FLOW BEHAVIOUR IN A MEMBRANE CROSS-FLOW FILTRATION CELL: EXPERIMENTAL OBSERVATIONS AND CFD MODELLING
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ABSTRACT

This paper presents experimental data and CFD-simulation results concerning the flow behaviour during cross-flow nanofiltration applied for concentrating antioxidants such as polyphenols and flavonoids from extracts of natural products. The effects of the cross-flow velocity and solute concentration on the permeate flux behaviour are discussed. The first one is studied numerically and discussed in rapport with existing experimental observations in literature. The CFD modelling is focused on an experimental flat-sheet membrane cell’s hydrodynamics – velocity and shear stress distribution on the membrane surface. The geometry of the cell includes tangential orientation of the feed pipe, resulting in a swirling flow in the cell, tangential to the membrane surface everywhere except for a very small region at the centre. The increase in the feed flow rate leads to greater shear forces to the membrane surface. This results in higher permeate flux attributed to reduced concentration polarization.

The effect of the solute concentration is studied experimentally using a 4 flat-sheet membrane rig for cross-flow filtration in both concentration and full recycle mode of operation. The permeate flux decline during nanofiltration of ethanolic extracts from Sideritis ssp. L. with membranes Duramem (molecular mass cut-off, MWCO 300 and 500 Da) is presented. It is related to concentration change and formation of a concentration polarization layer at the membrane–liquid interface, the effect being lower in case of better hydrodynamic conditions and lower feed concentrations.

Keywords: cross-flow nanofiltration, flow behaviour, CFD modelling, extracts of natural products, experimental flat-sheet membrane cell.

INTRODUCTION

Membranes are used on a large scale in separation technologies allowing separation at ambient temperature without chemical reaction. This is especially important in concentration of organic extracts of bioactive substances from medicinal plants, the subject area of the present work. It focuses on organic solvent nanofiltration (OSN), which has advantages for multicomponent systems containing sensitive to elevated temperatures components and allows for the regeneration of the organic solvent. The OSN is performed on laboratory scale in one of the two modes: dead-end filtration, where the entire solution under applied pressure is directed normally to the membrane surface and cross-flow filtration, where the flow direction is parallel to the membrane surface. The first mode is usually preferred for testing membranes because of the small volumes of solvent required. The second one is more consistent with the industrial scale. The majority of industrial membrane modules operate in a cross flow. A typical example is the spirally wound membrane module. It is employed predominantly in reverse osmosis (RO) and nanofiltration (NF), but also in ultrafiltration (UF) and microfiltration (MF).

The present investigation is incited by the increasing interest in membrane separation/concentration of biologically active compounds in extracts from medicinal plants with organic solvents. The OSN proves
to be less predictable than with aqueous solutions. The multi-component composition of the natural extracts leads to specific problems with fouling, including concentration polarization and cake layer resistance. This study is focused on two factors - solute concentration and cross-flow velocity. They are recognized to have close relation to the concentration polarization in cross-flow OSN. The aim of the research is to understand and control that phenomenon. The effect of the solute concentration is studied by measuring the volume fluxes and concentrations in a flat-sheet cross-flow filtration cell with a specific tangential inlet design. The experimental method is supplemented by numerical techniques for assessing the role of the cross-flow pattern in the cell.

The effect of the solute concentration is usually observed as a flux decline during nanofiltration, related to an additional membrane resistance. The membrane solute rejection leads to the development of a boundary layer in which the concentration of the rejected solutes exceeds the one in the bulk solution. This leads to increased osmotic pressure and reduced driving pressure difference across the membrane, resulting in reduced permeate flux. In case of multicomponent solutions, where compounds with very different solubility are present, saturation conditions may be reached at higher concentration with a subsequent cake formation, respectively fouling or scaling of the membrane. Regarding the concentration effect on rejection, the observations reported in the literature for OSN are more contradictory. Here it is important to account for the true rejections, concerning the increased concentration at the surface of the membrane in comparison with the bulk retentate concentration.

The cross-flow velocity is an important operating variable of the membrane separation process together with the trans-membrane pressure and feed concentration. It is usually commented in connection with tangential flow near the membrane surface in flat-sheet [1] or spirally wound [2] membrane modules. In batch type stirred filtration cells the corresponding variable studied is the stirrer’s speed [3]. A number of experimental investigations has proved the positive effect of the higher cross-flow velocity on the permeate flow during NF [1, 2], RO [1], UF [4], as well as MF [5]. The studied solutes are organic species like dyes in textile wastewater purification [2], bioactive compounds in plant extracts concentration [1, 3], fruit juices [4, 5] or sugar remelt syrup [6] clarification. The range of variation of the cross-flow velocity from different sources is presented in Table 1. The impact of the cross-flow velocity on permeate flux and solute rejection depends on the system solute-membrane, including component-ness of the solution, chemical composition, viscosity, membrane-solute interactions like adsorption, external or internal pore blocking etc. While the aqueous solutions are mostly studied, the reported investigations on organic solvents [7] are scarce. The observed increase in the permeate flow with the increase in the cross-flow velocity is usually significant [3], ranging from nonlinear [5], to nearly linear [2] or tending to constant [1] (i.e. a limiting value of the cross-flow velocity is reached; afterwards the latter has no more influence). There are few examples, when a minor effect of the cross-flow velocity is found [4] due to the narrow range of variation

![Fig. 1. Flux vs time variation with membrane Duramem 500, $C_f = 0.83$ mg ml$^{-1}$; Continuous full recirculation mode: (1), (3), (5); Concentration mode: (2), (4); Permeate to feed volume ratio ($V_p/V_f$): 0 (1); 0.28 (3); 0.57 (5); Average permeate flux ($J_p$) in L m$^{-2}$ h$^{-1}$: 22 (1); 16.78 (2); 17.3 (3); 13.98 (4); 13.11 (5).](image1)

![Fig. 2. Cumulative permeate volume vs time plot at different degrees of concentration ($V_p/V_f$).](image2)
of the latter. Increased cross-flow velocity improves the permeate flow, creating better hydrodynamic conditions in the vicinity of the membrane surface. The forced convection imposed by the cross-flow velocity generates turbulence, improves mixing and reduces the degree of solute-concentration polarization. The explanation can be searched in the increased shear stress at the membrane surface, which enhances the rate of removal of accumulated or deposited solutes responsible for fouling and the respective flux decay. Speaking in terms of fouling resistances [3, 5, 6], they are gradually decreased with higher cross-flow velocities, which is consistent with the observed increase in permeate flux. The gel-layer and concentration polarization resistance are found as power functions of the cross-flow Reynolds number [3]. As a result a reduced permeate concentration and improved mass transfer in the system is usually observed, namely an increasing permeate side solute mass transfer coefficient as a function of the effective cross-flow Reynolds number [2]. The influence of the cross-flow velocity on the rejection behaviour is usually less important - either no change [8] or slight increase [1] have been observed and attributed to the reduced concentration polarization.

The CFD simulation is a powerful tool in the investigations on membrane filtration. A lot of approaches and models have been offered and exploited in literature, reviewed in [9]. A frequent approach is modelling the flow in the retentate channel by assuming that the membrane is impermeable. The purpose of the simulation is to enhance membrane efficiency and minimize concentration polarization and fouling by increasing the shear stresses on the membrane surface. Recent studies include effects created by various geometries [10, 11], spacers [12, 13], gas sparging, baffles, etc. The present

Fig. 3. Flux \( J_p \) vs concentration \( C_r \) dependence with membrane Duramem 500.

Fig. 4. Average flux \( J_p \) and rejections \( R \) obtained at different feed concentrations \( C_f \).
The work applies this approach to a test flat-sheet cross-flow filtration cell with a specific tangential inlet [7] expected to enhance the cell flow efficiency.

**EXPERIMENTAL**

**Physical experiment**

Experimental observations on flux behaviour were performed in a cross-flow filtration of polyphenols from ethanolic extract of Sideritis ssp. (Mursalski tea). A METcell cross-flow system (Membrane Extraction Technology Ltd., UK) with four connected in series flat-sheet cells (membrane area 54 cm² each) was used at constant flow rate of 1.2 L min⁻¹ and transmembrane pressure of 20 bar. Duramem 500 membrane was used, details on the experimental set-up being given in [14]. The membrane was subjected to previous adaptation with the solvent. The initial sharp flux decline during nanofiltration of real extracts was excluded from the discussion (about 25 min, see Fig. 1). The permeate flux $J$ (L m⁻² h⁻¹) was obtained from the permeate volume $V_p$, determined at various times $t$:

$$J = \frac{V_p}{A \cdot t}$$  \hspace{1cm} (1)

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Table 1. Literature sources with cross-flow velocity effect on membrane filtration.

<table>
<thead>
<tr>
<th>Membrane process</th>
<th>Membrane module</th>
<th>Cross-flow velocity</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>NF</td>
<td>Spiral</td>
<td>0.5 - 2 m s⁻¹</td>
<td>[2]</td>
</tr>
<tr>
<td>NF</td>
<td>Flat-sheet</td>
<td>0.5 - 3.2 m s⁻¹</td>
<td>[1]</td>
</tr>
<tr>
<td>RO</td>
<td></td>
<td>0.77 - 1.25 m s⁻¹</td>
<td></td>
</tr>
<tr>
<td>MF</td>
<td>Flat-sheet</td>
<td>0.77 - 1.25 m s⁻¹</td>
<td>[5]</td>
</tr>
<tr>
<td>UF</td>
<td>Hollow fiber</td>
<td>10 - 20 L h⁻¹</td>
<td>[4]</td>
</tr>
<tr>
<td>UF</td>
<td>Flat-sheet – tangential</td>
<td>0.07 - 0.18 m s⁻¹</td>
<td>[3]</td>
</tr>
<tr>
<td>NF</td>
<td>Stirred cell (batch)</td>
<td>3.0 - 6.0 m s⁻¹</td>
<td>[6]</td>
</tr>
</tbody>
</table>


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Fig. 5. A flat-sheet cross-flow filtration cell.
Here A is the membrane area.

Solute rejections were evaluated based on the initial feed concentration \( C_f \) and the permeate concentrations \( C_p \) of total polyphenols:

\[
R = \left( 1 - \frac{C_p}{C_f} \right) \times 100 \% \tag{2}
\]

The total phenolic content was determined spectrophotometrically [15].

Experiments were performed in concentration mode (increasing retentate concentrations), as well as in constant concentration mode (full recycle of both permeate and retentate).

**Numerical simulation**

The effect of the cross-flow velocity was studied in the present work by CFD modelling of the feed flow in the filtration cell.

The geometry and meshing of the flat circular cell is shown in Fig. 5. A detailed scheme of the cell is given in [7]. The solution enters the cell tangentially from the side wall and exits the cell from the top centre, providing turbulent hydrodynamic conditions. The dimensions of the cell are: diameter of the inlet and outlet \( d_{\text{in/out}} = 3 \text{ mm} \); membrane area, \( A = 54 \text{ cm}^2 \); cell diameter \( D_{\text{cell}} = 82.9 \text{ mm} \); and height of the retentate channel, \( h_{\text{cell}} = 5 \text{ mm} \).

The following limitations were presumed:

- Steady-state flow was assumed in the 3D computational domain;
- The turbulent flow was represented by Reynolds-averaged Navier–Stokes (RANS) equations of continuity and momentum in Cartesian tensor form for incompressible Newtonian fluid [16]:

\[
\frac{\partial}{\partial x_i} (\rho u_i) = 0 \tag{3}
\]

\[
\frac{\partial}{\partial x_j} (\rho u_i u_j) = -\frac{\partial p}{\partial x_i} + \frac{\partial}{\partial x_j} \left[ \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_k}{\partial x_k} \right) \right] + \frac{\partial}{\partial x_j} \left( -\rho \overline{u'_i u'_j} \right), \tag{4}
\]

where \( u_i \) indicates time-averaged velocity component, \( p \) is pressure, \( \rho \) is liquid density, \( \mu \) stands for liquid dynamic viscosity, and \( \overline{\rho u'_i u'_j} \) represents Reynolds stresses.

- Closure of RANS equations was ensured by the realizable “k-ε” model of turbulence [16];

The equations were solved using a CFD code FLUENT based on finite volume technique:

- The fluid was water, which was assumed to be incompressible and isothermal with constant density and viscosity;
- No-permeation and no-slip conditions were assumed at the membrane walls;
- Velocity was specified at the inlet;
- At the outlet, a pressure boundary condition

![Fig. 6. Contour plots of velocity magnitude (m s\(^{-1}\)) and velocity magnitude vectors in a plane 1 mm above the membrane surface, feed flow rate 1.2 L min\(^{-1}\).](image-url)
was assumes and p = 0 Pa was set for computational purposes.

In conformity with the experimental observations, it was accepted that the permeate flow rate was negligible compared to the feed flow rate and did not distort significantly the flow field in the filtration cell. Therefore, the membrane surface was described as impermeable wall. The realizable “k-ε” model of turbulence was selected because it had shown improvement in comparison to the standard one for streamline curvature and vortices.

Mesh independency of the results was studied. Area-weighted average shear stress (Pa) at the membrane surface was determined with an increasing number of mesh elements. At the condition of scaled residuals of $10^{-4}$, a good compromise between computational time and parameter deviations was found for 160 000 mesh elements.

RESULTS AND DISCUSSION

Effect of solute concentration

Experimental data illustrating the effect of the solute concentration on the flux behaviour in cross-flow filtration of polyphenols from ethanolic extract of Sideritis ssp. is given in Figs. 1 to 3.

The plot of permeate flux vs. time of filtration in Fig. 1 concerns nanofiltration with consecutive constant or increasing retentate concentration of total phenolics (in mg ml$^{-1}$). The permeate flux decreases slowly with time, which is consistent with the formation of a concentration polarization layer and the increase in the concentration. In the constant concentration case, a full recycle of permeate and retentate was maintained, while in the second case the volume ratio permeate to feed ($V_p/V_f$) was increased (0 to 0.28 for the first and 0.28 to 0.57 for the second concentration period). The averaged permeate flux during the concentration period was evaluated from the cumulative volume vs. time plot with high correlation coefficient (Fig. 2).

Fig. 3 illustrates the flux vs. concentration dependence. The cross-flow filtration data at increasing solute concentrations on the retentate side are presented there. Decreased permeate fluxes and practically constant solute rejections are observed, as can be seen from Fig. 4.

Effect of cross-flow velocity

Contour plots of velocity magnitude (m s$^{-1}$) and velocity magnitude vectors in a plane 1 mm above the membrane are obtained at various fluid flow rates. Fig. 6 illustrates the case of feed flow rate of 1.2 L min$^{-1}$.

The following effects can be inferred:

- As shown by the velocity vectors that sweep the membrane surface, the tangential orientation of the feed inlet results in inner-cell swirling flow;
The flow is tangential to the membrane surface everywhere, except for a very small region at the centre. The simulation results in Fig. 6 reveal that the tangential inlet successfully serves its purpose to create a vortex in the cell without moving parts. The obtained flow pattern indicates that the inlet design ensures an effective cross flow, which hinders the development of a polarization layer.

The simulations demonstrate the effect of the feed flow rate on the shear forces to the membrane surface. Contour plots of the shear stress $\tau_{ij}$ at the membrane surface, represented in Eq. (4) by the expression

$$\tau_{ij} = \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right),$$

are shown in Fig. 7. The Reynolds number (Re) is determined at the cell inlet.

Fig. 8 presents a four-fold rise in the calculated average wall shear stress, when there is an increase in the flow rate from 40 L h$^{-1}$ to 150 L h$^{-1}$. The plot in Fig. 8 corresponds well to the experimental data in Fig. 9 [7], which shows that the permeate flux increases with the feed flow rate in OSN with the test cell described above at various TOABr (tetracyclammonium bromide) concentrations in toluene solutions.

CONCLUSIONS

A cross-flow filtration cell with flat membrane and tangential inlet flow is studied and the effects of the retentate flow rate and the solute concentration on the permeate flux behaviour are resolved. The permeate flux is observed to decrease with the increase in concentration on the retentate side, which is an expected result related to the concentration change at the membrane-liquid interface and the possible formation of a concentration polarization layer. As shown by experiments and CFD simulation, the effect can be reduced by the use of lower feed concentrations or by improving the hydrodynamic conditions. The numerical calculations confirm that the increased cross-flow velocity improves the permeate flow, creating better hydrodynamic conditions in the vicinity of the membrane surface. The obtained flow pattern in the cell points at the following:

- The tangential orientation of the feed inlet results in a swirling flow in the cell, which sweeps the membrane surface, ensuring without any moving parts tangential flow near the membrane, except for a very small region at the centre.
- The obtained flow distribution, as correlated with the experimental separation rate, confirms that the rise in the permeate flux with the increase in the feed flow rate is related to the increase in the shear forces at the membrane surface, which are likely to reduce the concentration polarization.

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