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PII: S0376-7388(14)00032-5
DOI: <http://dx.doi.org/10.1016/j.memsci.2014.01.019>
Reference: MEMSCI12673

To appear in: *Journal of Membrane Science*

Received date: 7 October 2013
Revised date: 19 December 2013
Accepted date: 7 January 2014

Cite this article as: R. Salcedo-Díaz, P. García-Algado, M. García-Rodríguez, J. Fernández-Sempere, F. Ruiz-Beviá, Visualization And Modelling Of The Polarization Layer In Crossflow Reverse Osmosis In A Slit-Type Channel, *Journal of Membrane Science*, <http://dx.doi.org/10.1016/j.memsci.2014.01.019>

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VISUALIZATION AND MODELLING OF THE POLARIZATION LAYER IN CROSSFLOW REVERSE OSMOSIS IN A SLIT-TYPE CHANNEL

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Abstract

The evolution of the concentration polarization layer during crossflow reverse osmosis in a slit channel has been studied. Digital Holographic Interferometry allows visualizing the polarization layer as an interference fringe pattern. An especial module with four windows has been designed to see the development of the polarization layer along the membrane channel. Several experiments with a constant transmembrane pressure of 6 bar, three different feed concentrations (3, 6 and 9 kg/m³) and three different Re (13, 38 and 111) had been carried out. The process has been modelled simulating the experimental conditions. The computed results were found to be consistent with the experimental ones. All the experiments show a continuous increase of the polarization layer along the channel, regardless of the crossflow velocity, except near the outlet of the cell due to an edge effect. This increase in the polarization layer is greater at lower Re, although it does not influence very much the permeate flux. In contrast, what substantially affects the permeate flux, much more than Re number, is the feed concentration due to its osmotic pressure.

Keywords:

Concentration polarization; Crossflow Reverse osmosis; Digital Holographic Interferometry; CFD Modelling; Polarization layer evolution

1. Introduction

Rejection of salt ions at the membrane surface in reverse osmosis processes (RO) leads to an increase in solute concentration near the membrane. This phenomenon is called concentration polarization (CP) and is one of the most important factors influencing the performance of membrane separation processes [1]. Concentration polarization is the primary reason for flux decline during the initial period of a

membrane separation, due to an osmotic pressure raise. In addition, CP can be also the precursor of subsequent fouling mechanisms in RO [2,3]. Knowledge and prediction of the concentration polarization effects, therefore, is crucial for designing reverse osmosis processes, predicting their performance, and especially for understanding the surface fouling phenomena [2,4,5].

The development of the CP layer in cross-flow RO is a complex process that is influenced by several factors such as solute properties, membrane properties and hydrodynamics (flow conditions, pressure, geometry of the channel, etc.) [6,7]. To properly predict the phenomena, it is necessary an accurate description of the variables involved in the CP development, although there is still a debate to state the most appropriate way to predict the concentration polarization in RO and in other membrane separation processes [4].

In a RO process, concentration polarization is coupled with permeate flux: CP is induced by permeate flux, which brings solute to the membrane but, at the same time, the permeate flux is limited by the resistance due to CP [4,6]. Besides this statement, in crossflow RO appears another fact that must be considered. As a result of solute continuous entrance to the membrane channel and solute rejection and accumulation, the concentration polarization layer grows gradually along the filtration channel [5,6,8,9].

Sablani et al. [8] made a critical review of works about concentration polarization in ultrafiltration and RO. Most of the papers were basically focused on theoretical studies or computational simulation of those processes. More recent studies have followed the same way and thus there is a lack of experimental research measuring the CP layer. The theoretical studies have made attempts to develop mathematical models for predicting the CP level in a membrane system. Some researchers use the classical film theory, which requires the knowledge of a mass-transfer coefficient, using a model that considers the mass transfer equations in the proximity of the membrane surface. Some of them assume a uniform solute concentration and permeate flux over the membrane surface [10] but others consider these variables as channel length dependent [6,11,12].

During the past decades the computational simulation of membrane processes became very popular due to the increasing computational power that allowed solving the mass transfer equations coupled with the governing transport equations by using numerical methods. Computational Fluid Dynamics (CFD) provides a powerful tool to determine the flow pattern adjacent to the membrane surface. A comprehensive review about the CFD methods applied to membrane processes has been made by Ghidossi et al. (2006) [13]. Most studies were made under laminar conditions since most of the membrane processes work under these conditions [14-17]. The slit-type configuration represents the basic model to characterize the flow in industrial membrane modules, thus restricting the flow to laminar conditions and causing severe problems of CP. Geraldes et al. [18] proposed a correlation between the concentration and the hydrodynamic boundary layer thickness which gives an insight into the

mechanisms involved in the growth of the boundary layers in the nanofiltration (NF) processes. Wiley and Fletcher [19] proposed a general purpose CFD model of concentration polarization and fluid flow. The model was extended to examine buoyancy effects in the salt water system under reverse osmosis conditions [20]. All these models show a trend towards greater completeness intending to enhance the performance of membrane separation processes.

Therefore, the accurate characterization of the build up and evolution of the CP layer is essential to fully understand the fouling mechanisms in RO processes. Different techniques for in situ monitoring the polarization phenomena in a membrane system have been developed over the years [21]. However, only a few papers can be found in which the concentration polarization layer is measured directly.

Interferometric techniques are one of those techniques cited by Chen et al. [21] that can be used to fully observe the CP. Digital Holographic Interferometry (DHI) has been used in this research and was previously applied to the study of the CP layer [22,23]. It is an interferometric technique very similar to classical Holographic Interferometry previously used [24-27] but replacing the holographic plate, which needs to be photographically developed, by the CCD chip of a video camera [28]. The interferometric techniques are based in the appearance of interference fringes related to changes in the refractive index of an object where a laser beam has passed through. Concentration polarization entails a change in the refractive index near the membrane surface. Therefore, the comparison of different states of the object studied (i.e. different images captured by the CCD) will cause the appearance of interference fringes, which will allow the visualization of the CP layer.

Both aforementioned aspects concerning concentration polarization in crossflow reverse osmosis (relationship between permeate flux and CP, development of the polarization layer along the membrane channel) are studied in this paper. Moreover, the use of an interferometric technique as DHI is a further step forward since it allows direct observation and measurement of the concentration profile within the concentration polarization layer as well as its evolution along the channel. This important information about the polarization phenomena notably improves those analytical works that use other experimental data (bulk concentration, flux and concentration of permeate, etc.).

This experimental research completes a previous one where concentration profiles and permeate fluxes in crossflow RO were studied [22]. A new longer module has been used to visualize the built up of the polarization layer along the membrane channel. The new module has four windows along the channel thus allowing the visualization of the concentration profiles at different locations of the membrane channel. The results obtained showed the tight relationship between CP, the permeate flux, the crossflow velocity and the development of the polarized layer along the permeation channel. The experimental conditions have also been simulated using a CFD model, developed to compute the permeate flux and the

concentration profiles in order to check the evolution of the polarization layer along the membrane channel. The results predicted by the simulations are in reasonable agreement with the experimental ones.

2. Experimental

2.1. Experimental set-up

Two different systems are linked together in the experimental assembly: the optical set-up for DHI and the reverse osmosis system. Both systems are the same to that explained in a previous paper [22]. These two assemblies are coupled on the same work table, being the RO module the common element.

The RO cell is specially designed to carry out the crossflow RO process satisfying the Holographic Interferometry requirements. The module is similar to that described in a previous paper [22], but with some marked changes to allow the study of the evolution of the concentration polarization layer along the permeate channel. The main difference is its size, more than twice longer than the previously used module. The new one consists also of two independent parts connected by bolted joints (Figure 1). The top part is a methacrylate cell, coated with a stainless steel housing to ensure physical stability. Pressure applied in the RO experiments could deform the transparent methacrylate cell, causing the appearance of interference fringes spurious to the concentration polarization. The stainless steel housing was used to avoid this possibility. The housing was provided with four circular windows, thus allowing the membrane surface to be visualized at different positions along the membrane channel.

The connection between the module and the reverse osmosis system is through a stainless steel tube of 4 mm of diameter. As the section of the tube and the channel are very different, an edge effect can disturb the flow through the module, especially in the windows closest to the inlet and outlet sides (windows one and four).

Figure 1. RO module

The membrane is placed in the bottom part, made of Teflon, and is fixed when both parts of the module are joined together, getting tightened between them. A porous plastic piece located below the membrane acts as a mechanic support. To ensure the correct sealed of the cell and prevent leakage, a silicone joint under the membrane is placed, surrounding the porous plastic piece. A 3 mm diameter hole in the center of the bottom part of the cell gathers together all the permeate, which leaves the module and then passes through a conductimeter.

The dimensions of the active membrane are 220 x 15 mm, with an effective area of 33 cm², and the channel height is 4.3 mm.

2.2. Materials

The membrane used is a polyamide thin film membrane, Filmtec XLE - 2540 from Dow. Suitable pieces for the size of the module (220 x 15 mm) were cut from the entire membrane. Each piece of membrane was used for several experiments. After each experiment, the cell was washed with distilled water until the permeate flux of water was recovered. When a decrease in the permeate flux of water was observed, the piece of membrane was replaced.

The experiments were carried out using solutions of Na₂SO₄ (Merck). Different feed concentrations (C₀), in the range of 3 – 9 kg/m³ were used to study the polarization layer in crossflow reverse osmosis. Physical properties (diffusion coefficient, density and osmotic pressure) of the solute solutions were obtained from the literature [22].

$$D \text{ (m}^2\text{/s)} = -3.9 \times 10^{-12} C \text{ (kg/m}^3\text{)} + 1.16 \times 10^{-9}$$

$$\rho \text{ (kg/m}^3\text{)} = 9.80 C \text{ (kg/m}^3\text{)} + 997.1$$

$$P \text{ (atm)} = 0.516 C \text{ (kg/m}^3\text{)}$$

Permeability and rejection of the membrane, Filmtec XLE-2540, were experimentally obtained.

2.3. Experimental methodology

The first step was to check the performance of the membrane. Water permeate flux (J_w) was measured before each experiment, checking the flow was correct. To clean the membrane, several washes with distilled water were needed; firstly, passing through the module in cross flow and then, applying a pressure of 6 bar, which was the pressure used for all the experiments. After this cleaning process, if the water permeate flux decreases more than a 10% respect to the last experiment performed, it was considered unacceptable and the piece of membrane was replaced.

Once the permeate flux was recovered, the optical set-up was properly aligned with one of the four windows of the cell and it was proceeded to capture the hologram, with water circulating in the RO system at the working pressure (6 bar). This hologram will be used as the reference state of the system.

Later, the module was emptied and the sodium sulphate solution was introduced into the system. Finally, the DHI program was initialized and, at the same time, the pump started up and both pressure and CF velocity were fixed.

When pressure is applied, a convective flux of solute to the membrane surface begins, thereby causing the accumulation of the solute next to the membrane surface. The increase of the concentration of the solution adjacent to the membrane surface changes its refractive index. The comparison between images corresponding to the actual state and the reference state (the hologram) causes the appearance of interference fringes. The methodology to evaluate these interferograms has been explained in a previous paper [22].

Each interference fringe in the interferogram corresponds to a certain concentration change in the solution and thus, the appearance of fringes will mean a progressive change of the solution concentration. The value of this concentration change depends on the relation between the concentration and the refractive index. Therefore, the measurement of the distance to the membrane of each fringe allows obtaining the concentration profiles in the polarization layer [29]. Figure 2 shows a scheme about how the fringes are converted into the corresponding concentration profiles.

Figure 2. Concentration profile obtained from an interferogram

The entire experiment lasted 75 minutes, using three different crossflow velocities during 25 minutes each one. The CF velocities used were 0.2, 0.7 and 2 cm/s, corresponding to a Re of 13, 38 and 111 respectively. Re number has been calculated using the hydraulic diameter as a characteristic length, defined by $D_h = 2h \cdot w / (h + w)$, where h is the height of the channel (4.3 mm) and w is its width (15 mm). The experiments began with the highest velocity (2 cm/s) and remained so for 25 minutes. After this

period the velocity was decreased (0.7 and 0.2 cm/s) and the process was repeated. Each part with a different CF rate was recorded separately.

Besides the CF rate, two more variables were studied: initial concentration (C_0) and distance from the entrance (x). Three values of initial concentration: 3, 6 and 9 kg/m³ and four distances from the entrance: 30, 80, 140, 190 mm (corresponding to each window of the module) were used. Therefore, one feed concentration and one distance were combined in each experiment with all CF velocities, used in downward way, as it has been explained. Each possible variables combination (C_0 and x) was repeated thrice to verify the reproducibility of the results.

Permeate flux and concentration were continuously measured during each experiment acquiring weight and conductivity data of the permeate solution. Permeate flux was calculated from the slope of the weight curve, and the concentration of the permeate was calculated from the relationship between its conductivity and concentration, experimentally measured [22].

$$C \text{ (kg/m}^3\text{)} = 8.15 \cdot 10^{-4} \mu \text{ (}\mu\text{S)} - 1.87 \cdot 10^{-3}$$

3. Numerical simulation

3.1. Mathematical model

The steady-state concentration polarization phenomenon in a crossflow RO process is often described by the one dimensional convection - diffusion equation (eq.(1)), coupled with the osmotic pressure law for the permeate flux calculation (eq.(2)).

$$JC_m + D \left(\frac{dC}{dy} \right)_{y=0} = JC_p \quad (1)$$

$$J = k_m (\Delta P - \Delta \pi) \quad (2)$$

K_m is assumed to be a constant based on the permeation flux of pure solvent.

The integration of eq. (1) results in the well known film theory,

$$\frac{C_m - C_p}{C_o - C_p} = \exp \left(\frac{J\delta}{D} \right) = \exp \left(\frac{J}{k} \right) \quad (3)$$

the term of the left hand side of eq. (3) is defined as the concentration polarization modulus.

This analytical model neglects axial solute convection near the membrane surface assuming that it does not influence the boundary layer thickness. This is the key limitation of this kind of models since the performance of the process is linked to the fluid movement through the cell volume. The model should take into account the hydrodynamics of the system, so solute mass balance and momentum transport

equations must be solved simultaneously, since the permeate velocity and the concentration at the membrane surface are interrelated.

In a steady-state, the governing equations for a two-dimensional incompressible laminar flow in the feed channel are the equations for the conservation of mass, momentum and solute mass fraction. These equations in 2D Cartesian coordinates are as follows:

Global continuity equation

$$\frac{\partial \rho u}{\partial x} + \frac{\partial \rho v}{\partial y} = 0 \quad (4)$$

Momentum conservation in the x-direction

$$\left(\frac{\partial(\rho u u)}{\partial x} + \frac{\partial(\rho u v)}{\partial y} \right) = -\frac{\partial p}{\partial x} + 2\frac{\partial}{\partial x} \left(\mu \frac{\partial u}{\partial x} \right) + \frac{\partial}{\partial y} \left[\mu \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] - \frac{2}{3} \frac{\partial}{\partial x} \left[\mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right] \quad (5)$$

Momentum conservation in the y-direction

$$\left(\frac{\partial(\rho u v)}{\partial x} + \frac{\partial(\rho v v)}{\partial y} \right) = -\frac{\partial p}{\partial y} + 2\frac{\partial}{\partial y} \left(\mu \frac{\partial v}{\partial y} \right) + \frac{\partial}{\partial x} \left[\mu \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] - \frac{2}{3} \frac{\partial}{\partial y} \left[\mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right] \quad (6)$$

Mass conservation of solute

$$\frac{\partial(\rho u m)}{\partial x} + \frac{\partial(\rho v m)}{\partial y} = \frac{\partial}{\partial x} \left[\rho D \left(\frac{\partial m}{\partial x} \right) \right] + \frac{\partial}{\partial y} \left[\rho D \left(\frac{\partial m}{\partial y} \right) \right] \quad (7)$$

The solution properties (density (ρ), viscosity (μ) and diffusivity (D)) are considered to be a function of the solute mass fraction while gravity effects are not considered.

This system of differential equations is solved for a rectangular channel (Figure 3) using the following set of boundary conditions.

Figure 3. Scheme of the simulated RO feed channel.

At the inlet ($x=0$), a fully developed velocity profile and solute mass fraction (m) are specified, giving

$$\begin{aligned} u &= u_o \left[1 - \left(\frac{y}{h} \right)^2 \right] \\ x = 0; \quad \forall y; \quad v &= 0 \\ m &= m_o \end{aligned} \quad (8)$$

At the impermeable non-membrane wall, tangential and normal velocities are set to 0, as well as the no solute flux condition is applied

$$\begin{aligned} y = h; \quad \forall x \quad u &= 0 \\ v &= 0 \\ \frac{\partial m}{\partial y} &= 0 \end{aligned} \quad (9)$$

At the membrane interface the no-slip condition ($u=0$) was also fixed, the v -velocity, v_m , is equal to the permeate volumetric flux, J , and for the solute concentration, the permeate flux must balance the convective and diffusive fluxes.

$$\begin{aligned} y = -h; \quad \forall x \quad u &= 0 \\ v &= v_m \\ \rho D \left(\frac{\partial m}{\partial y} \right) + \rho v_m m_m &= \rho v_m m_p \end{aligned} \quad (10)$$

The concentration on the two sides of the membrane are related via a rejection coefficient (R)

$$R = 1 - \frac{m_p}{m_m} \quad (11)$$

Hence, assuming that the density of the solution remains constant at both sides of the membrane, the condition for the solute mass fraction at the membrane boundary is

$$\rho D \left(\frac{\partial m}{\partial y} \right) + \rho v_m m_m R = 0 \quad (12)$$

This equation is equivalent to equation 1, where C is replaced by $\rho \cdot m$ and the permeate volumetric flux, J , by v_m . Permeate flux is calculated using the osmotic pressure law (eq. (2)).

3.2. Numerical simulations and model validation

The governing equations (eq. (4)-(7)) subject to the specified boundary conditions (eq. (8)-(12)) were solved numerically using commercial finite element software (Comsol Multiphysics®, version 3.4).

This software, which has been previously used by other authors [5,30,31] for filtration modelling, uses the Finite Element Method (FEM) as a numerical simulation technique. The FEM employs a

subdivision of the problem domain into a finite number of small regions called finite elements and can provide near-optimal approximate solutions to initial-boundary value engineering problems [15].

The system of Partial Differential Equations (PDEs) was introduced in the weak formulation. To formulate the weak form, each equation of the PDE system is multiplied by an arbitrary function, called test function. Then the PDEs are integrated by parts using the Green's formula, thus reducing the derivative order of the variables [32,32]. A stationary nonlinear solver was used to solve the equations using a maximum number of 25 iterations along with a relative tolerance of 10^{-6} . To have an accurate solution of the transport equations, a denser grid near the membrane was used since concentration polarization occurs only in the vicinity of the membrane and finite element simulations are limited by mesh accuracy [15].

The first set of calculations aimed to validate the model. To test the validity of the model presented above, simulations were carried out for the same rectangular channel and model parameters used by Fletcher and Wiley (2004) [20]. These results were also used by other authors to validate their model [33]. The results obtained in the tests simulations revealed an excellent agreement with those presented in [20].

Once the model was validated, it was used to simulate the experiments carried out. For this simulation only the feed channel of the RO cell was considered; the dimensions of the simulated domain are shown in Figure 4.

Figure 4. Scheme of the geometry considered in the simulations

The parameters of the model related to the membrane performance (K_m and R) were obtained from the experimental results. A mean value of $1.29 \times 10^{-11} \text{ m Pa}^{-1} \text{ s}^{-1}$ for K_m was used for all the simulation runs since the change in the permeation flux of pure water in all the experiments was very slight, while R was found to be dependent on the concentration of the bulk solution. The solution properties used are those defined in section 2.2.

4. Results and Discussion

The combination of three different initial concentrations and four windows gives a total of 12 experiments that were repeated three times to check reproducibility. In each experiment, the crossflow velocity was changed using three different values in order to test also the effect of this variable on the CP layer.

In all the experiments, measurement of the permeate flux and concentration profiles showed that the steady state was reached after a few minutes. In fact, the permeate flux reached an almost constant value after approximately 5 minutes, depending on the feed concentration and crossflow velocity used. In a similar way, the interferograms recorded with DHI showed a stable appearance, with a number of fringes almost constant after this time lapse, thus indicating the stabilization of the concentration profile.

Besides the permeate flux and the polarization layer, the permeate concentration was measured. The value of this concentration was always very low, thus resulting in rejections greater than 92% with slight differences among experiments, depending on C_0 . In particular, the mean values of the rejection coefficients (R) were 0.97, 0.96 and 0.93, for feed concentration of 3, 6 and 9 kg/m³, respectively.

4.1. Concentration polarization layer

The polarization layer can be visualized as an interference fringe pattern thanks to Digital Holographic Interferometry. When the pressure was applied and the experiment started, interference fringes began to appear in the membrane surface, which moved away from the membrane while new fringes continued appearing. After a few minutes, the interference fringe pattern remained almost stable and no new fringes went out, showing that the RO process had reached the steady state. Since each fringe represents a progressive growth in solution concentration, an increase in the number of fringes points out an increase of the polarization layer and, likewise, a stable interference fringe pattern shows a polarization layer in steady state.

When the crossflow velocity was changed, a similar performance was observed: the number of fringes was increasing during several minutes until it reached a constant value. Since the change of CF velocity was always to decrease, the shear effect of the flowing solution was always lesser and the number of fringes increased after a change in the CF velocity; i.e, the polarization layer always grew when the CF velocity decreased. This behaviour was observed in every experiment, regardless of the window of the module studied, i.e., regardless of the position along the channel.

Figure 5 shows the interferograms and the corresponding concentration profiles for all the experiments with $C_0 = 6$ kg/m³. At the top of the figure, it can be observed, for each window, the interferograms once the steady state is reached. They have been taken 15 minutes after establishing each CF velocity in every experiment, when the process had clearly reached the steady state. Each of the three

rows of interferograms corresponds to a different Re number, from 13 at the first row to Re 111 at the third one. Below the interferograms, the corresponding concentration profiles are shown. They are shown reversed from Figure 2 in order to visualize concentration as a function of the distance, i.e., a concentration profile.

Light deflection correction proposed by Rodrigues et al. [27], that can change the position of the fringes when drawing the respective concentration profiles, has been taken into account to draw the concentration profiles. However, since the ratio between the overall refractive index change in the polarization layer (Δn) and its thickness (δ) has always been lesser than 1 m^{-1} , no correction in the position of fringes from interferograms was needed.

Figure 5. Interferograms and concentration profiles for experiments with $C_0 = 6 \text{ kg/m}^3$ ($\blacklozenge$ - Re 13; $\blacktriangle$ - Re 38; $\bullet$ - Re 111)

The interferograms and concentration profiles in Figure 5 clearly show the influence of the crossflow velocity on the CP layer. At higher velocities, the CP layer is reduced due to the greater shear effect that diminishes the polarization layer along the whole channel. It is visibly clear from the concentration profiles that both concentration at the membrane surface (C_m) and polarization layer thickness (δ) are smaller as Re increases, for every distance from the inlet.

Besides the effect of the CF velocity, the evolution of the polarization layer along the membrane channel is also obvious. In all the experiments, both the number of interference fringes appeared and the distance from the furthest fringe to the membrane surface increased along the channel. In other words, the continuous entrance of solute and its accumulation make the CP layer grow continuously and both C_m and δ increase along the channel. However, in the last window ($x = 190$ mm), instead of this expected evolution, a decrease in both C_m and δ is observed. At this location, the proximity of the outlet of the cell (edge effect) could cause flow turbulence and, consequently, a raise of the shear flow thus reducing the polarization layer. Results from simulation (as explained below) are in accordance with this hypothesis.

For example, looking at the concentration profiles in Figure 5, the experimental values of the concentration at the membrane, C_m , with the lowest Re , are 7.8, 9.3, 9.9 and 8.7 kg/m³ at each distance from the inlet. Experimental values of δ evolved similarly and, for the same lowest Re , are 0.35, 0.60, 0.85 and 0.55 mm. The rest of the experiments shown in Figure 5 presented the same behaviour regardless of the CF velocity: the growth of the CP layer along the channel as a consequence of solute rejection and the continuous entrance of solute to the cell, as well as the decrease of the CP layer near the end of the channel due to the outlet effect.

The evolution of the CP layer along the membrane channel can be visualized more easily in Figure 6. It shows the evolution of C_m along the whole channel for $C_0 = 6$ kg/m³ for all the Re used. In order to a better visualization of the evolution of C_m , a point corresponding to the entrance to the cell, where C_m is still equal to C_0 , has been added. Values of the concentration at the membrane have been extrapolated from concentration profiles.

It can be observed (Figure 6) the development of the polarization layer along the channel and the important effect of turbulences at the outlet of the cell, which cause the fall in C_m . Moreover, there is a difference in the behaviour of C_m for each crossflow velocity. At the higher Re , the concentration at the membrane surface tends to stabilize closer to the inlet end; thus, C_m values at the second and third window are very similar. However, when Re is 13, there is still an increase of C_m from $x = 80$ mm (second window) to $x = 140$ mm (third window). The reason is that a higher Re implies a faster movement of the solute in the axial direction, which prevents a continued accumulation of solute along the boundary layer.

Figure 6. Evolution of C_m along the membrane channel

The results for the rest of experiments at different C_0 (3 and 9 kg/m³), follow the same trends just explained. Figure 7 shows a summary of the concentration profiles measured in all the experiments, including those for $C_0 = 6$ kg/m³ already shown. The concentration profiles corresponding to the same C_0 and Re are depicted together, so the evolution of the concentration profile along the channel can be observed in the same graphic. For all the experimental conditions, the concentration profile grows along the membrane length, with the exception aforementioned ($x = 190$ mm). The concentration profile corresponding to that distance is reduced down below the profile corresponding to $x = 80$ mm, thus indicating that the CP layer at the channel outlet tends to approximate the magnitude of the CP layer near the channel inlet. This behaviour experimentally observed points out the important influence of the development of the flow axial to the membrane surface on the growth of the concentration polarization layer in RO processes.

Figure 7. Evolution of the concentration profiles along the membrane for all the experimental conditions. ($\square$) 30 mm; ($\times$) 80 mm; (Δ) 140 mm; ($\circ$) 190 mm

The evident influence of the crossflow velocity on the concentration profiles is also observed in Figure 7, since for all C_0 , both C_m and polarization layer thickness decrease with the increasing Re. It can be observed that, for a certain Re, δ grows with the feed concentration. When Re is 13, the layer thickness is about 0.6, 0.8 and 1.0 mm for each C_0 (3, 6 and 9 kg/m³, respectively) considering the polarization layer fully developed; i.e., at $x = 140$ mm. However, as Re increases, those differences in δ become lesser and, at the highest Re, there are hardly differences in the layer thickness; for Re = 111, δ is about 0.2 mm for every initial concentration. This behaviour takes place because a slow flow of solute moving tangentially to the membrane surface let a higher accumulation of solute, which is higher at higher concentrations.

It is generally assumed that the mass transfer coefficient is related with Re through a correlation of the type $Sh=f(Re, Sc)$. Therefore, when Re increases, the mass transfer coefficient ($k=D/\delta$), included in the Sherwood number, also increases; i.e. there is a decrease in δ .

The numerical simulation of the process, using the CFD model described in section 3, provides a reasonable agreement with the experimental results. Figure 8 shows the experimental and simulated concentration profiles for the experiments with $C_0 = 6 \text{ kg/m}^3$. They are presented independently, like the interferograms of Figure 5, for the sake of clarity. It can be observed that the computed concentration profiles agree quite well at $x = 80$ and 140 mm , where the polarization layer is almost fully developed. However, simulated concentration profiles in the windows nearer to the inlet and outlet of the cell show a worst agreement with the experimental ones. In all cases, the experimental profiles are lower than the simulated ones. As explained before, at these positions there could be some turbulence caused by the change in the hydrodynamic conditions; i.e., a change in the flow section between the membrane channel and the pipes connecting the cell with the rest of the RO system. Therefore, the flow could become unstable and break the velocity profile, a fact that is not assumed in the simulations, where a stable profile is considered.

Furthermore, turbulence of the entrance of the channel could also contribute to the lower experimental concentration profiles along the whole channel. In fact, the differences between experimental and simulated results are higher for the highest cross flow velocity, where there will be more edge effect and more turbulence.

Figure 8. Experimental and simulated concentration profiles for the experiments with $C_0 = 6 \text{ kg/m}^3$. (◇) experimental; (—) calculated.

4.2. Permeate flux

The permeate flux data contribute with more information about the variables involved in the development of the polarization layer. Figure 9 shows the experimental values of the permeate flux and those predicted by the simulations, for every C_0 and Re used. The values presented in Figure 9, are the average values along the membrane channel, and it can be observed the influence of the variables under study (C_0 and Re) on J .

On the one hand, a higher Re number leads to a higher permeate flux. As it has been previously explained, the crossflow velocity has a notable influence on the concentration polarization, particularly reducing the thickness of the polarization layer. Thus, C_m was also reduced and consequently the resistance to J (eq. (2)) decreased. However, the influence of the Re number on permeate flux was not as important as it would be expected and only slight differences in J were observed.

Figure 9. Permeate flux, experimental and simulated, for all the experiments

On the other hand, there was a remarkable influence of the feed concentration on J . An increase in twice or thrice the C_0 , led to a decrease of the same order in J , for every C_0 . Initial concentration (and so C_m) greatly affected permeate flux due to the rise of the osmotic pressure resistance.

Finally, it was observed that the simulated permeate fluxes were very similar to experimental values, with a mean error of 4 %. These results confirm that the employed CFD model properly predicts both permeate flux and the evolution of the polarization layer provided that the hydrodynamics of the system was stable.

5. Conclusions

Digital Holographic Interferometry has been proved to be a suitable technique to study the polarization layer in a reverse osmosis process. Concentration profiles have been measured directly at different positions of a new module large enough to allow the monitoring of the evolution of the polarization layer along the membrane channel. The RO process has been simulated with a model that couples the solute mass transfer with the momentum transport equations. The set of equations has been numerically solved using commercial CFD software. The comparison between the experimental results and those predicted by the simulation shows a reasonable good agreement between them.

The interferograms captured during the process and their corresponding concentration profiles confirm that the CP layer grows along the feed channel due to the combination of the continuous entrance of solute to the cell and the accumulation of solute in the CP layer. This phenomenon obviously occurs regardless of the feed concentration or the crossflow velocity. However, higher velocities lead to an earlier stabilization of the polarization layer due to a faster flow of solute moving tangentially to the membrane surface, which prevents the growing solute accumulation along the whole channel. Moreover, high CF velocity (or turbulence due to an edge effect) causes low concentration polarization reducing the boundary layer thickness, δ , and the concentration of the solution in contact with the membrane surface, C_m . This same behaviour was also observed in the results computed with the model.

The reduction of the polarization layer due to a high CF velocity causes also an increase in the permeate flux, although this effect is not so significant. Indeed, permeate flux data obtained from the experiments and the simulation show that it is mostly affected by the feed concentration. The osmotic pressure of the solution causes a notable increase in the resistance to the flux, becoming the main factor influencing the performance of the RO process.

In summary, the development of the polarization layer is a complicated phenomenon that is influenced by all the main variables of the process: driving force (difference of pressure between the feed and permeate side of the membrane), resistance to the flux (membrane characteristics, osmotic pressure) and hydrodynamics (CF velocity, channel dimensions). All of them are influenced by each other and there exists a sort of balance. Hydrodynamics (cross flow velocity or turbulence) basically affects the mass transfer (k) by reducing the polarization layer thickness (δ). However, this reduction in δ is not enough to notably improve the performance of the process and to increase permeate flux. The osmotic pressure, dependent of C_m (or C_0), is the variable that has a stronger influence on it.

Therefore, the attempts to reduce polarization layer or increase mass transfer (use of spacers to increase turbulence, a higher pressure or CF velocity, buoyancy...) seem to have a limit in the improvement of the process, due to the osmotic pressure of the solution. However, a detailed study of

them could contribute to the understanding of the phenomena of the concentration polarization. In certain processes, Digital Holographic Interferometry can be a suitable technique to that purpose.

Acknowledgements

This research has been sponsored by the Plan Nacional de I+D+i CTQ2009-14109 (Ministerio de Ciencia e Innovación).

Nomenclature

C	solute concentration (kg m^{-3})
D	diffusion coefficient of solute in water (m^2s^{-1})
J	permeate flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)
J_w	pure water permeate flux ($\text{m}^3 \text{m}^{-2} \text{s}^{-1}$)
k	mass transfer coefficient (ms^{-1})
K_m	water permeability of the membrane ($\text{m Pa}^{-1} \text{s}^{-1}$)
m	solute mass fraction
ΔP	imposed pressure difference across the membrane (Pa)
P	pressure
R	rejection coefficient
Re	Reynolds number (dimensionless)
Sc	Schmidt number (dimensionless)
Sh	Sherwood number (dimensionless)
u	velocity in the x direction (axial to the membrane surface) (m s^{-1})
u_o	maximum value of the velocity profile at the inlet (m s^{-1})
v	velocity in the y direction (normal to the membrane surface) (m s^{-1})

Greek letters

$\Delta\pi$	osmotic pressure difference across the membrane (Pa)
δ	polarization layer thickness (m)
ρ	fluid density (kg m^{-3})
μ	fluid viscosity (Pa s)

Subscripts

p	value at the permeate side of the membrane
m	value at the wall on the feed channel side of the membrane
0	value at the entrance of the channel

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Figure Captions

Figure 1. RO module

Figure 2. Concentration profile obtained from an interferogram

Figure 3. Scheme of the simulated RO feed channel.

Figure 4. Scheme of the geometry considered in the simulations

Figure 5. Interferograms and concentration profiles for experiments with $C_0 = 6$ kg/m³ (◆- Re 13; ▲- Re 38; ● - Re 111)

Figure 6. Evolution of C_m along the membrane channel

Figure 7. Evolution of the concentration profiles along the membrane for all the experimental conditions. (□) 30 mm; (x) 80 mm; (▲) 140 mm; (o) 190 mm

Figure 8. Experimental and simulated concentration profiles for the experiments with $C_0 = 6$ kg/m³. (◇) experimental; (—) calculated.

Figure 9. Permeate flux, experimental and simulated, for all the experiments

$$\text{Ec. 1.} \quad J C_m + D \left(\frac{dC}{dy} \right)_{y=0} = J C_p$$

$$\text{Ec. 2.} \quad J = k_m (\Delta P - \Delta \pi)$$

$$\text{Ec. 3.} \quad \frac{C_m - C_p}{C_o - C_p} = \exp\left(\frac{J \delta}{D}\right) = \exp\left(\frac{J}{k}\right)$$

$$\text{Ec. 4.} \quad \frac{\partial \rho u}{\partial x} + \frac{\partial \rho v}{\partial y} = 0$$

$$\text{Ec. 5.} \quad \left(\frac{\partial(\rho u u)}{\partial x} + \frac{\partial(\rho u v)}{\partial y} \right) = -\frac{\partial p}{\partial x} + 2 \frac{\partial}{\partial x} \left(\mu \frac{\partial u}{\partial x} \right) + \frac{\partial}{\partial y} \left[\mu \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] - \frac{2}{3} \frac{\partial}{\partial x} \left[\mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right]$$

$$\text{Ec. 6.} \quad \left(\frac{\partial(\rho u v)}{\partial x} + \frac{\partial(\rho v v)}{\partial y} \right) = -\frac{\partial p}{\partial y} + 2 \frac{\partial}{\partial y} \left(\mu \frac{\partial v}{\partial y} \right) + \frac{\partial}{\partial x} \left[\mu \left(\frac{\partial u}{\partial y} + \frac{\partial v}{\partial x} \right) \right] - \frac{2}{3} \frac{\partial}{\partial y} \left[\mu \left(\frac{\partial u}{\partial x} + \frac{\partial v}{\partial y} \right) \right]$$

$$\text{Ec. 7.} \quad \frac{\partial(\rho u m)}{\partial x} + \frac{\partial(\rho v m)}{\partial y} = \frac{\partial}{\partial x} \left[\rho D \left(\frac{\partial m}{\partial x} \right) \right] + \frac{\partial}{\partial y} \left[\rho D \left(\frac{\partial m}{\partial y} \right) \right]$$

$$\text{Ec. 8.}$$

$$u = u_o \left[1 - \left(\frac{y}{h} \right)^2 \right]$$

$$x = 0; \quad \forall y$$

$$v = 0$$

$$m = m_o$$

$$\text{Ec. 9.}$$

$$u = 0$$

$$y = -h; \quad \forall x$$

$$v = 0$$

$$\frac{\partial m}{\partial y} = 0$$

$$\text{Ec. 10}$$

$$u = 0$$

$$y = -h; \quad \forall x$$

$$v = v_m$$

$$\rho D \left(\frac{\partial m}{\partial y} \right) + \rho v_m m_m = \rho v_m m_p$$

$$\text{Ec. 11.} \quad R = 1 - \frac{m_p}{m_m}$$

$$\text{Ec. 12} \quad \rho D \left(\frac{\partial m}{\partial y} \right) + \rho v_m m_m R = 0$$

Highlights

- Polarization layer in crossflow RO has been visualized by Holographic Interferometry
- Evolution of the polarization layer (PL) along the channel has been directly observed
- PL is mostly affected by the CF velocity, but it hardly influences permeate flux
- Feed concentration reduces considerably the permeate flux due to the osmotic pressure
- Experimental results have been compared with calculated data from CFD