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Generalized criterion for the onset of particle deposition in CFMF via DOTM – Modeling and Experimental Validation

Farhad Zamani^{a,b}, Filicia Wicaksana^{b,c}, A.H. Taheri^{a,b}, Adrian W.K. Law^{a,b}, A.G. Fane^{a,b},
W.B. Krantz^{b,d}

^a School of Civil and Environmental Engineering, Nanyang Technological University 50
Nanyang Avenue, Singapore 639798, Singapore

^b Singapore Membrane Technology Centre, Nanyang Environment and Water Resource
Institute, 1 Cleantech Loop, Clean Tech One, Singapore 637141, Singapore

^c Department of Chemical and Materials Engineering, the University of Auckland, Auckland,
New Zealand 1142

^d Department of Chemical & Biological Engineering, University of Colorado, Boulder, CO
80309-0424, USA

Abstract:

The concept of a critical permeation flux for the onset of particle deposition in crossflow microfiltration (CFMF) is well-established. However, the critical flux is known to be a function of process parameters such as the particle size, bulk concentration and crossflow velocity. The critical modified Peclet number (Pe_{crit}) is explored here as a generalized criterion for the onset of particle deposition that incorporates the effects of these process parameters as well as axial position along the membrane. Proper determination of Pe_{crit} requires accurate prediction of the concentration polarization boundary layer thickness δ_c and shear-induced diffusion coefficient D_s . The classical Lvque model is adapted to allow for the effect of the permeation flux on the velocity profile. Moreover, the assumptions of a constant concentration at the membrane surface c_w and constant D_s that have been made in prior studies are relaxed in an improved numerical solution to the convective diffusion equation that is used to predict δ_c and D_s . The critical permeation flux is determined from particle deposition data for 6 and 10 μm latex spheres taken via Direct Observation Through the Membrane (DOTM) characterization. A constant value of $Pe_{crit} = 4.00 \pm 0.08$ is found to characterize the effects of particle diameter, bulk concentration and crossflow velocity as well as axial position on the onset of particle deposition.

Keywords: Crossflow microfiltration (CFMF); Membrane Fouling; Critical Flux; Critical Peclet Number; Direct Observation Through the Membrane (DOTM)

1. Introduction

The concept of critical flux is now well established for crossflow microfiltration processes [1]. Above the critical flux, there is a net transport and deposition of particles (foulant) on the membrane when convection towards the membrane exceeds the diffusive back transport. The various back transport mechanisms, including Brownian diffusion and shear-induced diffusion, are known to be a function of the surface shear in the boundary layer, the relationship between flux and back diffusion points to a characteristic Peclet number (advective transport/diffusive transport) for crossflow microfiltration (CFMF). This approach has been discussed by Bacchin et al. [2, 3] who suggested that fouling conditions could be identified by a critical Pe number (ratio of critical flux to the mass transfer coefficient). In this paper we examine this approach using carefully measured particle deposition data for supra-micron particles in a well-defined flow channel.

The failure of the Brownian motion diffusivity mechanism to explain the supermicron particle deposition in CFMF [4, 5] resulted in introducing the shear-induced diffusivity (SID) mechanism to explain the back diffusion behavior of supermicron particles in shear flows [6]. Prior to that, it had been observed that supermicron particles experience lateral migration in shear flows [7]. Since then, the SID mechanism has been confirmed by observations such as increase in viscosity of a suspension by shearing it in a Couette flow [8]. In addition to SID, other mechanisms such as inertial lift [5, 9] and ‘flowing cakes’ (transport of particles along the surface) [10, 11] have also been proposed for supermicron particle deposition in CFMF [12]. Bacchin and coworkers [13] took surface interactions (e.g., electrokinetic effects) into account along with hydrodynamic effects to explain the behavior of colloidal particles near a membrane surface. For the models that consider the motion of a single particle owing to inertial lift and/or transport of the particle along the surface in the flow of a suspension, particle-particle interactions are ignored, which is not a realistic assumption for more concentrated suspensions. The concentration of rejected particles is usually very high close to the membrane surface in CFMF, which is the region of interest for studying the motion of the particles. On the other hand, the shear-induced diffusion coefficient, D_s in the SID models, is an all-inclusive parameter that is a measure of all mechanisms for diffusive particle motion (i.e., perpendicular to the direction of the crossflow). This diffusive motion can arise from mutually induced velocity fields, particle-particle interactions, lift forces and body forces [7, 14].

The Graetz [15] or Lvque [16] solutions that were originally used for the somewhat analogous heat transfer problem have been widely used to solve the mass transfer in laminar flow CFMF in order to determine the mass transfer coefficient, concentration boundary layer thickness and wall concentration [6, 17, 18]. The differences between the various prior studies that used the Lvque solution arise from the use of different diffusion coefficients. Numerical [19, 20] and computational fluid dynamics (CFD) [21] approaches have also been used to model the momentum and concentration boundary layers in membrane applications. The strengths and weaknesses of these models will be discussed in the next section.

In this paper a comprehensive simulation using a modified SID model [18] that is solved by the finite difference method has been employed to predict the wall concentration distribution along the membrane surface. It will be shown that in contrast to the Lvque solution the wall concentration is not the same at different locations along the membrane surface. In addition, based on the results from the DOTM technique the local critical fluxes (J_{crit}) at two different points near the inlet and exit of the crossflow channel have been measured. The experimental data from the DOTM technique combined with the results from the finite difference simulation support the existence of a critical modified Peclet number (Pe_{crit}) concept that has been suggested in prior studies [2, 3, 22].

2. Model development and numerical solution

CFMF can be considered to be the flow of an incompressible fluid between parallel flat walls, one of which is porous (i.e., the membrane). To model the mass transfer in this flow, both the velocity and concentration profiles need to be described in some way. Various approximations for the velocity profile have been used in different studies. The velocity profile corresponding to Poiseuille flow that does not consider any effects of the permeation flux through the wall has been used in the Graetz solution [15]. The Lvque solution [16] uses a linear approximation of the velocity distribution within a thermal boundary layer that can be employed in the same way for a concentration boundary layer.

In crossflow microfiltration the transverse velocity (due to the permeation) is usually negligible compared to the axial (crossflow) velocity. As a result, the transverse velocity component is usually neglected in solving the equations-of-motion in these systems. However, the concentration polarization boundary layer where the mass transfer of the particles occurs is usually very thin compared to the channel height; hence, very close to the wall, the axial and transverse velocities can be comparable. Therefore, in order to model the

mass transfer more accurately, a velocity profile that incorporates the effects of both the transverse and axial components of the velocity field is required.

2.1 Velocity profile in a channel with flux through the membrane

The geometry, coordinate system and other details of the CMFM channel are shown in Fig.1 in which x and y denote the coordinates in the axial and transverse directions, respectively, and v_w denotes the volumetric permeation flux at the wall. The dimensionless transverse coordinate is denoted by λ (y/h); this implies that λ varies between 0 (the solid wall) and 1 (the membrane surface). When the width of the channel (w) is much larger than the height (h), the flow can be considered to be two-dimensional. A sufficiently long entry region is provided so that the velocity profile within the region of interest can be considered to be fully developed. The flow is assumed to be steady and laminar. The velocity components in the x and y directions are denoted by u and v , respectively. The typical profile of the velocity components (u and v) and mass concentration (c) are shown in Fig. 1. It will be shown later that the particle deposition begins in locations where the modified Peclet number exceeds a critical Peclet value (Pe_{crit}). It also will be explained that the effective wall concentration (c_w) should be distinguished from the deposited cake concentration (c_c) for supermicron particles (Fig. 1). Since the onset of particle deposition near the local critical flux was studied here, the model does not consider any immobile cake layer thickness. Therefore, the axial velocity at the membrane wall was assumed to be zero. As a result, the boundary conditions constitute no-slip and no-permeation at the solid wall ($u, v=0$ at $\lambda=0$) and no-slip and a velocity determined by the permeation flux at the membrane ($u=0, v=v_w$ at $\lambda=1$).

Berman [23] used a perturbation solution to solve the equations-of-motion that incorporated a transverse velocity component arising from permeation through a porous wall. Details of the perturbation solution for this problem can be found in prior studies [20, 23]. The perturbation solution which used by Berman [23], was also used here in which a no-slip condition was assumed at the membrane surface. From the perturbation solution the velocity components can be calculated from the following [20]:

$$u(x, \lambda) = \left(u_0 - \frac{v_w x}{h}\right) (f'_0(\lambda) + Re_w f'_1(\lambda)) \quad (1)$$

and

$$v(\lambda) = v_w (f_0(\lambda) + Re_w f_1(\lambda)) \quad (2)$$

where Re_w denotes the wall Reynolds number ($v_w h/\nu$), ν is the fluid kinematic viscosity and u_0 is the mean axial velocity in the channel; $f_0(\lambda)$ and $f_1(\lambda)$ are functions of λ that are obtained from the perturbation solution:

$$f_0(\lambda) = -2\lambda^3 + 3\lambda^2 \quad (3)$$

and

$$f_1(\lambda) = \frac{-2}{35}\lambda^7 + \frac{1}{5}\lambda^6 - \frac{3}{10}\lambda^5 + \frac{27}{70}\lambda^3 - \frac{8}{35}\lambda^2 \quad (4)$$

This velocity profile given by Eqs. (1)-(4) is an input for the numerical solution for the mass transfer near the membrane surface in the next section.

Fig. 1. Schematic of CFMF showing the parallel plate channel geometry, the axial and transverse velocity profiles, concentration profile, concentration polarization boundary layer, and cake formation layer; particle deposition begins downstream from the channel inlet owing to the local dependence of the critical permeation flux.

2.2 Mass transfer of supermicron particles near the membrane surface

Close to the membrane surface in CFMF applications at a filtration flux below or near the critical flux [1, 3], there is a balance between the convection of particles ($v_w c_w$) toward the membrane and the back-diffusion of rejected particles from the membrane ($D \partial c / \partial y$). This can be written as a boundary condition at the membrane surface as follows:

$$D \frac{\partial c}{\partial y} = v_w c_w \quad (5)$$

where y is the normal direction to the membrane. The diffusion coefficient (D) for the rejected particles incorporates both the Brownian-induced and the shear-induced diffusion coefficients, D_B and D_s , respectively [24, 25]:

$$D = D_B + D_s = \frac{k_B T}{6\pi\mu r_p} + D_s \quad (6)$$

where k_B is the Boltzmann constant, T is the temperature, μ is the viscosity and r_p is the particle radius. It has been shown that for supermicron particles, the Brownian diffusion coefficient is negligible compared to the shear-induced diffusion coefficient (i.e., $D_B \ll D_s$) [7, 8]. Different empirical relationships have been used to approximate D_s ; most of these relationships are based on the results of the experiments conducted by Eckstein et al. [7]. For suspensions with a feed volume fraction (ϕ_b) in the range of $0.2 < \phi_b < 0.5$ (this range may be applied to the wall volume fraction ϕ_w), Zydney and Colton [6] approximated D_s by:

$$D_s = 0.03 r_p^2 \gamma_w \quad (7)$$

where γ_w denotes the shear rate at the membrane surface. They used this diffusion coefficient in the Lvque solution to obtain the local and average fluxes at the membrane surface. This will be referred to as the SID model.

A modified shear-induced diffusion coefficient was used by Li et al. [18] for more dilute suspensions with a wall volume fraction in the range $0 < \phi_w < 0.2$ and is given by:

$$D_s = 0.1 \phi_w r_p^2 \gamma_w \quad (8)$$

Note that D_s is dependent on the suspension volume concentration at the wall (ϕ_w). This diffusion coefficient was also used as an input to the Lvque solution. Since in the Lvque solution the wall concentration must be known and constant over the membrane surface, Li et al. assumed a wall volume fraction of 0.2 ($\phi_w = 0.2$) [18]. This will be referred to as the SID-MOD model.

Here, the velocity profile obtained via a perturbation analysis is used that allows for the effect of the permeation flux in a differential mass balance that incorporates shear-induced diffusion as well as convective mass transfer. Realistic flux conditions are applied at the impermeable and permeable solid boundaries. Neither the concentration nor the shear-induced diffusion coefficient is specified at the boundaries; the local values for both quantities are determined from a finite difference solution. The shear-induced diffusion coefficient is determined from the SID model for high concentrations and from the SID-MOD model for low concentrations:

$$\begin{cases} D_s = 0.1 \varphi_w r_p^2 \gamma_w & \text{if } 0 < \varphi_w < 0.2 \\ D_s = 0.03 r_p^2 \gamma_w & \text{if } 0.2 < \varphi_w < 0.5 \end{cases} \quad (9.a)$$

$$(9.b)$$

Note that the volume fraction (φ_w) and mass concentration (c_w) at the wall are distinguished from the volume fraction (φ_c) and mass concentration (c_c) of the cake (deposited layers of particles that have no motion relative to each other). That is, because the shear-induced diffusion of particles near the membrane surface is due to particle-particle interactions [7, 8], there is no flow induced particle-particle interaction within the cake region. Hence, φ_w denotes the volume fraction in the dynamic region close to the membrane surface but above the static region of deposited particles in the cake (i.e., see Fig.1). Note that there is a discontinuity in Eq. (9) at $\varphi_w = 0.2$ that arises from the difference between the SID and SID-MOD models. However, it will be shown in the results section that for this study of CFMF involving dilute suspensions the wall concentration never exceeds much more than 0.2; hence, only Eq. (9a) was used in the numerical solution. Note that the relation between mass concentration (c_w) and volume fraction (φ_w) is $c_w = \varphi_w \rho_p$ in which ρ_p is the particle density.

In CFMF systems the channel height (h) is much smaller than the channel length (L). Therefore, axial diffusion in the convective diffusion equation is negligible compared to transverse diffusion. By introducing the dimensionless variables $u^* = u/u_0$, $v^* = v/v_w$, $c^* = c/c_0$, $x^* = x/L$ and $\lambda = y/h$, the convective diffusion equation and boundary conditions can be written as follows:

$$\frac{u_0}{L} u^* \frac{\partial c^*}{\partial x^*} + \frac{v_w}{h} v^* \frac{\partial c^*}{\partial \lambda} = \frac{D_s}{h^2} \frac{\partial^2 c^*}{\partial \lambda^2} \quad (10)$$

$$\frac{\partial c^*}{\partial \lambda} = 0 \quad \text{at } \lambda = 0 \text{ for } 0 < x^* < 1 \quad (11)$$

$$\frac{\partial c^*}{\partial \lambda} = \frac{h}{D_s c_b} v_w c_w \quad \text{at } \lambda = 1 \text{ for } 0 < x^* < 1 \quad (12)$$

$$c^* = 1 \quad \text{at } x^* = 0 \text{ for } 0 < \lambda < 1 \quad (13)$$

The geometry, boundary conditions and coordinate systems used are shown in Fig. 1. To discretise the convective diffusion equation and boundary conditions given by Eqs. (10) to (13), a backward difference approximation is used for the convective terms on the left-hand side of Eq. (10) and a central difference approximation is used for the diffusive terms on the right-hand side of Eq. (10). The number of grid points in the axial direction (x -direction) is denoted by M and the subscript m denotes the grid number in this direction ($m=0$ and $m=M$ for the inlet and exit of the channel, respectively). For the transverse direction (y -direction) the grid number k varies between 0 (the solid wall) and K (the membrane surface); that is,

c_{km}^* denotes the dimensionless concentration of the k^{th} row in the y -direction and the m^{th} column in the x -direction. $\Delta x^* = 1/(M-1)$ and $\Delta \lambda = 1/(K-1)$ are the dimensionless grid spacings in the axial and transverse directions, respectively. The finite difference equations for different grid points in a column, denoted by F_k , can be written as follows:

$$F_1 = \frac{2D_s L}{u_0 h^2 \Delta \lambda^2} c_{1m}^* - \frac{2D_s L}{u_0 h^2 \Delta \lambda^2} c_{2m}^* = 0 \quad \text{for } k = 1 \quad (14)$$

$$F_k = \left[\frac{-Lv_w v_{km}^*}{u_0 h \Delta \lambda} + \frac{-D_s L}{u_0 h^2 \Delta \lambda^2} \right] c_{k-1m}^* + \left[\frac{u_{km}^*}{\Delta x^*} + \frac{Lv_w v_{km}^*}{u_0 h \Delta \lambda} + \frac{2D_s L}{u_0 h^2 \Delta \lambda^2} \right] c_{km}^* + \left[\frac{-D_s L}{u_0 h^2 \Delta \lambda^2} \right] c_{k+1m}^* \quad (15)$$

$$+ \left[\frac{u_{km}^*}{\Delta x^*} \right] c_{km-1}^* = 0 \quad \text{for } 2 \leq k \leq K-1$$

$$F_K = \left[\frac{-Lv_w}{u_0 h \Delta \lambda} + \frac{-2D_s L}{u_0 h^2 \Delta \lambda^2} \right] c_{K-1m}^* + \left[\frac{-Lv_w}{u_0 h \Delta \lambda} + \frac{2D_s L}{u_0 h^2 \Delta \lambda^2} \right] c_{Km}^* = 0 \quad \text{for } k = K \quad (16)$$

From Eq. (9) it can be seen that $D_s (= 0.1 c_{Km}^* c_{bp}^2 \gamma_w / \rho_p \text{ for } 0 < c_{Km}^* c_{bp} < 0.2)$ is a function of the local wall concentration (c_{Km}^*). Since Eqs. (14) to (16) constitute a system of nonlinear equations, Newton's method is employed [26]. A backward difference scheme has been used for discretization of the convective diffusion equation. The concentration in each column of points (column k) is dependent only on the left column (column $k-1$). Thus, the system of equations is solved for each column whereby the solution for column $k-1$ is used as the input for the system of equations for column k . A block diagram that illustrates the algorithm and protocol for solving the convective diffusion equation using Newton's method is shown in Fig. 2.

Fig.2. Algorithm (Newton's method) used for solving the nonlinear convective diffusion equation.

In Fig.2 tol refers to the convergence tolerance that is set to 10^{-6} for all the simulations in this study. C_m^* , $F(C_m^*)$ and $J(C_m^*)$ are the dimensionless concentration, finite difference equations and Jacobian, respectively, of the m^{th} column when the solution is cast in the following matrix form:

$$C_m^* = \begin{bmatrix} c_{1m}^* \\ c_{2m}^* \\ \vdots \\ c_{Km}^* \end{bmatrix}, F(C_m^*) = \begin{bmatrix} F_1(C_m^*) \\ F_2(C_m^*) \\ \vdots \\ F_K(C_m^*) \end{bmatrix}, J(C_m^*) = \begin{bmatrix} \frac{\partial F_1(C_m^*)}{\partial c_{1m}^*} & \frac{\partial F_1(C_m^*)}{\partial c_{2m}^*} & \cdots & \frac{\partial F_1(C_m^*)}{\partial c_{Km}^*} \\ \frac{\partial F_2(C_m^*)}{\partial c_{1m}^*} & \frac{\partial F_2(C_m^*)}{\partial c_{2m}^*} & \cdots & \frac{\partial F_2(C_m^*)}{\partial c_{Km}^*} \\ \vdots & \vdots & \ddots & \vdots \\ \frac{\partial F_K(C_m^*)}{\partial c_{1m}^*} & \frac{\partial F_K(C_m^*)}{\partial c_{2m}^*} & \cdots & \frac{\partial F_K(C_m^*)}{\partial c_{Km}^*} \end{bmatrix} \quad (17)$$

where $F_1(C_m^*)$ to $F_K(C_m^*)$ are defined by Eqs. (14) to (16). $\max \{F(C_m^*)\}$ refers to the largest component of the column matrix $F(C_m^*)$. To choose a proper grid spacing in the simulation, a grid sensitivity analysis has been performed based on the effect of the grid number on the concentration at the middle of the channel wall on the membrane side. Fig.3 shows a plot of

the aforementioned concentration as a function of the number of transverse (panel a) and axial grid points (panel b). The legend summarizes the fixed parameters for this sensitivity analysis. Fig. 3a indicates that the accuracy of the solution depends significantly on the axial grid number (K) but changes by less than 1% for values of $K > 4000$. Fig. 3b indicates that the accuracy does not depend significantly on M . Hence, the values of M and K are fixed at 1000 and 4000, respectively, in order to ensure a solution within 1% accuracy in the simulations.

Fig.3. The effect of the transverse (panel a) and transverse (panel b) grid numbers on the concentration at the middle of the channel wall constituting the membrane.

The principal features of the model developed here and the manner in which it relaxes assumptions made in prior studies are summarized in the following:

1. It employs a velocity profile obtained from a perturbation solution that incorporates the effect of the permeate flux through the membrane on the solution for the concentration profile near the membrane surface; this effect is neglected in the Lvque solution.
2. The wall concentration (c_w) is not specified but rather is predicted by a numerical solution to the describing equations in contrast to the Lvque solution that requires specifying the concentration on the membrane surface.
3. The shear-induced diffusion coefficient is calculated locally via a numerical solution to the nonlinear system of equations in contrast to prior studies that assumed a constant value.

Incorporating these improvements is shown to be important by comparing the predictions of the model developed here to those of the more limiting models developed in prior studies.

3. Material and methods

A schematic of the Direct Observation Through the Membrane (DOTM) setup is shown in Fig. 4. A gear pump (Cole Parmer) was used to circulate the feed suspension through the CFMF channel (109 mm length x 33.5 mm width x 2 mm height). The permeate was extracted using a low capacity peristaltic pump (Minipuls 3, Gilson). The variations of transmembrane pressure (TMP) and flux during the experiment were monitored using a digital balance (PL4002, Mettler) and three pressure transducers (Cole Parmer) connected to a data-logging system (LabVIEW). More details of the DOTM setup and CFMF module are given elsewhere [27].

Fig. 4. Schematic of the Direct Observation Through the Membrane (DOTM) setup.

Transparent Anopore inorganic disc membranes (Anodisc, Whatman, Germany) with a diameter of 47 mm and a nominal pore size of 0.2 μm were used in this study. In order to facilitate the placement of the membrane in the acrylic CFMF module, the membrane was framed between two sheets of paper that created a square active membrane area (2.7 cm x 2.7 cm) in the middle of the flow channel (Fig. 5a). Each experiment for a fixed set of system parameters was conducted with using a new membrane. Polystyrene latex beads were used as model particles (Fluka, Sigma Aldrich, USA). Since the concentrations used in this study were very low, the kinematic viscosity, ν , of the suspensions was assumed to be that of pure water (i.e., $10^{-6} \text{ m}^2/\text{s}$).

3.1. Experimental protocol

Flux-stepping tests were conducted by incrementally increasing the flux at 15-minute intervals. A series of filtration runs was performed at crossflow velocities (CFVs) of 0.07 - 0.13 m/s, feed concentrations (c_b) of 0.5 and 1 g/L, and particle sizes of 6 μm and 10 μm (d_p). Five cases defined by different combinations of these parameters were studied as summarized in Table 1. The highest Reynolds number reached in these five cases was 520 (related to Case 2); thus, the flow regime was laminar. In order to investigate the effect of axial distance on particle deposition, DOTM images were captured with a colour video camera (TK-C921BEG, JVC) and AxioVision software (Carl Zeiss) at two locations: 0.7 cm (Point 1) and 2.2 (Point 2) cm from the feed inlet (Fig. 5a). These locations were pre-set by using a motorized stage (mechanical stage 75 x 50 mot standard, Carl Zeiss) coupled with an electronic coaxial drive. The recorded images then were analysed to determine the rate of particle deposition. The total membrane surface area viewed under the microscope (A) was $280 \times 210 \mu\text{m}^2$ (Fig. 5b).

Fig. 5. (a) Placement of the transparent Anopore membrane in the channel; (b) the total membrane surface area viewed under the microscope.

To determine the local critical flux ($J_{crit}(x)$) for the different cases and axial positions, the following relationship, which is obtained from a particle mass balance [28, 29], was used:

$$\frac{\Delta N}{\Delta t} = \frac{3}{4} \frac{\theta c_b A}{\pi \rho_p r_p^3} (v_w - J_{crit}(x)) \quad (18)$$

The local critical flux is shown by $J_{crit}(x)$ to imply that the critical flux is function of the distance (x) from the inlet of the channel as well as other hydrodynamic parameters [2]. N is the number of particles, A is the membrane area and θ is the fractional deposition constant (the fraction of particles convected to the membrane that are deposited on it) [29]. Eq. (18) relates the particle deposition rate ($\Delta N/\Delta t$) to the difference between the permeate flux and

critical flux ($v_w - J_{crit}(x)$). The flux-stepping was continued for at least three more steps after deposition began for each case. At each flux step, the particle deposition rate for both Points 1 and 2 was determined from the images captured at the two time points (t_1 and t_2), as illustrated in Fig. 6. Eq. (18) implies that if the particle deposition rate ($\Delta N/\Delta t$) is plotted versus the permeate flux (v_w), the intercept of the regression line with the horizontal axis (flux axis) corresponds to the highest flux at which the particle deposition rate is zero (i.e., the critical flux). The resulting critical fluxes determined in this manner for the five cases are presented and discussed in Section 4.

Table 1. Particle diameter (d_p), concentration (c_b) and crossflow velocity (CFV) values for the five cases in this study.

Case No.	d_p (μm)	c_b (g/L)	CFV (m/s)
Case 1	10	0.5	0.07
Case 2	10	0.5	0.13
Case 3	6	0.5	0.075
Case 4	6	1.0	0.075
Case 5	6	1.0	0.1

Fig. 6. Permeation flux (v_w) and transmembrane pressure (TMP) as a function of time for the flux-stepping protocol; the two micrographs show the particle deposition at 79 and 85 seconds from which the particle deposition rate can be determined as illustrated.

4. Results and discussion

In these experiments involving the use of DOTM in CFMF, the particle deposition at a constant overall permeation flux was found to be a function of axial location. This can be observed in Fig. 7 that shows the particle deposition at different locations along the membrane at nearly the same time for which the experiment conditions are given on the figure: left edge or inlet (Fig. 7a); Point 1 (Fig. 7b); middle of the channel (Fig. 7c); Point 2 (Fig. 7d); and right edge or exit (Fig. 7e). This suggests that the critical flux should be determined locally to determine the onset of particle deposition in CFMF. These data will be analyzed quantitatively in the subsequent discussion.

Fig. 7. The membrane surface coverage at the same time at different distances from the channel inlet: (a) left edge or inlet; (b) Point 1; (c) middle of the channel; (d) Point 2; (e) right edge or exit.

Providing a quantitative explanation for this apparent local critical flux in CFMF was a major goal of this study. Prior models could not address this question owing to limiting assumptions. In particular, the model developed in this study does not require specifying the concentration at the membrane surface and allows for the concentration dependence of the shear-induced diffusion coefficient in both the axial and transverse directions. As such, the model can predict the dependence of the critical flux on the coupled hydrodynamics and mass transfer in the concentration polarization boundary layer. The modelling results for the critical flux can be generalized by casting them in terms of the critical modified Peclet number. The modified Peclet number is the dimensionless ratio of the transverse convection (i.e., owing to the permeation flux) to the diffusion of the rejected particles in crossflow membrane filtration. It was first introduced by Bhattacharya and Hwang [22] to characterize the concentration polarization boundary layer in nanofiltration and reverse osmosis. A critical Peclet number (Pe_{crit}) in the concentration polarization boundary layer has been suggested by Bacchin and co-workers [2, 3] that provides a dimensionless criterion for assessing when the convective flux of particles towards the membrane exceeds their back-diffusion, thereby causing the onset of particle deposition. The critical modified Peclet number is defined as follows:

$$Pe_{crit} = \left(\frac{J_{crit}(x)\delta_c}{D_s} \right)_{crit} \quad (19)$$

where δ_c is the concentration polarization boundary layer thickness that can be a function of axial position. This suggests that local particle deposition should occur if $Pe > Pe_{crit} \cong 1$. Moreover, if the critical modified Peclet number is to fundamental significance for CFMF, its value should be independent of the operating conditions (i.e., the conditions for the different cases). This hypothesis will be tested by employing the model developed in this study to determine the local concentration polarization boundary layer thickness and shear-induced diffusion coefficient. The predicted local values of Pe_{crit} will be used to interpret the observed particle deposition determined by the DOTM characterization. The model developed in this study will be used to assess the error incurred in using the SID and SID-MOD models to predict Pe_{crit} .

4.1 Determination of critical fluxes

The method for determining the critical flux was explained in Section 3.1. Fig. 8a shows the particle deposition rate as a function of the permeation flux for Point 2 for each of the five cases. The intercept of the best fit line for each set of points with the horizontal (permeate flux) axis is identified with the critical flux for that case. The critical fluxes along with the R^2 values for the linear regression obtained via this procedure are shown in the legend of Fig. 8a.

As expected, a higher CFV, larger particle diameter and a lower feed concentration resulted in a higher critical flux. The same method for calculating the critical flux at Point 1 was used. The results for both Points 1 and 2 are summarized in Fig. 8b, which clearly shows that the critical fluxes are lower for point 2, which is farther from the channel inlet than Point 1. These results confirm that the critical flux is a local concept whose value is strongly dependent on the hydrodynamics. The critical flux measurements are reproducible with a relative error within ± 2.5 %. These critical fluxes will be used to determine the critical modified Peclet number for the different cases at the two locations.

Fig. 8. (a) Particle deposition rate as a function of permeate flux at Point 2 from which the critical flux is determined for the different cases; (b) Comparison between the critical fluxes determined at Points 1 and 2 for the different cases

4.2 Comparison between predictions of the different models

Fig. 9 shows a plot of the shear-induced diffusion coefficient D_s (panel a) and the dimensionless concentration polarization boundary layer thickness δ_c^* ($= \delta_c/h$) (panel b) as a function of the dimensionless distance from the channel inlet for the model developed in this study (dashed line), the SID model (dash-dot line) and the SID-MOD (solid line). The comparison is shown only for Case 1 (Point 1) for which the local critical permeate flux is $41.4 L/m^2h$ ($1.15 \times 10^{-5} m/s$). Fig. 9a shows that D_s is constant along the membrane length for both the SID and SID-MOD models owing to the assumption of a constant wall

concentration. In contrast, D_s increases with increasing distance from the channel inlet for the model developed in this study since it does not specify the wall concentration but determines it from the numerical solution. The SID and SID-MOD models overestimate D_s and thereby predict a thicker concentration polarization boundary layer thickness δ_c than does the model developed here (Fig. 9b). In Section 4.3 the critical modified Peclet numbers predicted by the three solutions will be compared.

Fig. 9. Shear-induced diffusion coefficient D_s (panel a) and the dimensionless concentration polarization boundary layer thickness δ_c^* ($= \delta_c/h$) (panel b) as a function of the dimensionless distance from the channel inlet for the model developed in this study (dashed line), the SID model (dash-dot line) and the SID-MOD (solid line) for Case 1.

4.3 Critical modified Peclet number

The critical modified Peclet number, Pe_{crit} , has been advanced as a possible key parameter to characterize the mass transfer of supermicron particles in CFMF. The value of Pe_{crit} depends on the method used to solve the convective diffusion equation. Fig. 10 shows a plot of Pe_{crit} based on the critical flux determined via the procedure shown in Fig. 8a and based on the values of D_s and δ_c predicted by the SID model at Points 1 and 2 for the five cases. The Pe_{crit} values differ between the five cases as well as at the two points for each case. In principle, Pe_{crit} should be the same irrespective of the operating conditions and the axial position since this dimensionless group characterizes the balance between convection and diffusion when particle deposition begins. Fig. 11 shows the same plot for Pe_{crit} predicted by the SID-MOD model. Again, the values of Pe_{crit} vary widely for the different cases and the two observation points. This means that the SID and SID-MOD models do not lead to a critical Peclet number. However, Fig. 12 shows the values of Pe_{crit} predicted by the model developed in this

study for the five cases and the two observation points. In contrast to Figs. 10 and 11, $Pe_{crit} = 4 \pm 0.08$ for all the results shown in Fig. 12. If Pe_{crit} properly characterizes the conditions defining the onset of particle deposition, its value should be a constant irrespective of the operating conditions. Moreover, the fact that $Pe_{crit} \cong 1$ indicates that this dimensionless group properly characterizes the conditions for which the convection and back-diffusion of particles just balance each other. The model developed in this study predicts a constant value of Pe_{crit} because it more accurately predicts the values of D_s and δ_c that appear in Pe_{crit} owing to relaxing the constraints of a constant c_w and D_s embodied in the SID and SID-MOD models. Hence, in contrast to the critical flux that has been shown to vary locally in these CFMF studies, Pe_{crit} is a global dimensionless parameter whose value characterizes the onset of particle deposition irrespective of axial position, particle diameter, bulk concentration of particles and crossflow velocity.

Fig. 10. Critical modified Peclet numbers predicted by the SID model at Points 1 and 2 for the five cases corresponding to different operating conditions.

Fig. 11. Critical modified Peclet numbers predicted by the SID-MOD model at Points 1 and 2 for the five cases corresponding to different operating conditions.

Fig. 12. Critical modified Peclet numbers predicted by the model developed in this study at Points 1 and 2 for the five cases corresponding to different operating conditions.

5. Conclusions

The concept of a critical flux above which particle deposition commences is well-established. However, several studies have shown that the critical flux is dependent on the process parameters such as the particle diameter, bulk concentration of particles and crossflow velocity. This study has explored the concept of the critical modified Peclet number, a dimensionless group that provides a measure of the ratio of the convection to diffusion of the particles. The critical modified Peclet number incorporates both the local concentration polarization boundary layer thickness and the local diffusion coefficient. Since these two quantities cannot be easily measured, it is necessary to predict them from a solution to the convective diffusion equation. Prior studies have employed the Lvque solution that does not incorporate the effect of the permeation flux on the velocity profile within the concentration polarization boundary layer and have assumed a constant concentration and a constant diffusion coefficient along the membrane surface. As such, these models cannot predict the concentration polarization boundary layer thickness and diffusion coefficient accurately. In this study the effect of the permeation flux on the velocity profile was incorporated into the model and neither the concentration nor the shear-induced diffusion coefficient was specified along the membrane surface. The resulting nonlinear convective diffusion equation was solved numerically to determine the concentration polarization boundary layer thickness and shear-induced diffusion coefficient as a function of axial

distance. These predictions along with the values of the critical flux were used to determine the critical modified Peclet number at two axial positions for five cases involving different values for the particle diameter, bulk particle concentration and crossflow velocity. The resulting critical modified Peclet number was found to be a constant of order one irrespective of axial position or the process parameters for the five cases. This suggests that the critical modified Peclet number provides a global criterion for the inception of particulate fouling in contrast to the critical flux that can depend on the axial position as well as the process parameters.

This study provides strong support for the use of the critical modified Peclet number as a criterion for the onset of particle deposition in CFMF. However, this study considered only the deposition of two different sizes of latex microspheres at two bulk concentrations for five crossflow velocities. The concept of a critical modified Peclet number should apply to different types of particles if the onset of deposition is determined by a balance between convection and diffusion of particles to and from the membrane surface, respectively. However, if additional mechanisms are involved such as eddy diffusion (i.e., turbulent flow), Brownian diffusion or electrostatic interaction between the membrane and the particles, the concept of the critical modified Peclet number as defined in this study undoubtedly will need to be modified. This study establishes that in the absence of the aforementioned additional mechanisms, the onset of particle deposition in CFMF is determined by a unique value of the modified Peclet number irrespective of the axial position, particle diameter, bulk concentration or crossflow velocity. Further work is required to assess the general applicability of our new modeling approach and the factors influencing Pe_{crit} .

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Nomenclature

A	Area of the membrane	m^2
c	Mass concentration	kg/m^3
c^*	Dimensionless mass concentration	-
c_b	Feed mass concentration	Kg/m^3
c_c	Mass concentration of the cake	kg/m^3
c_w	Mass concentration at the wall	kg/m^3
D_B	Brownian-induced diffusion coefficient	m^2/s
D_s	Shear-induced diffusion coefficient	m^2/s
h	Channel height	m
J	Jacobian matrix	-
J_{crit}	Critical flux	m/s
k_B	Boltzmann constant	$\text{m}^2\text{kg/s}^2\text{K}$
K	Grid points number in the transverse direction	-
L	Channel length	m
M	Grid points number in the axial direction	-
N	Particle number deposited on the membrane	-
r_p	Particle radius	m
t	Time	s
T	Temperature	K
u	Axial velocity component	m/s
u^*	Dimensionless axial velocity component	-
u_0	Mean axial velocity	m/s
v	Transverse velocity component	m/s
v^*	Dimensionless transverse velocity component	-
v_w	Volumetric permeation flux at the wall	m/s
x	Axial coordinate	m
x^*	Dimensionless axial coordinate	-
Δx^*	Dimensionless grid spacing in the axial	-

	direction	
y	Transverse coordinate	m
w	Channel width	m
<i>Greek symbols</i>		
γ_w	Shear rate at the wall	1/s
δ_c	Concentration boundary layer thickness	m
θ	Fractional deposition constant	-
λ	Dimensionless transverse coordinate	-
$\Delta\lambda$	Dimensionless grid spacing in the transverse direction	-
μ	Viscosity	kg/m s
ν	Kinematic viscosity	m ² /s
ρ_p	Particle density	kg/m ³
φ	Volume fraction	-
φ_b	Feed volume fraction	-
φ_c	Volume fraction of the cake	-
φ_w	Volume fraction at the wall	-
<i>Superscript</i>		
*	Denotes dimensionless parameters	
<i>Subscripts</i>		
<i>crit</i>	Denotes critical value	
<i>w</i>	Denotes the wall of the membrane	
<i>Abbreviations</i>		
CFMF	Crossflow microfiltration	
CFV	Crossflow velocity	
SID	Shear-induced diffusivity	
SID-MOD	Modified shear-induced diffusivity	

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FIGURE CAPTIONS

Fig. 1. Schematic of CFMF showing the parallel plate channel geometry, the axial and transverse velocity profiles, concentration profile, concentration polarization boundary layer, and cake formation layer; particle deposition begins downstream from the channel inlet owing to the local dependence of the critical permeation flux.

Fig. 2. Algorithm (Newton's method) used for solving the nonlinear convective diffusion equation.

Fig. 3. The effect of the transverse (panel a) and transverse (panel b) grid numbers on the concentration at the middle of the channel wall constituting the membrane.

Fig. 4. Schematic of the Direct Observation Through the Membrane (DOTM) setup.

Fig. 5. (a) Placement of the transparent Anopore membrane in the channel; (b) the total membrane surface area viewed under the microscope.

Fig. 6. Permeation flux (v_w) and transmembrane pressure (TMP) as a function of time for the flux-stepping protocol; the two micrographs show the particle deposition at 79 and 85 seconds from which the particle deposition rate can be determined as illustrated.

Fig. 7. The membrane surface coverage at the same time at different distances from the channel inlet: (a) left edge or inlet; (b) Point 1; (c) middle of the channel; (d) Point 2; (e) right edge or exit.

Fig. 8. (a) Particle deposition rate as a function of permeate flux at Point 2 from which the critical flux is determined for the different cases; (b) Comparison between the critical fluxes determined at Points 1 and 2 for the different cases

Fig. 9. Shear-induced diffusion coefficient D_s (panel a) and the dimensionless concentration polarization boundary layer thickness δ_c^* ($= \delta_c/h$) (panel b) as a function of the dimensionless distance from the channel inlet for the model developed in this study (dashed line), the SID model (dash-dot line) and the SID-MOD (solid line) for Case 1.

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Fig. 11. Critical modified Peclet numbers predicted by the SID-MOD model at Points 1 and 2 for the five cases corresponding to different operating conditions.

Fig. 12. Critical modified Peclet numbers predicted by the model developed in this study at Points 1 and 2 for the five cases corresponding to different operating conditions.

TABLE CAPTIONS

Table 1. Particle diameter (d_p), concentration (c_b) and crossflow velocity (CFV) values for the five cases in this study.