Membrane fouling quantification by specific cake resistance and flux enhancement using helical cleaners

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Abstract
Membrane fouling in a cross-flow ultrafiltration (UF) is investigated using a tubular membrane for three different feed solutions containing oil droplets and particles. The initial build-up of a highly concentrated fouled cake layer on the membrane surface results in sharp flux decline which acting as secondary filtration layer over the membrane surface. In the present work, the fouled cake layer resistance is quantified in terms of specific cake resistance (α), which depends strongly on the operating parameters. Cake resistance, α, calculated from experimental data using different models, is used as an operational indicator to quantify membrane fouling. Two sets of experiments are performed, first by operating tubular membrane as it is, and later by introducing a static helical turbulence promoter. Results showed that the influence of pressure and feed concentration on α is more important than that of feed temperature and cross-flow velocity. Oil droplets sizes, which vary mainly with temperature, play an important role in the interpretation of results. Droplets coalescence and flocculation of suspended particles and the aggregation of droplets on particles explain these variations. The cake layer is significantly reduced by using a novel static helical type turbulence promoter inside the tubular membrane module allowing operation at low pressure and low cross-flow velocity, thus, aid in reducing membrane fouling without significantly increasing energy consumption. Computational Fluid Dynamics (CFD) calculations of full Navier-Stokes equations are also performed to gain insight into flow hydrodynamics and for estimating shear stresses when the turbulent promoter is deployed. The result indicates that roughly 4-5 times higher shear stress on the membrane surface is generated depending on the applied cross-flow velocity. It potentially explains significant cake layer alteration with variations in specific resistance and corresponding flux enhancement as observed in experiments.
**Keywords:** Membrane fouling, ultrafiltration, helical turbulence promoter, CFD, hydrodynamics, DNS.

1. **Introduction**

Membrane processes can be classified into four types depending on the solutes sizes to be separated and/or the membrane molecular weight cut-off or pore size. Two filtration modes are possible: namely, dead-end and cross-flow. The main common feature of these processes is the permeation flux decline (at constant pressure) due to concentration polarization (CP) and fouling phenomenon, which are the major bottleneck that hinders their wide application. The decline in flux is attributed to several resistances-in-series, composed of three main resistances: namely, membrane resistance, CP resistance (reversible) and fouling layer resistance (reversible/irreversible) due to internal (standard), partial and external pore plugging.

Several studies have used specific cake resistance ($\alpha$) as a parameter to evaluate the degree of reversible and irreversible membrane fouling [1-9]. Different results focusing on the influence of $\alpha$ on operating parameters such as pressure, temperature, velocity, concentration, particles size and shape, ionic strength, pH, and cake layer porosity and its resistance have been found by different authors. During rice wine dead-end microfiltration (MF), Sripui et al. [1] reported that $\alpha$ may vary with different operating conditions such as pressure and concentration and depends on many factors such as particles shape and density, size distribution and porosity. They found that $\alpha$ decreased as the ratio of particles having sizes larger than 45 $\mu$m increased, increased linearly with the increase of total suspended solids (SS) concentration for particle sizes of the rage of 1-20 $\mu$m, had minimal influence for particles greater than 20 $\mu$m, and is independent of concentration for particles larger than 45 $\mu$m. McCarthy et al. [7] presented some findings on the influence of $\alpha$ on different operating parameters in both cross-flow and dead-end filtration modes. They reported that the main dependence of $\alpha$ variation is the particles size of different suspensions which, in turn, could vary with some operating parameters. Wang et al. [4] studied the influence of different operating parameter conditions on $\alpha$ using a yeast suspension in cross-flow MF. They found that $\alpha$ increased with the increase of cross-flow velocity, pressure, and concentration to an optimum value of 3 g/L, and decreased with increasing temperature. In dead-end MF using Bacillus subtilis as feed solution, Graves et al. [6] also found that increasing the pressure increases $\alpha$, but results in little increase in flux.
Different models have been used to evaluate $\alpha$. Listiarini et al. [2] used the curves of MFI (modified fouling index) by plotting $t/V$ (t: time and V: filtrate volume) versus V to determine $\alpha$ from the slope in dead-end nanofiltration (NF). They found that the adsorption resistance and the resistance due to pore blocking are negligible compared to the cake resistance as NF pores are too small compared to the particles in suspension. Khan et al. [3] also used the MFI model to determine $\alpha$. They found that $\alpha$ was strongly correlated to the fouling rates and that cake resistance contribution was predominant compared to irreversible fouling resistance. They stated that $\alpha$ could be considered as a reliable indicator to predict the degree of membrane fouling. On the other hand, in dead-end filtration of a mixed liquor of suspended solids, Chang and Kim [5] found that the membrane cake resistance decreased as the feed concentration decreased but $\alpha$ is not in contradiction with theory for low concentrations. Values of $\alpha$ were determined experimentally from the plot $(1/J^2; J$ is the flux) versus time. The membrane cake resistance was found to be a predominant factor governing the overall flux decline. From this contradiction, they concluded that $\alpha$ can’t be a proper parameter to estimate the rate of fouling, especially at low feed concentrations, ignoring that the main reason could be the size of the particles and not the concentration.

Teoh et al. [10] developed a new procedure to determine the membrane resistance $R_m$ and $\alpha$ from constant pressure cake filtration of CaCO$_3$ and Kaolin suspensions. The new procedure allows establishing the relationship of $\alpha_{av}$ versus the cake compressive stress values based on the knowledge of the instantaneous filtration rate, and compares them with $t/V$ versus V plots and compression/permeability measurements. They found that $\alpha_{av}$ increased with the pressure drop across the filter cake. Kim and Yuan [11] also developed a simple hydrodynamic model to evaluate the membrane cake resistance to quantify $\alpha$. In their model they used dimensionless $\alpha$ by multiplying its value with the square of the radius of outer porous shell and represented it versus the occupancy fraction. In their study, they questioned the validity of the Carman-Kozeny equation to estimate $\alpha$ for certain values of aggregates porosity. By developing the model, they explained the fact that Happel and Carman-Kozeny equations are almost indistinguishable. Pontie et al. [12] investigated fouling on three different molecular weight cut-off (100 kDa, 30 kDa and 10 kDa) membranes for regenerated cellulose with humic acid by varying pH of the feed solution. Neutral pH was found to have the highest fouling, whereas the lowest occurred at basic pH. Tighter membrane was found to be relatively less fouled compared with higher molecular weight cut-off
membranes. Introduction of clay particles in the feed solution with humic acid significantly altered the fouling behaviors and altered specific resistance quite dramatically.

Different authors used several techniques aiming to reduce CP and fouling, such as use of helical baffles [13-16], spacers [17, 18], feed pulsation in a baffles tubular membrane system [19], backpulsing [20, 21], use of rotational membranes [22], or corkscrew vortices formed in helical flow passage by the interaction of dean vortices and axial flow component [23], and air or gas sparging during filtration [24-26]. All these authors observed a significant increase in flux. They reported that the flux improvement was attributed to a reduction of the cake/gel layer formed on the surface of the membrane by increasing the cross-flow velocity, and thus the shear stress. Based on previous studies [1-9] it appears that $\alpha$ depends on several parameters and in general more sensitive with the type of feed and the membrane utilized for specific filtration purpose. Minimize $\alpha$ would result in lowering the fouling tendency and will enhance the flux recovery. In the present work, a novel static turbulent promoter is designed to minimize $\alpha$ and enhance the flux recovery through a cross-flow filtration using a tubular membrane. The static promoter is hydrodynamically designed such that it does not induce significant pressure drop and it produces enough turbulence on the feed lumen side to mitigate fouling, thus, aid in reducing $\alpha$. An inorganic tubular membrane made of zirconia active layer deposited on a carbon support is evaluated in terms of $\alpha$ as a parameter to quantify cross-flow ultrafiltration (UF) membrane fouling. Three different feed solutions primarily targeting produced water (imitating waste water from oil industry) are used in the present study; mainly an oil-in water (OiW) emulsion, SS and mixed suspensions (MS, i.e. produced water) containing oil droplets and SS. The significance of OiW emulsion filtration and associated technological hurdles and membrane development are recently presented in [27]. The behavior of $\alpha$ to predict membrane fouling for the different solutions, its influence on permeation flux for different operating parameters with and without using the helical-type turbulence promoter is further investigated. Mathematical and empirical models are used to calculate $\alpha$ using experimental data. In addition, flow hydrodynamics through the static turbulence promoter is also investigated through Computational Fluid Dynamics (CFD). It provides a fundamental understanding on shear stress and velocity distribution that quantifies fouling mitigating behavior as seen in experiments.

2. Theory
UF performance is affected by the hydraulic resistance of the membrane $R_m$ (m$^{-1}$) and the combined resistance (cake resistance) $R_c$ (m$^{-1}$) induced by reversible and/or irreversible processes, e.g. CP, cake or gel layer formation, and fouling. Therefore, the permeation flux $J$ (L.m$^{-2}$.h$^{-1}$) at steady state (stability of flux versus time after the build-up of a uniform cake layer on the surface of the membrane) is described by Darcy’s law (resistances-in-series model):

$$J = \frac{\Delta P}{\mu(R_m + R_c)} = \frac{1}{A} \frac{dV}{dt}$$

where $\Delta P$ is the applied pressure (Pa), $\mu$ is the dynamic viscosity (Pa.s), $V$ is the filtrate volume (m$^3$), $t$ is the time (s), and $A$ is the membrane surface area (m$^2$).

While $R_m$, which is related to the physical properties of the membrane, is considered to be constant as shown in Eq. (2) obtained from Hagen-Poiseuille law (assuming cylindrical capillary pores of uniform diameter and perpendicular to the membrane surface), the resistance $R_c$ is expressed as a function of $\alpha$ by (Eqs. (3-5)):

$$R_m = \frac{L_p}{K} = \frac{8\pi L_p}{\pi \rho_p n \rho_{p}} = \frac{8\pi L_p}{2 \tau \rho_p}$$

$$R_c = \frac{\alpha C_o}{A}$$

where $C_o$ is the mass of accumulated matter per unit volume of permeate (kg.m$^{-3}$) and could be defined as the volume fraction occupied by particles/colloids in the feed suspension $x_o$ (-), $\tau$ is the tortuosity factor (-), $L_p$ is the pore length or membrane thickness (m), $K$ is the medium permeability (m$^2$), $P_p$ is the pore density or the number of pores per unit of membrane area (m$^{-2}$), $r_p$ is the pore radius (m), and $\varepsilon$ is the cake porosity.

$R_c$ is also defined as a function of the cake mass (amount of deposited material) per unit area $m_c$ (kg.m$^{-2}$) [5, 28, 29], Eq. (4); or as a function of the cake layer thickness $\delta$ (m) [11, 30] Eq. (5).

$$R_c = \alpha m_c$$

$$R_c = \alpha \delta$$

With $\delta = 2 n r_{pr}$, or $\delta = \frac{m_c}{\rho_p (1 - \varepsilon)}$ [28]

where $r_{pr}$ is the particle radius (m), $n$ is the number of mono-cellular layers, and $\rho_p$ is the particle density (kg.m$^{-3}$).

Contrary to the classical dead-end operation, in cross-flow mode, it is more difficult to measure experimentally the cake mass per unit area during the filtration as there is no mass balance relating
the cake mass and the flux, therefore it is not easy to obtain $\alpha$. McCarthy et al. [7] reviewed some difficult procedures used by different authors to measure the cake mass such as weighing the membrane before and after filtration or removing the cake layer by pushing a cleaning rod inside a tubular membrane. As a result, $\alpha C_0$ values are most commonly determined using the classical cake filtration model (from the slope of $t/V$ vs. $V$).

If we assume that the deposit consists of isometric grains, $\alpha$ is inversely proportional to the cake layer porosity to the power of three (Eq. (6)); or, as a function of the particle diameter $d_p$ (m), the cake porosity and particle density (Carman-Kozeny relationship for spherical particles, mostly applied for incompressible cakes with a porosity range between 0.3 and 0.6) (Eq. (7)):

$$\alpha = \frac{h_k a_g^2 (1 - \varepsilon)^2 \delta}{\varepsilon^3 L}$$  \hspace{1cm} (6)

$$\alpha = \frac{180 (1 - \varepsilon)}{\rho_p d_p^2 \varepsilon^3}$$  \hspace{1cm} (7)

where $h_k$ is the Kozeny constant, $a_g$ is the grain specific surface area which is defined either by surface area divided by volume ($m^2.m^{-3}$ or $m^{-1}$) or surface area divided by mass ($m^2.kg^{-1}$), and $L$ is the cake length (m). These equations show clearly the tendency of the decrease of $\alpha$ with the increase of the cake porosity (-).

Looking at the different equations, the unit of $\alpha$ could be defined as (m.kg$^{-1}$) according to Eqs. (3), (4) and (7) or as (m$^{-2}$) according to Eqs. (3), (5) and (6). Eq. (3) is valid for both units depending on the definition of the bulk concentration in the equation which could be expressed as concentration ($kg.m^{-3}$) [1, 4, 7-9], or as the bulk volume fraction occupied by particles or colloids in the feed suspension (dimensionless) [11, 30, 31-33]. It is important to mention here that the trends of $\alpha$ variation with the different operating parameters and the interpretation of results presented in this paper remain the same regardless of the unit used.

3. Materials and methods

3.1. Experimental unit and membranes

Feed solutions were filtered in a cross-flow (tangential flow) UF apparatus equipped with an inorganic composite tubular membrane, as represented schematically in Fig. 1. Both permeate and concentrate were recirculated in the feed tank to keep the feed concentration constant. The pressure was controlled using a diaphragm valve. The homogeneity of feed solutions was maintained by continuously stirring and thermo-regulating the feed tank. In the case of OiW emulsions, unstable
solutions were continuously stirred at constant temperature in the feed tank for 10-15 min before starting the experiments to assure the homogeneity of the feed solution. When oil droplets are still observed on the surface of the feed tank walls (coalescence), the feed solution was recirculated in the feed tank using a centrifugal pump at high velocity until reaching a homogeneous solution before starting the experiments.

Figure 1: A schematic diagram of the experimental unit, and picture of the turbulence promoter fabricated using stainless steel.

Tubular membrane made of a zirconia active layer deposited on a carbon support was used in this study. The membrane samples were analyses for surface topography by scanning electron micrograph (SEM) instrument. Figs. 2(A) and 2(B) show the outer porous support layer and inner lumen surface of the membranes. The thickness of the tubular membrane is about 2 mm (Fig. 2(C)). The inner dense layer of about 8-10 µm thickness is visible in Fig. 2(D) and can be further distinguished into two different dense layers. Innermost dense layer (facing lumen) is around 2 µm thick and the porous sandwiched layer between the two (outer support and the innermost lumen layer) is about 6-8 µm thick. The mean pore size of the membrane provided by the manufacturer and confirmed by porometry test in our laboratory is 0.02 µm. The length and internal diameter of the membrane are 23.5 cm and 0.6 cm, respectively. The effective membrane surface area is 44.3 cm². These types of inorganic membranes are robust and can be cleaned with strong chemicals such as acids and produce permeate quality free of oil regardless of the operating parameters [34].
Figure 2: SEM of the tubular membrane. (A) outer Surface, (B) inner lumen, (C) total thickness, and (C) sandwich layer between outer and inner lumen of the membrane. The internal diameter of the membrane is 6 mm.

3.2. Suspensions and determinations

The synthetic OiW emulsion was made of a heavy crude oil. Various oil concentrations were added in DI water in order to reach desired feed water concentrations. On the other hand, the mixed suspension solutions were prepared by adding the desired concentration of particles in the OiW emulsion. Well-defined amounts and sizes of polystyrene particles was used. Droplets and particles size distribution was quantified using a Malvern Mastersizer/E Laser Granulometer. Permeate quality was analyzed by extraction and infrared absorptiometry (OCMA-220 Horiba) and HPLC.

3.3. Experimental protocol

The duration of each filtration experiment was 2 h. Membrane cleaning was performed by a simple backwash for 1 min duration using DI water (for suspended particles solutions) and using 30% sodium hydroxide and 30% nitric acid solutions (for OiW emulsions) after each filtration experiment (2 h). The new membrane permeability was achieved with one cleaning cycle in all experiments suggesting that no severe irreversible fouling occurred. All experiments have been
performed in duplicate or triplicate and the average values are presented in the figures. The experimental errors were within 4%.

3.4. Turbulence promoter

The static turbulence promoter (helical baffle type) used in this study corresponds to wound stems in circular helices consisting of variable number of helices per unit length, as shown in Fig. 1. The static turbulence promoter was carefully inserted inside the tubular membrane element, as shown in Figure 3. To avoid vibrations inside the module due to fluid flow, their diameter is equivalent to the inner membrane tube so that the helices heads touch the membrane surface when installed. To the difference of the systems previously proposed by [15, 35, 27], this new design consists of welding each helix to the consecutive one with an angle of 90° (Fig. 1b). This system generates more turbulence and better mixing aiming to reduce CP and reduces pressure drop inside the membrane module.

3.5. Hydrodynamics computations

The hydrodynamics difference created by the presence of turbulent promoter is investigated through CFD. Direct Numerical Simulations (DNS) were performed [36,37 ] as it allows temporal and spatial flow resolution to the smallest (Kolmogorov) turbulent scales without use of any turbulence model. However, computational requirements for doing DNS are quite intense, therefore, all computations were carried on a supercomputer (Shaheen II, KAUST in-house supercomputing facility [38]) using parallel distributed environment on 1024 processors. Full solution of Navier-Stokes equations solution under incompressible flow assumption was performed:

\[ \nabla \cdot \mathbf{u} = 0 \]  \hspace{1cm} (8)

\[ \frac{\partial \mathbf{u}}{\partial t} + (\mathbf{u} \cdot \nabla) \mathbf{u} = -\nabla P + \frac{1}{Re} \nabla^2 \mathbf{u} \]  \hspace{1cm} (9)

where \( \mathbf{u} \) is the velocity field vector (m.s\(^{-1}\)), \( \nabla \) is the three-dimensional space gradient in dimensionless Cartesian coordinates, and \( \rho \) is the fluid density (kg.m\(^{-3}\)). The fluid (here water at 20 °C) was assumed to follow Newtonian behavior. Since the permeate flow (~10 μm/s) is typically very small compared to the cross-flow (in the order of 0.05% per spacer cell), the fiber wall was assumed impermeable, which allows the use of no-slip boundary condition (\( \mathbf{u} = 0 \)). At
the channel inlet, constant flow velocity $U_0$ (corresponding to experimental conditions) was specified normal to the inlet face. The exit of the channel was set as an outlet flow condition [39].

Figure 3: Computer aided design model of the helical turbulent promoter reconstructed using digital measurements. (A) Model inside the computation domain. Two set of helical promoters were reconstructed. $D = 6$ mm corresponds to the case when the promoter is tightly fit with membrane lumen and $D = 5$ mm corresponds to loose fit of the promoter. (B) Mixed 3D tetrahedral and hexahedral mesh generated for space discretization. (C) Isometric cross-section view of boundary layer mesh used in the simulations. The internal diameter of the membrane is 6 mm.

The solution of the system of Eqs. (8) and (9), along with the specified boundary conditions, was carried out on the commercial solver, ANSYS Fluent (ANSYS, Inc., Version 18), with second order space and time discretization using finite-volume approach [39, 40]. The Computer Aided Design (CAD) model for the static turbulence promoter was created using SolidWorks software (Dassault Systèmes, SOLIDWORKS Corporation, Version 2016) by digitally measuring the dimensions. Fig. 3(A) shows the reconstructed model along with the computation domain. The DNS required meshing was achieved by using mix element shapes of the control volumes (Fig. 3(B)). Near the surface, fine mesh was chosen to appropriately resolve the evolving boundary layer, as shown in Fig. 3(C).
The grid independence and the validation of the DNS solver was performed by comparing the shear stress for the case when turbulent promoter is not used with the analytical relations available in literature (presented in Section 4.2, Fig. 11A). DNS results compared quite well with the shear stress magnitude for four inlet velocity conditions. Later using the same resolved computational grid, DNS was performed for the case when turbulent promoter is inserted inside the membrane element. To ensure the correct shear stress resolution on the turbulent promoter, the boundary mesh size was kept the same as on the tubular wall surface and smoothly increases away from the surface. This ensures correct resolution of the hydrodynamics for the static turbulent promoter case as the used mesh size and model parameters are already validated for the case without static turbulence promoter.

4. Results and discussion

4.1. Membrane and cake layer resistances

When \( R_c \) is independent of \( \alpha \), it is a function of operating parameters and membrane characteristics. From Eq. (1), \( R_c \) can be found as a function of driving pressure:

\[
R_c = \frac{\Delta P}{\mu J} - R_m
\]  

Fig. 4, plotted using experimental data, shows the variation of \( R_c \) with applied pressure. It is noticeable that \( R_c \) has almost the same order of magnitude as the membrane resistance \( R_m \), which shows the advantage of the cross-flow filtration mode. In dead-end filtration, \( R_m \) is negligible compared to \( R_c \) [6]. Development of the cake layer is related to the applied pressure and is stabilized by the shear stress induced by cross-flow velocities. The increase of applied pressure enhances the permeation flux and forces more solute towards the membrane surface resulting in an increase of the cake layer thickness and its resistance. This phenomenon is more significant when the feed concentration is higher. Above 3 bars, the increase of \( R_c \) is more significant, from \( 4.10^{12} \text{ m}^{-1} \) to \( 6.5.10^{12} \text{ m}^{-1} \). But, at high velocity and low pressure, the cake resistance is more stabilized. Shear stress on the cake layer is so influential that the properties of the latter are independent of pressure [41]. When the pressure increases, \( R_c \) increases even at high cross-flow velocities.
**Rc** also depends on pressure due to cake compressibility and it is related by an empirical equation [42,43]:

$$R_c = e(\Delta P)^s$$  \hspace{1cm} (11)

where $e$ is a constant related primarily to the shape and size of particles forming the cake layer, and $s$ is the cake compressibility, which can vary from zero (incompressible cake) to a value near one (highly compressible cake). If $s$ is nil, $R_c$ is independent of the pressure; this case is observed for low feed concentrations. If $s = 1$, $R_c$ is proportional to the pressure and the curve passes through the origin; this is possible for highly concentrated solutions.

Mallubhotla and Belfort [44] found values of $s$ varying between 0.61 and 0.71 while testing the compressibility of the deposited foulants on the surface of the membrane. In MF of a bentonite solution using different membranes, Rjimati and Grasmick [43] found values of $s$ and $e$ varying between 1.24 and 1.97, and 13 and 279, respectively. They concluded that this great variation is due to the membrane nature and its permeability. They also found that $R_c$ is independent of Reynolds number ($R_e = \rho ud/\mu$; $d$ is the filtration element internal diameter (m)) and pressure for low concentration solutions, which confirm our approach.

4.2. **Influence of operating parameters on specific cake resistance ($\alpha$)**
Temperature is the main parameter influencing the process that is involved while filtering the OiW emulsion, since the droplets sizes are significantly influenced by temperature variation. Two classes of droplets sizes of about 10 μm and 80 μm were observed at 20 °C and 25 °C, respectively. At 30 °C, the large droplets disappeared completely and the mean droplets size is reduced to the range of 1 μm. These size ranges explain the total rejection of oil droplets by the membrane (mean pore size = 0.02 μm). No oil was found in permeate even at high feed concentration and temperature. The membrane pore size is significantly smaller than the smallest oil droplet and particles present in the solution, which explains the absence of membrane pore adsorption (internal fouling). When suspended solids (SS) having sizes of about 40-50 μm mean size are added to the OiW solution forming a mixed suspension (MS), the particle sizes become significantly higher. Microscopic observations showed that oil droplets agglomerate on the SS particles leading to larger clusters with sizes independent of temperature, leading to higher fluxes compared to those obtained while filtering OiW or particles separately (Fig. 5). Similar phenomenon was observed in previous studies [32, 41]. The influence of using a static turbulence promoter inside the membrane module will be discussed in the next section.

![Figure 5: Flux vs. time (1 bar, 20 °C, 1.2 m/s, 2 g/L oil, 2 g/L SS).](image)

Fig. 6(a) shows that α of the MS remains constant when temperature increases from 20 to 35 °C, because temperature does not have any effect on the particles size. Whereas for the OiW emulsion, α decreases with increasing the temperature which can be attributed to the coalescence
of droplets on the membrane surface. Indeed, the coalescence process decreases $\alpha$ as larger clusters form a stagnant cake layer protecting the membrane from pore clogging (Fig. 6a). Between $20 \, ^\circ C$ and $25 \, ^\circ C$, the decrease is significant; $\alpha$ decreases from $16 \times 10^{-9} \, m^{-2}$ to $9 \times 10^{-9} \, m^{-2}$; while for temperatures higher than $25 \, ^\circ C$, $\alpha$ decreases slowly from $9 \, m^{-2}$ to $6 \, m^{-2}$. The remaining smaller droplets in the suspension do not coalesce and lead to a constant $\alpha$. This phenomenon cannot be observed when the particles size is independent of temperature (SS case). At higher temperature, the deposit formed mainly by small oil droplets is more homogeneous, resulting in a decrease of $\alpha$. Although the smallest droplets are much smaller than the smallest membrane pore size, they tend to penetrate the membrane structure (droplets deformation by the effect of the driving pressure) [45] and could cause partial pore clogging.

![Figure 6](image)

Figure 6: Influence of (a) temperature ($1 \, \text{bar}, 1.2 \, \text{m/s}, 2 \, \text{g/L}$), (b) particle fraction (concentration) ($1 \, \text{bar}, 1.2 \, \text{m/s}, 20 \, ^\circ C$), (c) applied pressure ($1.2 \, \text{m/s}, 20 \, ^\circ C, 2 \, \text{g/L}$), on specific cake resistance ($\alpha$).
Fig. 6(b) shows that $\alpha$ is an inverse function of concentration as the coalescence process is probably favored by the increase of feed water concentration. The lower concentration of particles allows the membrane to run at higher flux. High concentrations have little influence on $R_c$; the deposit is homogeneous and acts as a dynamic membrane which can potentially retain the small particles, generating a reduction of membrane porosity and permeability.
Figure 7: (a) Cake formation in cross-flow filtration mode: the retentate flows parallel to the membrane surface reducing the thickness of the boundary layer; (b) deposition of droplets and particles on the membrane surface at different temperatures showing aggregation of droplets on particles phenomenon (bottom: 30 °C using helical turbulence promoter).

Furthermore, it can be observed that $\alpha$ of the MS deposit remains independent of the applied pressure while it decreases with the increase in pressure for the SS deposit, as shown in Fig. 6(c). This probably results from flocculation of SS on the membrane surface that induces a deposit of larger particles. The same trend is found with OiW droplets deposit alone, but with lower values especially at low pressure, which would suggest that the cake is not compressible. During filtration of OiW emulsion or SS separately, droplets and particles having the same nature and characteristics form a more homogenous and compressible cake layer, which limits the benefit of increasing the pressure and velocity. We notice that we obtained parallel lines for different temperatures. The relatively constant $\alpha$ for the MS could be due to an incompressibility of the cake composed of mixed droplets-particles and a lack of agglomeration in larger clusters under these conditions. In addition, it was found that 2 bars is the critical pressure above which $\alpha$ for MS becomes higher than that of OiW droplets or SS deposits only, which is somewhat paradoxical (Fig. 6(c)). High pressures favor the coalescence of the deposited droplets and diminish the cake specific area. Pressure generates forces which act to stick the deposit on the surface of the membrane and are opposite to the lift forces as well as the turbulent shearing forces that develop due to cross-flow velocity (Fig. 7). The cake thickness is fixed by the balance of these forces. This phenomenon has more effect with the use of a helical turbulence promoter inside the membrane module which generates higher turbulence, creates unsteadiness and lowers $\alpha$ (Fig. 6(c)), resulting in an increased and more stable permeation flux (Fig. 5).

These results confirm the assumption by Kim and Yuan [11] that cake layers composed of relatively large colloids with small roughness can considerably reduce the cake layer resistance, avoiding no-slip boundary conditions on external surfaces of the rough colloids. These results suggest that $\alpha$ variation is significantly influenced by the particles sizes and their nature, and the compressibility of the cake layer, which in its turn is governed by the balance of the pressure, cross-flow velocity and the lift (diffusion) forces. The cake mass per unit area decreases with increasing cross-flow velocity and increases with increase the applied pressure.
In a dead-end filtration study, Sripui et al. [1], Teoh et al. [10] and Kim and Lee [8] found that α increased with pressure, indicating a compressible cake layer. However, in cross-flow filtration mode, this is not applicable as the deposited particles are continuously swept away from the membrane surface lowering the cake resistance and reaching a steady state flux after stabilizing the cake (uniform cake). It is believed that the contradiction of these results is mainly due to the fact that α is determined by a relationship developed for dead-end filtration using cross-flow filtration data, which may not be always coherent.

In another study, Wang et al. [4] also found that α increased with pressure and velocity in cross-flow MF. According to these authors, the main reason is that the larger particles were carried back into the bulk suspension by the effects of pressure and velocity, allowing smaller particles to deposit on the surface of the membrane and leading to a more compact cake layer, which causes the increase of α, which is in agreement with theory. However, as the MS is a complex solution, it is difficult to correlate α with the sizes of formed flocs, which significantly vary with operating parameters and hydrodynamic conditions.

On the other hand, Fig. 8(a) shows that α remains independent of cross-flow velocity, which in its turn stabilizes the deposit without leading to a significant droplets coalescence. α of the OiW emulsion is higher, probably due to the coalescence of smaller droplets making the cake layer more compressible. The deposition of a heterogeneous layer on the membrane surface becomes more homogenous by the conjugated effects of the shear stress and the driving pressure (Fig. 7). The uniformity of the cake layer favored by the shear forces makes it more difficult for the deposited particles to flow back into the bulk solution again, contrary to Wang’s et al. [4] assumption. The particle agglomeration is favored by the increase of concentration. From a hydrodynamic concept, the feed flow through a uniform porous sphere could be characterized by, first, a slow interior flow driven by pressure and second, a fast exterior flow driven by shear stress forces regenerated around the permeable surface [46]. Aggregates of mixed oil and particles assisted in building up a porous cake layer on the membrane surface with low α and subsequently protecting the membrane from severe fouling [47].
The use of a turbulence promoter inside the membrane module has a significant effect on $\alpha$. The presence of static turbulence promoter increases the cross-flow velocity and improves the local hydrodynamic conditions by producing higher vorticity near the surface of the membrane, and maximizes the feed directivity to the membrane surface without significant increase of energy consumption. Figs. 9 (A-C) show the flow velocity magnitude computed through DNS for the case when the helical turbulence promoter fits the internal diameter of membrane perfectly ($D = 6$ mm) and for the case when it is slightly loose ($D = 5$ mm). The computations of both these cases were carried out for the feed flow inlet velocities ($U_o$) ranging from 0.8 m/s to 1.6 m/s as used in experiments. The case when the promoter fits the membrane perfectly, higher velocities at the membrane surface are observed compared to the case when the fitting is slightly loose. The fluid momentum is lost through the opening created between the promoter wall and the membrane.

Figure 8: Influence of (a) cross-flow velocity (1 bar, $20^\circ$C, 2 g/L), (b) flow rate using helical turbulence promoter (1 bar, $20^\circ$C) on specific cake resistance ($\alpha$).
surface for the loose fit promoter, resulting in lower fluid velocities near the membrane surface (Figs. 9C and 9D). Further, for both cases, the fluid flows helically as shown in the subsequent streamtrace plot (Figs. 9E and 9F). The feed fluid is forced to follow the helical path in both cases, however for the loosely fitted promoter, feed fluid leaks along the membrane wall. Therefore, for fitted turbulence promoter the feed flows faster leading to detachment of particles away from the surface.

The helical promoter produces a helical flow and generates turbulence near the membrane surface creating a vortex and enhancing scouring leading to a reduction of the thickness of the deposit by increasing the shear forces against the membrane surface, which explains the increase and rapid stability of the steady-state flux (Fig. 5). Similar flux values at about three-fold lower velocity were achieved when using comparable turbulence promoters [15]. On the other hand, α is inversely proportional to flow rate, as shown in Fig. 8(b). It is noticeable that this phenomenon was not observed in the absence of helical baffles, which explains the role of turbulence promoter in eliminating or significantly reducing the particles flocculation and agglomeration processes due to higher shear forces.

The primary purpose of the turbulence promoter is to enhance surface shear stress to mitigate foulant deposition. For tubular membrane, with no turbulent promoter present, the shear stress θ depends on the energy degradation regime and could be calculated by [48, 49]:

\[
\frac{f}{2} = \frac{\theta}{\varrho u^2} \tag{12}
\]

where \(f/2\) is the friction factor, which is a function of Reynolds number and can be obtained by:

\[
\frac{f}{2} = \frac{8}{R_e} \quad \text{for } R_e \leq 4,000 \tag{13}
\]

\[
\frac{f}{2} = 0.023R_e^{-0.2} \quad \text{for } 5,000 \leq R_e \leq 200,000 \tag{14}
\]
Figure 9: Fluid velocity magnitude (A) $U_o = 0.8 \text{ m/s and } D = 6 \text{ mm}$, (B) $U_o = 1.6 \text{ m/s and } D = 6 \text{ mm}$, (C) $U_o = 1.6 \text{ m/s and } D = 5 \text{ mm}$, (D) $U_o = 0.8 \text{ m/s and } D = 5 \text{ mm}$, (E) Streamtrace plot overlapped with x-velocity component for $U_o = 0.8 \text{ m/s and } D = 5 \text{ mm}$, and (F) Streamtrace plot overlapped with x-velocity component for $U_o = 1.6 \text{ m/s and } D = 6 \text{ mm}$.

For the case when the turbulent promoter is inserted, DNS provides a reasonable estimate on shear stress generated on the wall of the tubular fiber. Figs. 10(A-D) show contours of shear stress at the membrane surface for both tightly and loosely fitted promoter at 0.8 m/s and 1.6 m/s feed velocities. For both cases, the relative minimum and maximum magnitude of shear stress level achieved are similar. More helicity in shear stress profile is maintained for the tightly fitted
promoter, while for loosely fitted promoter it tends to diffuse along the wall due to fluid leakage. However, for tightly fitted promoters, the distribution of shear stress is more uniform, and higher values are achieved along the surface of the membrane for all feed velocities. Therefore, from an application perspective, it is desirable to have the promoter perfectly fit inside the lumen of the tubular membrane to achieve desirable cleaning performance.

Finally, the average shear stress is computed as depicted in Fig. 11A to quantify the cleaning effect. The average shear stress computed by DNS for the case when no promoter is also presented as validation of solver. The predictions are reasonably accurate, and the trend follows the analytical predictions. For promoter cases, roughly 3 times higher average shear stress for the tightly fitted promoter, while roughly 2.5 times higher shear stress is predicted for the loosely fitted promoter. In addition, the trends are linear with respect to feed velocity, indicating that at higher feed flow rates the turbulent promoter will be more effective. The effectiveness of helical baffle can also be represented by plotting experimental permeate flux as a function of analytical shear stress, as shown in Fig. 11(b). It shows that a plateau is reached at about 4 Pascal, but when the helical baffle is installed the flux roughly becomes proportional to the shear stress and its value is higher, which confirms our approach.
5. Conclusions

Specific cake resistance ($\alpha$) variation, and its influence on steady state permeation flux, is used as an operational indicator to quantify membrane fouling in cross-flow UF of different solutions containing oil-in water droplets, suspended solids, and a mixed suspension. It was found that $\alpha$ depends strongly on particles and droplets sizes, pressure, feed concentration, in some cases on temperature, and is almost independent of cross-flow velocity due to shearing forces. $\alpha$ is also significantly influenced by the cake nature and its compressibility. However, $\alpha$ is inversely proportional to the feed flow rate when a turbulence promoter is used. Cross-flow membrane processes often rely on the presence of shears to reduce the accumulation of foulants from the surface of the membrane. The turbulence promoter (modified helical baffles type) used in this study enhanced the flux significantly without limiting the process or mass transfer as demonstrated by computational studies. Coalescence phenomenon of the deposited droplets, flocculation of suspended solids and aggregation of droplets on solids explain the variations of $\alpha$ with the operating parameters. An increase of pressure favors the coalescence of droplets and particles in suspension. Aggregation of droplets on solids leads $\alpha$ to become independent of pressure and
temperature. Results also showed that the use of dead-end filtration models for cross-flow filtration experiments is inadequate as it was demonstrated by the different $\alpha$ variations (in cross-flow) compared to other studies using dead-end mode.

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**Nomenclature and abbreviations**

*List of symbols*

- $A$ Membrane surface area ($m^2$)
- $a_g$ Grain specific surface area: defined either by surface area divided by mass ($m^2.kg^{-1}$) or surface area divided by volume ($m^2.m^{-3}$ or $m^{-1}$)
- $C_o$ Mass of accumulated matter per unit volume of permeate ($kg.m^{-3}$)
- $CP$ Concentration polarization
- $d$ Filtration element internal diameter ($m$)
- $d_p$ Particle diameter ($m$)
- $e$ Constant related primarily to the size and shape of the particles of the cake layer
- $f, g$ Constants
- $f/2$ Friction factor
- $h_k$ Kozeny constant
- $J$ Permeation flux ($L.m^{-2}.h^{-1}$)
- $K$ Medium permeability ($m^2$)
- $L$ Cake length ($m$)
- $L_p$ Pore length or membrane thickness ($m$)
- $MF$ Microfiltration
- $MS$ Mixed suspension
- $m_c$ Cake mass per unit area ($kg.m^{-2}$)
- $n$ Number of mono-cellular layers
### NF Nanofiltration

### OiW Oil-in water

### $R_e$ Reynolds number

### $R_c$ Cake resistance – reversible and/or irreversible fouling (m$^{-1}$)

### $R_m$ Membrane resistance (m$^{-1}$)

### $P_p$ Pore density: is the number of pores per unit of membrane area (m$^{-2}$)

### $r_p$ Pore radius (m)

### $r_{pr}$ Particle radius (m)

### SS Suspended solids

### $s$ Cake compressibility

### $t$ Time (s)

### $u$ Cross-flow velocity (m.s$^{-1}$)

### UF Ultrafiltration

### $V$ Filtrate volume (m$^3$)

### $x_o$ Volume fraction occupied by particles in the feed suspension (-)

### $x_c$ Cake fraction volume

### Greek symbols

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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<tbody>
<tr>
<td>$\alpha$</td>
<td>Specific cake resistance (m.kg$^{-1}$) or per unit length of deposit (m$^{-2}$)</td>
</tr>
<tr>
<td>$\delta$</td>
<td>Cake layer thickness (m)</td>
</tr>
<tr>
<td>$\Delta P$</td>
<td>Applied pressure (Pa)</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>Cake porosity (-)</td>
</tr>
<tr>
<td>$\mu$</td>
<td>Dynamic viscosity (Pa.s) or (kg.m$^{-1}$.s$^{-1}$)</td>
</tr>
<tr>
<td>$\nabla$</td>
<td>3-D space gradient in dimensionless Cartesian coordinates</td>
</tr>
<tr>
<td>$\tau$</td>
<td>Tortuosity factor</td>
</tr>
<tr>
<td>$\vartheta$</td>
<td>Shear stress (Pa)</td>
</tr>
<tr>
<td>$\rho$</td>
<td>Liquid density (kg.m$^{-3}$)</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>Particles density (kg.m$^{-3}$)</td>
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</tbody>
</table>
References


Graphical abstract
Highlights

- Specific cake resistance ($\alpha$) variation and its influence on UF performance are investigated
- The influence of operating parameters on $\alpha$ is discussed
- A helical-type turbulence promoter is used to enhance the flux by decreasing $\alpha$
- CFD results explained the hydrodynamics behind flux improvement
- $\alpha$ could be considered as a reliable operational indicator to quantify membrane fouling
Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:
CRediT author statement

Noreddine Ghaffour: Helical-type turbulence promoter design, fabrication and testing, Methodology, Analysis, Paper writing and Editing, Final reviewing, Supervision. Adnan Qamar: Modeling and simulation, Data validation, Analysis, Writing, Reviewing and Editing.