equal to the local wall shear stress  $\tau_w(x)$ 

$$\tau(x,z) \approx \tau_w(x) = -\frac{R}{2} \cdot \frac{\mathrm{d}P(x)}{\mathrm{d}x} \tag{4}$$

where P(x) is the local pressure in the filter channel. In the case of laminar flow in cylindrical filter channel, the local pressure gradient dP(x)/dx is related with the average tangential flow rate Q(x) as

$$\frac{\mathrm{d}P(x)}{\mathrm{d}x} = -\frac{8\mu_b}{\pi R^4}Q(x) \tag{5a}$$

where  $\mu_b$  is the viscosity of suspension in the bulk. In the case of turbulent cross-flow the relation is

$$\frac{\mathrm{d}P(x)}{\mathrm{d}x} = -\frac{0.009\,\mu_b^{0.25}\,\rho_b^{1.75}}{R^{4.75}}\,Q(x)^{1.75} \tag{5b}$$

where  $\rho_b$  is the density of bulk suspension. It is assumed that Eqs. (5a) and (5b) are valid despite of the presence of thin CP layer.

The value of Q(x) decreases with x because of filtrate formation

$$\frac{\mathrm{d}Q(x)}{\mathrm{d}x} = -2\pi R J(x) \tag{6}$$

where J(x) is the local filtrate flux. However, for the usual case of relatively slow filtration (i.e., when the average filtrate flux over the filter channel length  $\overline{I}(L)$  is relatively low,  $\overline{I}(L) \ll Q_0$ ), for the sake of evaluation of  $\tau_w$ , it can be assumed that the value of local flow rate is practically unaffected by filtrate formation,  $Q(x) \approx Q_0$ . Hence, the applied pressure gradient is practically constant and equal to

$$\frac{\mathrm{d}P(x)}{\mathrm{d}x} \approx -\frac{8\mu_b}{\pi R^4} Q_0 \tag{7a}$$

in the case of laminar cross-flow and to

$$\frac{\mathrm{d}P(x)}{\mathrm{d}x} \approx -\frac{0.009\,\mu_b^{0.25}\rho_b^{1.75}}{R^{4.75}}\,Q_0^{1.75} \tag{7b}$$

in the case of turbulent cross-flow. Thus, as it follows from Eqs. (4), (7), the value of the shear stress in a thin CP layer is practically constant along the filter channel and equal to

$$\tau \approx 4\mu_b Q_0 / (\pi R^3) \tag{8a}$$

in the case of laminar cross-flow and to

$$\tau \approx 0.0045 \mu_b^{0.25} \rho_b^{1.75} Q_0^{1.75} R^{-3.75}$$
(8a)

According to Eqs. (8), the value of shear stress in a thin CP layer is an operating parameter depending on  $Q_0$ . Hence, the modeling of filtration requires to determine the local rheological properties in this layer (defined via Eq. (3)) for a single value of  $\tau$  (defined via one of Eqs. (8)). For a given value of  $\tau$ , known (either modeled or experimentally measured) dependency  $\dot{\gamma}(\phi, \tau)$ , can be introduced into Eq. (2)

$$u(x,z) = \int_{h_d(x)}^{z} \dot{\gamma}(\phi,\tau) \,\mathrm{d}z \tag{9}$$

where  $\phi = \phi(x, z)$  is unknown.

An extension of the usual assumption of conventional filtration theory (filtrate flux does not vary across the filter cake, i.e.,  $J \neq J(z)$ ) for the case of cross-flow filtration with CP layer formation allows to apply Darcy equation in order to relate the local filtrate flux with local filtration properties of CP layer and deposit:

$$\mu_f J(x) = -k(x,z) \frac{\mathrm{d}\Pi(x,z)}{\mathrm{d}z} \tag{10}$$

where  $\mu_f$  is the filtrate viscosity, k(x, z) is the local hydraulic permeability and  $\Pi(x, z)$  is the local osmotic pressure in the CP layer or the solid pressure in the deposit. Eq. (10) is applied for description of deposit (immobile) and CP layer (flowing): in the current model, usual presentation of filtrate percolation across the CP layer as a backward diffusion of particles (Gaddis (1992), Bacchin et al. (2002)) is replaced by Darcy equation, where osmotic pressure substitutes for solid pressure. According to the conventional filtration theory, for a given system of particles, k and  $\Pi$  are material properties that depend only on the local particle volume fraction. Further in this section, we assume that the particle volume fraction dependencies  $k(\phi)$  and  $\Pi(\phi)$  are not affected by the tangential flow of CP layer or tangential stress applied to the deposit (in other words, shear-induced diffusion coefficient is negligible, or hydrodynamic component of osmotic pressure is negligible; the model extension for the case of non-negligible shear-induced effects is provided in the Appendix B). Then, Eq. (10) can be rewritten as

$$\mu_f J(x) = -k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} \cdot \frac{\mathrm{d}\phi}{\mathrm{d}z}$$
(11)

where  $\phi = \phi(x, z)$  is unknown.

The following model derivation is equivalent to that done by Bacchin et al. (2002). Expression of dz from Eq. (11)

$$dz = -\frac{k(\phi)}{\mu_f J(x)} \cdot \frac{\partial \Pi(\phi)}{\partial \phi} d\phi$$
(12)

with its following substitution into Eq. (9) yields

$$u(x,z) = -\frac{1}{\mu_f J(x)} \int_{h_d(x)}^{z} \dot{\gamma}(\phi,\tau) \cdot k(\phi) \cdot \frac{\partial \Pi(\phi)}{\partial \phi} d\phi$$
(13)

Substitution of Eqs. (12) and (13) into Eq. (1) yields

$$Q_0\phi_0 = \frac{2\pi R}{\mu_f^2 J(x)^2} \int_{h_d(x)}^{h_d(x)+h_{CP}(x)} (\phi - \phi_0)k(\phi) \frac{\partial\Pi(\phi)}{\partial\phi} \left[ \int_{h_d(x)}^z \dot{\gamma}(\phi,\tau) k(\phi) \frac{\partial\Pi(\phi)}{\partial\phi} d\phi \right] d\phi + Q(x)\phi_0 \quad (14)$$

The upper limit of the first integral corresponds to the boundary between the CP layer and the bulk suspension, where  $\phi = \phi_0$  (it is assumed that relatively low filtrate flux has no significant influence on the bulk concentration). The lower integration limit  $h_d(x)$  corresponds to the "lower" boundary of CP layer. Therefore, when the deposit is absent, it corresponds to the membrane surface position, where  $\phi = \phi_w(x)$ . When the deposit is present on the membrane, it corresponds to the CP layer – deposit surface position, where  $\phi = \phi_{sg}$ . The particle concentration in the point of sol-gel transition  $\phi_{sg}$  does not depend on x but depends on  $\tau$  (i.e., an element of external membrane fouling flows, so, belongs sol and not to gel, if its local particle concentration corresponds to the yield stress, which is lower that the wall shear stress  $\tau_w$ ). Hence, Eq. (14) can be rewritten as

$$Q(x) = Q_0 - \frac{2\pi R}{\phi_0 \mu_f^2 J(x)^2} \int_{\phi_0}^{\phi_w(x)} (\phi - \phi_0) k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} \left[ \int_{\phi}^{\phi_w(x)} \dot{\gamma}(\phi, \tau) k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} d\phi \right] d\phi \quad (15a)$$

for values of x corresponding to the membrane covered by flowing CP layer, or

$$Q(x) = Q_0 - \frac{2\pi R}{\phi_0 \mu_f^2 J(x)^2} \int_{\phi_0}^{\phi_{sg}} (\phi - \phi_0) k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} \left[ \int_{\phi}^{\phi_{sg}} \dot{\gamma}(\phi, \tau) \, k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} \, \mathrm{d}\phi \right] \mathrm{d}\phi \quad (15b)$$

for values of x corresponding to the membrane covered by solid deposit.

For any point on the membrane surface, the local value of  $\phi_w(x)$  can be obtained from the expression for the pressure distribution between the CP layer, the deposit (if it is present on the membrane surface) and the membrane:

$$\mu_f R_m J(x) = P(x) - \Pi(\phi_w(x)) - P_f(x)$$
(16)

where  $R_m$  is the hydraulic resistance of filtration membrane,  $\Pi(\phi_w(x))$  is the local pressure drop across the deposit and the CP layer, and  $P_f(x)$  is the pressure at filtrate side. In the present model, is assumed that  $P_f(x)$  is not controlled (as it is done, for example, in the uniform transmembrane pressure filtration units); therefore,  $P_f(x) = 0$ . The variation of P(x) along the filter channel can be found form

$$P(x) = P_0 - \frac{2\tau}{R}x\tag{17}$$

with the help of Eqs. (7a) or (7b) for the case of laminar or turbulent cross-flow, respectively.

Eqs. (6), (15a), (15b), (16) and (17) are main model equations that are sufficient for calculation of local flux J(x) at different filtration conditions. For the sake of convenience, all assumptions used for their derivation are summarized in the Appendix A.

### **MODEL SOLUTION**

For a given suspension (given value of  $\phi_0$  and given dependencies  $k(\phi)$ ,  $\Pi(\phi)$  and  $\dot{\gamma}(\phi,\tau)$ ) subjected to a given shear stress  $\tau$  (that determines  $\phi_{sg}$  and  $\dot{\gamma}(\phi,\tau)$ ), the value of double integral in Eq. (15a) if a function, which depends only on  $\phi_w$  (which varies with *x*). The value of the double integral in Eq. (15b) is a constant (which depends only on  $\phi_{sg}$ ). Therefore, Eqs. (15a) and (15b), can be rewritten as

$$Q(x) = Q_0 - \frac{2\pi R}{\phi_0 \mu_f^2} \cdot \frac{M(\phi_w(x), \tau)}{J(x)^2}$$
(18a)

$$Q(x) = Q_0 - \frac{2\pi R}{\phi_0 \mu_f^2} \cdot \frac{M(\phi_{sg}, \tau)}{J(x)^2}$$
(18b)

where *M* is the material properties-dependent function of concertation  $\phi$  defined as

$$M(\phi_{w},\tau) = \int_{\phi_{0}}^{\phi_{w}} (\phi - \phi_{0}) k(\phi) \frac{\partial \pi(\phi)}{\partial \phi} \left[ \int_{\phi}^{\phi_{w}} \dot{\gamma}(\phi,\tau) k(\phi) \frac{\partial \pi(\phi)}{\partial \phi} d\phi \right] d\phi$$
(19a)

and  $M(\phi_{sg}, \tau)$  is the said constant

$$M(\phi_{sg},\tau) = \int_{\phi_0}^{\phi_{sg}} (\phi - \phi_0) k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} \left[ \int_{\phi}^{\phi_{sg}} \dot{\gamma}(\phi,\tau) k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} d\phi \right] d\phi$$
(19b)

The system of Eqs. (6), (16), (17) and (18a) is sufficient for the modeling of crossflow filtration without deposit formation. It yields an ordinary differential equation

$$\frac{\mathrm{d}\phi_{w}}{\mathrm{d}x} = \frac{\frac{\phi_{0}}{\mu_{f}R_{m}^{3}} \left[P_{0} - \frac{2\tau}{R}x - \Pi(\phi_{w})\right]^{4} - \frac{4\tau}{R}M(\phi_{w},\tau)}{\left[P_{0} - \frac{2\tau}{R}x - \Pi(\phi_{w})\right] \frac{\mathrm{d}M(\phi_{w},\tau)}{\mathrm{d}\phi_{w}} + 2M(\phi_{w})\frac{\mathrm{d}\Pi(\phi_{w})}{\mathrm{d}\phi_{w}}}$$
(20)

Numerical solution of Eq. (20) with the initial condition  $\phi_w(0) = \phi_0$  yields  $\phi_w(x)$  distribution for the part of membrane, where  $\phi_w(x) < \phi_{sg}$  is satisfied. Then, J(x) dependency can be calculated using Eq. (16) and (17) from obtained  $\phi_w(x)$  dependency using known  $\Pi(\phi)$  dependency.

At certain filtration conditions, a deposit appears at a certain critical distance from the entrance to the filter channel  $x_{cr1}$ , where  $\phi_w(x_{cr1}) = \phi_{sg}$  is satisfied. For the downstream part of the filter channel (with  $x \ge x_{cr1}$ ), filtration is described by the system of Eqs. (6) and (18b), having the following analytical solution

$$J(x) = \left[J^{-3}(x_{cr1}) + \frac{3\phi_0\mu_f^2}{2M(\phi_{sg},\tau)}(x - x_{cr1})\right]^{-\frac{1}{3}}$$
(21)

where values of  $J(x_{cr1})$  and  $x_{cr1}$  are determined from the solution of Eq. (20). The  $\phi_w(x)$  dependency (local deposit concentration on the membrane surface) can be further calculated from  $\phi_w(x)$  and  $\Pi(\phi)$  dependencies with the help of Eqs. (16) and (17).

In the case of sufficiently long filter channel, decreasing of J(x) and P(x) with x can reduce the value of  $\phi_w(x)$  below  $\phi_{sg}$  at  $x = x_{cr2}$  (the second critical distance from the entrance). For the downstream part of the filter channel (at  $x > x_{cr2}$ ) filtration will be again described by Eq. (20) with  $\phi_w(x_{cr2}) = \phi_{sg}$  as an initial condition.

The areas of applicability of Eqs (20) and (21) are summarized in Fig. 2.



Fig. 2. Areas of applicability of model equations.

Finally, the local particle concentration distribution across the CP layer and the deposit  $\phi(x, z)$  can be calculated at any value of x with the help of obtained J(x) and  $\phi_w(x)$  dependencies, which are introduced into the integral form of Eq. (12):

$$z(x,\phi) = \frac{1}{\mu_f J(x)} \int_{\phi}^{\phi_W(x)} k(\phi) \frac{\partial \Pi(\phi)}{\partial \phi} d\phi$$
(22)

Eq. (22) is also used for the calculation of local deposit thickness  $h_d(x) = z(x, \phi_{sg})$  and local CP layer thickness  $h_{CP}(x) = z(x, \phi_0) - z(x, \phi_{sg})$ .

#### MODEL RESULTS AND DISCUSSION

## Impact of material and rheological properties on filtration, new measure of the filterability

For a filter channel part starting at the cell entrance and having a length  $l < x_{cr1}$  combination of Eqs. (6) and (18a) yields the following expression for the length-averaged filtrate flux  $\overline{I}(l)$ 

$$\overline{J}(l) = \frac{1}{\phi_0 \mu_f^2} \cdot \frac{M(\phi_w(l), \tau)}{J(l)^2 l}$$
<sup>(23)</sup>

Besides of evident dependencies (filtration rate decreases with increasing of bulk concentration  $\phi_0$  and filtrate viscosity  $\mu_f$ ), Eq. (23) also suggests that filtration depends on  $M(\phi_{w}, \tau)$ , and that filtrate flux is higher, when  $M(\phi_{w}, \tau)$  is higher. As it follows from Eq. (21), these dependencies also hold for the filter channel part covered by deposit: in particular, filtration rate is higher at higher  $M(\phi_{sg}, \tau)$ . It can be concluded that, irrespective of  $\phi_w(x)$  and J(x), filtration is faster, when  $M(\phi, \tau)$  is higher in a particle concentration range  $[\phi_0, \phi_{sg}]$ . Therefore, M is a measure of filterability of suspension in a cross-flow filtration experiment.

According to Eqs. (19a) and (19b), for a given  $\phi$ , the value of  $M(\phi, \tau)$  increases with the increasing of permeability  $k(\phi)$  and decreasing of compressibility and viscosity (increasing of  $\partial \Pi(\phi)/\partial \phi$  and  $\dot{\gamma}(\phi, \tau)$ , respectively). These are expectable correlations of filterability with material properties of a filtered sample. In addition, as soon as all multipliers of integrands in Eqs. (19a) and (19b) are positive,  $M(\phi, \tau)$  is an increasing function of  $\phi$ .

According to Eq. (19a), for a CP layer and a deposit with Newtonian rheological properties (that is  $\dot{\gamma}(\phi, \tau) = \tau/\mu(\phi)$  and  $\phi_{sg} \neq f(\tau)$ ),  $M(\phi, \tau)$  is directly proportional to  $\tau$ . However, for non-Newtonian CP layer and deposit (that exhibit shear-thinning and yield stress) the increasing of  $M(\phi, \tau)$  with  $\tau$  is sublinear, and the maximal value of  $M(\phi_{sg}, \tau)$  attained in the experiment increases with  $\tau$  since  $\phi_{sg} = f(\tau)$ .

Detailed discussion of the influence of operating parameters and sample material properties on filtration kinetics requires assumed functional forms of material properties (either  $\Pi(\phi)$ ,  $k(\phi)$  and  $\dot{\gamma}(\phi,\tau)$  or  $M(\phi,\tau)$ ) for a particle concentration range  $[\phi_0, \phi_{sg}(\tau_{max})]$ , where  $\tau_{max}$  is a maximal value of wall stress expected in the experiment. It is worth noting that according to the model, sample properties at  $\phi > \phi_{sg}(\tau_{max})$  (i.e., deposit properties) do not influence the steady-state kinetics of cross-flow filtration (though they can influence a solute rejection during the filtration and the membrane cleaning after the filtration).

# Inverse and direct problems solution: characterization of cross-flow filterability and scaling of cross-flow filtration

As it follows from the previous section, the cross-flow filtration rate at different operating conditions can be predicted with the help of Eqs. (19a) and (20) or (19b) and (21) together with Eqs. (16) and (17) for suspension with known material properties  $\Pi(\phi)$ ,  $k(\phi)$  and  $\dot{\gamma}(\phi, \tau)$ , which completely characterize the filterability. Experimental determination of  $\Pi(\phi)$ ,  $k(\phi)$  and  $\dot{\gamma}(\phi, \tau)$  for different suspensions is a feasible but time consuming routine. Moreover, these dependencies must be determined in the concentration range  $[\phi_0, \phi_{sg}]$ , which generally corresponds to low osmotic pressure range (therefore, particular methods are suggested for the  $\Pi(\phi)$  and  $k(\phi)$  determination).

Using the model, it is possible to characterize filterability of suspension during a cross-flow filtration in a different way, by determining *M* from cross-flow filtration data. New method for determination of filterability can be introduced as follows. Material properties of a studied sample can be presented and the model equations can be rewritten for a new variable, which is local osmotic (or solid) pressure in the CP layer or deposit  $\Pi$  (instead of the used before local particle concentration  $\phi(x, z)$ ):  $\phi(\Pi)$ ,  $k(\Pi)$  and  $\dot{\gamma}(\Pi, \tau)$ . The pressure varies across the CP layer and the deposit  $\Pi = \Pi(x, z)$ , it changes from  $\Pi = \Pi_0$  at the bulk – CP layer interface to  $\Pi_w$  at the membrane surface, and it is equal to  $\Pi_{sg}$  at the deposit surface. There are following relations of these values with previously defined limiting values of  $\phi$ :  $\Pi_0 = \Pi(\phi_0)$  (constant),  $\Pi_w = \Pi(\phi_w)$  (varies with *x*), and  $\Pi_{sg} = \Pi(\phi_{sg})$  (does not depend on position but depends on  $\tau$ , because  $\phi_{sg}$  depends on  $\tau$ ). Following the same routine as it was used for the derivation of Eqs. (15a) and (15b), the filterability *M* can be now presented as

$$M(\Pi_{w},\tau) = \int_{\Pi_{0}}^{\Pi_{w}} (\phi(\Pi) - \phi(\Pi_{0})) k(\Pi) \left[ \int_{\Pi}^{\Pi_{w}} \dot{\gamma}(\Pi,\tau) k(\Pi) d\Pi \right] d\Pi$$
(24a)

Likewise, Eq. (18a), Eq. (24a) is applicable when  $\Pi_w(x) < \Pi_{sg}$ , while in the point of sol-gel transition the value of *M* is

$$M(\Pi_{sg},\tau) = \int_{\Pi_0}^{\Pi_{sg}} (\phi(\Pi) - \phi(\Pi_0)) k(\Pi) \left[ \int_{\Pi}^{\Pi_{sg}} \dot{\gamma}(\Pi,\tau) \cdot k(\Pi) \mathrm{d}\Pi \right] \mathrm{d}\Pi$$
(24b)

Using new definition of M, Eq. (23) can be simply rewritten as

$$\bar{J}(x) = \frac{1}{\phi_0 \mu_f^2} \cdot \frac{M(\Pi_w(x), \tau)}{J(x)^2 x}$$
(25)

with the same condition of applicability  $x < x_{cr^1}$ . The following rewritten form of Eq. (21)

$$J(x) = \left(J^{-3}(x_{cr1}) + \frac{3\phi_0\mu_f^2}{2M(\Pi_{sg},\tau)}(x - x_{cr1})\right)^{-\frac{1}{3}}$$
(26)

describes local filtration flux for  $x \ge x_{cr1}$ . Finally, Eq. (16) can be rewritten as

$$\mu_f R_m J(x) = P(x) - \Pi_w(x) - P_f(x)$$
(27)

If the local flux distribution along the filter channel J(x) is measured in a cross-flow filtration experiment at given values of  $P_0$  and  $\tau$ , then the distribution of M along the channel (i.e.,  $M(x)/(\phi_0^{\mu_f^2})$ ) can be obtained via the data fitting with the help of

Eqs. (25) and (26). Also,  $\Pi_w(x)$  can be calculated with the help of Eq. (27) with for known dependencies P(x) and  $P_f(x)$ . Hence, the dependency  $M(\Pi, \tau)/(\phi_0 \mu_f^2)$  on  $\Pi$  can be constructed for the used value of  $\tau$  (for a certain range of  $\Pi$ , which can be enlarged via application of different values of  $P_0$  at the same value of  $\tau$ ). This dependency is a measure of filterability of a studied sample in a cross-flow filtration for the given value of  $\tau$  and for any set of other operating parameters (applied pressure  $P_0$ , filter channels' length *I* and internal radius *R*, membrane resistance  $R_m$ ).

When  $M(\Pi, \tau)/(\phi_0 \mu_f^2)$  is determined, it can be used for the calculation of filtration rate J(x) for  $x < x_{cr1}$  with the help of the following equation

$$\frac{\mathrm{d}\Pi_{w}}{\mathrm{d}x} = \frac{\phi_{0}\,\mu_{f}^{3}R_{m}\left[P_{0} - \frac{2\tau}{R}\,x - \Pi_{w}\right]^{4} - \frac{4\tau}{R}M(\Pi_{w})}{\mu_{f}R_{m}\left[P_{0} - \frac{2\tau}{R}\,x - \Pi_{w}\right]\frac{\mathrm{d}M(\Pi_{w})}{\mathrm{d}\Pi_{w}} + 2M(\Pi_{w})}$$
(28)

which is derived using the same routine as it was used for Eq. (20). Numeric solution of Eq. (28) with initial condition  $\Pi_{W}(0) = \Pi_{0}$  and application of Eq. (27) yields J(x) and the value of  $x_{cr1}$  at the point with  $\Pi_{W}(x_{cr1}) = \Pi_{sg}$  (deposit formation). Following filtration is described by Eq. (26) as soon as  $M(\Pi_{sg}, \tau)/(\phi_{0}\mu_{f}^{2})$  is known from the aforementioned experiment.

### APPENDIX A. MODEL ASSUMPTIONS

The following assumptions are used for the model derivation.

- 1. Steady-state filtration is considered.
- 2. Flow is fully developed (at least in the CP layer).
- 3. Filtration membrane is a continuum with constant hydraulic resistance.
- 4. Membrane is fully retentive.

5. Pore blocking and other "pore fouling" phenomena are not considered. If they are present, clean membrane resistance is replaced by fouled membrane resistance (that does not comprise the CP and deposit resistance).

6. Thickness of CP layer (and that of the deposit) is negligible as compared to the filter channel dimensions.

7. In the CP layer, tangential gradients of cross-flow rate and particle concentration are negligible as compared to respective normal gradients.

8. Local values of material properties  $(k(\phi) \text{ and } \Pi(\phi))$  are not affected by tangential flow.

9. In CP layer and deposit, dependencies  $k(\phi)$ ,  $\Pi(\phi)$  and  $\dot{\gamma}(\phi)$  are equivalent to those for suspensions having same particle concentration (e.g., particle classification or other phenomena potentially influencing granular of chemical composition of CP layer and deposit are not considered).

10. The shear stress in CP layer is constant and equal to the wall shear stress.

11. Poiseuille relationship for friction factor is valid despite of the presence of CP layer and deposit.

12. Filtrate flux is negligible as compared to cross-flow flux; therefore, tangential pressure gradient is practically constant over the filter channel (including the CP layer).

13. For given *x*, filtrate flux is constant across the CP layer, deposit and membrane.

14. Filtrate permeation in CP layer can be described by Darcy equation for granular media.

# APPENDIX B. MODEL EXTENSION FOR THE CASE OF NON-NEGLIGIBLE SHEAR-INDUCED EFFECTS

When the modelling of cross-flow filtration is based on consideration of convectiondiffusion balance, the influence of local shear rate on the particle diffusion away from the membrane is accounted with the help of the coefficient of shear-induced diffusion. In the current paper, an equivalent approach (which is usually applied in conventional filtration-consolidation theory) is used for description of local material balance in cross-flow filtration: the local flux towards the membrane is proportional to the hydraulic pressure gradient (which is equal to the osmotic or solid pressure gradient with the opposite sign) and impeded by the local hydraulic resistance of the CP layer (deposit):

$$\mu_f J(x) = -k(x,z) \frac{\mathrm{d}\Pi(x,z)}{\mathrm{d}z} \tag{B1}$$

where J(x) is the filtrate flux,  $\mu_f$  is the filtrate viscosity, k(x, z) is the local hydraulic permeability and  $\Pi(x, z)$  is the local osmotic pressure. The local osmotic pressure is a function of particle concentration  $\phi(x, z)$ , however, it can also depend on the local shear rate  $\dot{\gamma}(x, z)$ , and the influence of the shear rate on the local osmotic pressure is equivalent to the influence of the shear rate on the coefficient of diffusion. Therefore, the influence of the local shear rate in the CP layer on the cross-flow filtration rate can be accounted as

$$\mu_f J(x) = -k(x,z) \frac{\mathrm{d}\Pi(\phi,\dot{\gamma})}{\mathrm{d}z} \tag{B2}$$

Both  $\phi$  and  $\dot{\gamma}$  depends on x and z, while k only depends on the systems concentration  $k = k(\phi)$ . However,  $\dot{\gamma} = \dot{\gamma}(\phi, \tau)$ , and Eq. (B2) can be rewritten as

$$\mu_f J(x) = -k(x, z) \frac{\mathrm{d}\Pi(\phi)}{\mathrm{d}z} = -k(\phi) \frac{\partial\Pi(\phi)}{\partial\phi} \cdot \frac{\mathrm{d}\phi}{\mathrm{d}z}$$
(B3)

where dependence of osmotic pressure on shear rate is implicit. As soon as Eq. (B3) is equivalent to Eq. (11), the following model derivation for the case of non-negligible shear enhanced diffusion results in exactly the same equations Eqs. (12) - (23).

### ABBREVIATIONS

- CP concentration polarization
- sg sol-gel transition

## NOMENCLATURE

- *h*<sub>CP</sub> local thickness of CP layer (m)
- *h*<sub>d</sub> local thickness of deposit (m)
- J local filtrate flux (m·s<sup>-1</sup>)
- $\overline{I}$  average filtrate flux over a part of the filter channel (m·s<sup>-1</sup>)
- k local permeability (m<sup>2</sup>)
- L length of filter channel (m)
- *I* length of a part of the filter channel (m)
- *M* filterability, material function that governs cross-flow filtration ( $m^{4} \cdot Pa^{2} \cdot s^{-1}$ )
- *P* total pressure in the filtration channel (Pa)
- $P_0$  total pressure at the entrance to the filter channel (Pa)
- *P<sub>f</sub>* pressure at filtrate side (Pa)
- Q average tangential flow rate in the filter channel (m<sup>3</sup>·s<sup>-1</sup>)
- $Q_0$  average tangential flow rate at the entrance to the filter channel (m<sup>3</sup>·s<sup>-1</sup>)
- *R* inner radius of filter channel (m)
- $R_m$  membrane resistance (m<sup>-1</sup>)
- *u* local tangential flow rate  $(m \cdot s^{-1})$
- x axial distance from the entrance of the filter channel (m)

 $x_{cr1}$  first critical distance, axial distance from the entrance of the filter channel, where the deposit appears on the membrane surface (m)

 $x_{cr2}$  second critical distance, axial distance from the entrance of the filter channel, where the deposit disappears from the membrane surface (m)

*z* normal distance from the membrane surface (m)

## **GREEK LETTERS**

- $\dot{\gamma}$  local shear rate (s<sup>-1</sup>)
- $\mu_b$  viscosity of bulk suspension in the filter channel (Pa·s)
- $\mu_f$  filtrate viscosity (Pa·s)
- Π local osmotic pressure of particles in CP layer or solid pressure in deposit (Pa)
- $\Pi_0$  osmotic pressure in feed suspension (Pa)
- $\Pi_{sg}$  osmotic pressure in point of sol-gel transition (Pa)
- $\Pi_w$  osmotic pressure (solid pressure) on the membrane surface (Pa)
- $\rho_b$  density of bulk suspension in the filter channel (kg·m<sup>3</sup>)
- *τ* shear stress (Pa)
- $\tau_w$  wall shear stress (Pa)

- *φ* particle volume fraction (dimensionless)
- $\phi_0$  particle volume fraction in feed suspension (dimensionless)
- $\phi_{sg}$  particle volume fraction in point of sol-gel transition (dimensionless)
- $\phi_w$  particle volume fraction on the membrane surface (dimensionless)

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