

Filtration method characterizing the reversibility of colloidal fouling layers at a membrane surface: Analysis through critical flux and osmotic pressure

Benjamin Espinasse, Patrice Bacchin^{*}, Pierre Aimar

Laboratoire de Génie Chimique, Université Paul Sabatier, 31062 Toulouse cedex 9, France

Received 4 September 2007; accepted 16 January 2008

Available online 19 January 2008

Abstract

A filtration procedure was developed to measure the reversibility of fouling during cross-flow filtration based on the square wave of applied pressure. The principle of this method, the apparatus required, and the associated mathematical relationships are detailed. This method allows for differentiating the reversible accumulation of matter on, and the irreversible fouling of, a membrane surface. Distinguishing these two forms of attachment to a membrane surface provides a means by which the critical flux may be determined. To validate this method, experiments were performed with a latex suspension at different degrees of destabilization (obtained by the addition of salt to the suspension) and at different cross-flow velocities. The dependence of the critical flux on these conditions is discussed and analysed through the osmotic pressure of the colloidal dispersion.

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Keywords: Critical flux; Ultrafiltration; Colloids; Membrane; Irreversibility; Fouling; Osmotic pressure

1. Introduction

As the price of membranes continues to decrease, operating costs are representing a larger share of the total cost of membrane processes and are limiting the development of membrane technologies. In membrane filtration, these costs are primarily linked to fouling in two points: one is relative to the direct reduction of productivity and the other is linked to the selectivity of the operation. The formation of a deposit on the membrane surface generally changes its properties. This is an important problem for applications that are very sensitive to the surface properties, as in food and pharmaceutical industries [1]. When a natural dispersion is ultra-filtered, fouling is often the consequence of the concentration of colloids [2] (macromolecules or sub-micronic particle). Now, colloid dispersions exhibit a specific behaviour because of surface interactions. These surface interactions are multiple in nature (electrostatic repulsion, van der Waals attraction or hydrophobic–philic interaction) and have different interaction lengths; thus leading to a complex

system when colloidal dispersions are concentrated. It has been shown [3] that the accumulation of matter on the membrane surface can be characterised with regard to its reversibility. A reversible accumulation will “disappear” when the trans-membrane pressure (TMP) is decreased, while an irreversible one (with the form of a deposit or a gel) will remain on the membrane surface when the pressure is released. In the case of irreversible fouling, the method for removing the deposit layer is mechanical or chemical cleaning. The limit between reversible and irreversible fouling often appears to operators when they increase the permeation flux; the threshold value for which the reversible accumulation of matter turns into an irreversible cake can be called a critical flux. It was defined in the literature in 1995 [4,5] as the flux above which an irreversible deposit appears at the membrane surface. Its experimental determination is then of practical importance to optimize the operating conditions of a membrane process [3].

Mass accumulation on a membrane surface can be reversible if the matter is accumulated but stays dispersed, or does not stick to the surface (a reduction in the applied pressure reduces the concentration polarisation by diffusion). The osmotic model is well accepted to establish the J versus TMP relationship. However, the concept of osmotic pressure is well established for

^{*} Corresponding author. Fax: +33 5 61 55 61 39.

E-mail address: bacchin@chimie.ups-tlse.fr (P. Bacchin).

molecules and salts (and their limiting effects on nanofiltration and reverse osmosis) but not as much for colloidal dispersions in processes such as ultrafiltration. A number of studies have provided experimental evidence of the osmotic pressure of colloidal dispersions [6] with various measurement methods. Different authors have linked the osmotic pressure to the properties of the colloids [7–9]. The main difference between molecular and particulate osmotic pressure lies in the range of pressure (much lower for particles). Colloidal systems such as dispersions of latex particles can “resist” compression and “create” an osmotic pressure from around 0.1 bar to a few bars [10] according to the particle size and their inter-particle interactions. For some critical value of the osmotic pressure (or at the associated critical volume fraction, generally around 0.4–0.6 bar), the compressed dispersion is no longer stable and particles make an irreversible state transition from a dispersed to a condensed phase [11] where particles are all in contact with each other. This irreversible transition from a dispersed to a condensed phase can occur at the membrane surface when the permeate flux is high enough, resulting in a deposit or gel layer formation at the membrane surface. Consequently, deposit layer formation is intrinsically linked to the osmotic pressure and to the permeate flux that leads to aggregation of the particles on the membrane surface (i.e., the critical flux) [5].

The critical flux was defined in 1995 by two different teams both theoretically and experimentally [4,5,12]. Theoretically the critical flux was defined as the *flux above which an irreversible deposit appears at the membrane surface*. For colloidal suspensions this phenomenon is generally a balance of particle–particle or particle–membrane repulsive forces and permeate drag forces. Above a given value of flux, when the repulsive forces are overcome by the permeate drag forces, a deposit forms on the membrane surface and creates an additional resistance to the permeate flow through the membrane.

Based on a theoretical force balance approach, when the critical flux is overcome a decrease in pressure will not lead to a spontaneous decrease in resistance. Howell et al. [13] confirmed the existence of a critical and sub-critical flux where, at a constant flux, the pressure remains constant with time. No deposit appears for these operating conditions on the membrane surface, regardless of the duration of filtration. On the other hand, setting a flux above the critical flux will lead to an increase in pressure with time until a steady state condition has been reached. The concept of critical flux is also used in membrane bioreactor studies to determine the filtration regime by analyzing the variations in pressure for constant flux experiments [14]. Researchers often consider the critical flux to have been reached when the pressure cannot attain steady state with time for that given flux [15]. Other investigators [16], in contrast, measured the transmembrane pressure necessary to maintain a given permeation flux across a microfiltration membrane for silica suspensions by using a flux stepping method (Fig. 1).

These experiments determined two kinds of critical flux: the weak form (to separate non linear pressure–flux variations to linear variations with a slope being inferior to the one of the water flux) and the strong form (to separate non linear pressure–

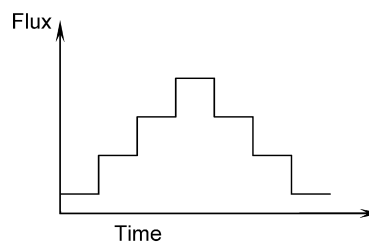


Fig. 1. Filtration procedure with flux stepping used in literature [4] to find the critical flux.

flux variations to linear variations similar that the one reached for water flux).

Other techniques are also used to detect deposit formation: evolution of concentration in the suspension [17] in a batch operation can allow estimation of the accumulated material via mass balance. Direct microscope observation [18] has also been used to study the evolution of a deposit on a membrane surface for particles having a size larger than a few microns, but as an indirect method, the accuracy and relevance are unsatisfactory.

The aim of this work is to present a square wave filtration method to determine the extent of reversibility of fouling during membrane filtration. The general principle of this method has been briefly presented in earlier studies [19]. This method gives a more accurate value of the critical flux than other techniques. The improvement is based on the use of a square wave filtration method, which makes it possible to determine the irreversibility along a range of permeate fluxes. This method allows the detection of and distinction between the reversible and irreversible portions of fouling when a flux reduction is observed. The reversible part is described by an osmotic pressure contribution, whereas the irreversible part is analyzed as a hydraulic resistance (deposit or gel-like layer). This method allows a pragmatic definition of the critical flux, and is validated here for different conditions of filtration and colloidal stability.

2. Materials and methods

2.1. Filtration rig

The ultrafiltration set-up used in this study is shown in Fig. 2. The pressure is controlled by a current-to-pressure transducer (CPT, Rosemount, Baar, Switzerland) with an accuracy of 0.02 bar and regulated with a PID (proportional-integral-derivative) regulator. The filtration rig is temperature controlled at 25 °C and the permeation flux is measured with an electronic balance (Adventurer, Ohaus, Nanikon, Switzerland) linked to a computer. Crossflow is measured with a flow-meter (Promag A, Endress-Hausser, Reinach, Switzerland) with a precision of 3%.

The ultrafiltration module contains one inner skinned Carbosep tubular membrane (Orelis, Miribel, France). Its molecular weight cut off was 15 kDa.

The total active membrane surface was 0.0226 m², with a length of 1 m and a hydraulic diameter of 6 mm. In this rig, the flow velocity can be varied from 0.30 to 1.27 m s⁻¹. The Reynolds numbers associated with these flow rates are 1952 and 8458, respectively.

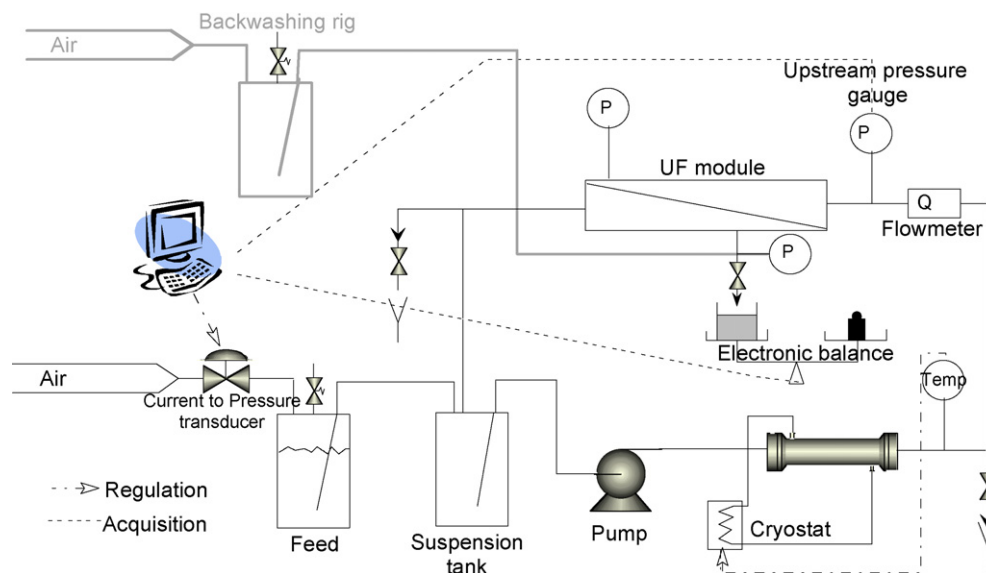


Fig. 2. Flow sheet of the filtration rig used for the critical flux determination.

Table 1
Evolution of zeta potential with the ionic strength

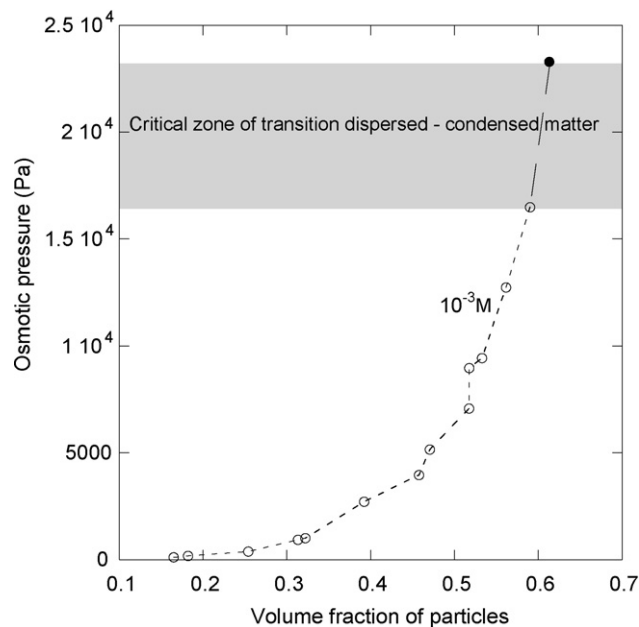
Ionic strength (M)	Zeta potential (mV)	
0	-49 ± 1	-71 ± 2
10^{-4}	-57 ± 1	-76 ± 2
10^{-3}	-61 ± 1	-71 ± 2
10^{-2}	-76 ± 1	-80 ± 2

The concentration of the suspension in the filtration rig is: 0.7 g L^{-1} . The retentate of the suspension is recycled and the permeate is automatically refilled by a solution of same ionic strength. The length of every pressure step is variable. The length of each step is determined by the time necessary to reach the equilibrium of flux. We consider equilibrium to have been reached when the variation of flux is less than 2% per hour. In this work, the time necessary to reach the equilibrium ranges from 30 min to 4 h. The pressure in the system is obtained with compressed air that pressurizes the feed tank. The pressure is accurately regulated in the rig by a 'current to pressure transducer' (CPT) controlled with a computer software interface.

2.2. Latex suspension

Latex particles used in these experiments are hard PVC spheres with a zeta potential ranging from $-60 \pm 2 \text{ mV}$ at pH 3.5 to $-85 \pm 2 \text{ mV}$ at pH 9. In the working conditions (pH 6–7) the average zeta potential is $-71 \pm 2 \text{ mV}$ with a mean particle diameter of 115 nm (Zetasizer 4, Malvern Instruments, Worcestershire, UK) when no salt is added (Table 1).

The particles are 100% rejected by the membrane. The salt concentration, in all experiments was below the critical coagulation concentration (0.1 M in KCl) to avoid particle aggregation in initial dispersion. In this work, salt acts on the stability of particles that decreases when salt concentration increases. To have an experimental evidence of this effect, the osmotic

Fig. 3. Results of measurements of the osmotic pressure vs volume fraction of particles of PVC latex in deionised water and $[\text{KCl}] = 10^{-3} \text{ M}$.

pressure of the latex suspension was measured by a chemical compression method previously described in literature [8]. Details of this method can be found in literature [10,20]. The results for an ionic strength of 10^{-3} M of KCl are presented in Fig. 3 with a classical increase in osmotic pressure with the latex volume fraction. In this work, the increase of the osmotic pressure is related to the increase of the repulsive electrostatic interactions between colloids as particles are brought closer together. It was proven [20] that the osmotic pressure is sensitive to the variation of ionic strength; an increase in ionic strength leads to a decrease of the osmotic pressure (as the repulsive interactions decrease).

At a given volume fraction the particles are no longer stable and they turn from a dispersed to a condensed phase (the

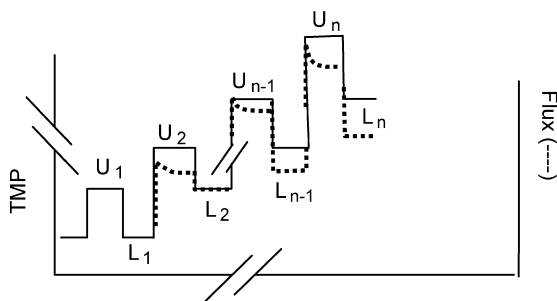


Fig. 4. Principle of the square wave technique; pressure and flux vs time; upper and lower steps.

dispersion appears as a solid). This transition has been verified by re-dispersing 1 g of the compressed suspension into 200 ml of distilled water under agitation for 24 h. Afterwards, the bulk suspension was analysed (by turbidity measurement) to evaluate the proportion of particles that re-dispersed. When no particles were re-suspended (black symbol in Fig. 3), it is concluded that the phase transition had occurred during the compression (the latexes are condensed during the osmotic pressure measurement test). This transition is linked to the concept of critical flux, which corresponds to an irreversible transition between a reversible to an irreversible deposit on the membrane.

3. Results and discussion

3.1. Principles and first results of the Square wave barovelocimetry method (SWB)

The principle of this filtration technique is to alternate step-wise the applied force (the pressure) with positive and negative variations, as presented in Fig. 4, and to continuously measure the permeate velocity. The U steps correspond to the upper steps and the L steps to the lower steps.

At each time step, a steady state permeate flux is reached. The stabilisation of the system (i.e., steady state) takes from a few minutes to several hours depending on system evolution. The method is similar to the electrochemistry method of “square wave voltammetry” and is defined as the square wave baroflumetry.

The interest of this technique is to evaluate the loss in flux between two steps of pressure: the flux is compared between step L_n and U_{n-1} which have the same pressure. The fouling associated to a decrease phenomena that took place at step U_n is considered as totally reversible if the flux is the same at step U_{n-1} and step L_n, and partly irreversible if not.

The classical *J* versus TMP filtration curve corresponds in Fig. 5 to the dotted line (upper pressure steps). The SWB method provides additional information with lower pressure steps that will allow for the deduction of the reversibility of the accumulated layer.

The hydraulic resistance of fouling layers can be classically determined through the integrated form of Darcy law:

$$J = \frac{\Delta P}{\mu(R_m + R_f)} \quad (1)$$

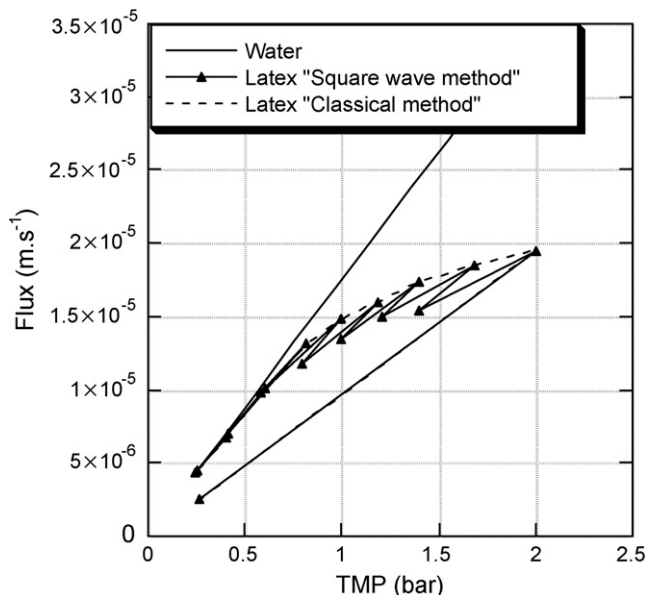


Fig. 5. Flux vs TMP for the “square wave” technique; PVC latex, $Q_c = 0.79 \text{ m s}^{-1}$, $[\text{KCl}] = 10^{-3} \text{ M}$.

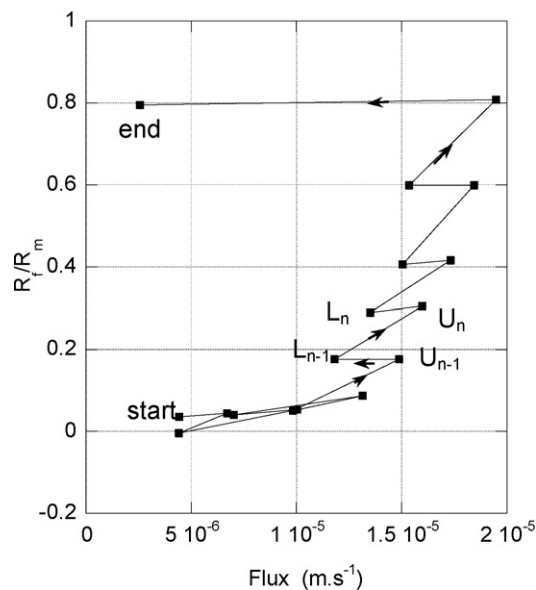


Fig. 6. Evolution of total fouling resistance vs pressure obtained with the “square wave technique.”

The results are presented in terms of fouling resistance, R_f , over membrane resistance, R_m , in Fig. 6 as a function of the permeation flux on the y-axis in Fig. 5.

Fig. 6 is read from the lower left point (start) and follows the arrows. At the beginning of filtration the fouling resistance increases with increasing pressure, but a decrease of pressure allows it to go back to the preceding resistance value; there is no hysteresis as the increase in resistance is reversible. Otherwise, an increase of resistance can be observed between steps $n - 2$ and $n - 1$, such that resistance remains constant when the pressure is decreased at L_{n-1} (the increase of resistance is consequently irreversible). The significance of this pattern is the appearance of an irreversible deposit on the membrane surface.

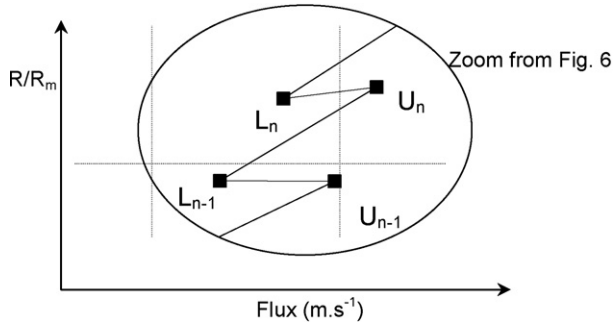


Fig. 7. Detail of Fig. 6 showing a full pressure step in term of R/R_m vs flux.

3.2. Analysis of the filtration in term of irreversible and reversible resistance

3.2.1. Calculation of the irreversible resistance R_{if}

From the representation R_f/R_m , it is possible to determine a grade of reversibility of the matter accumulated at the membrane surface: the term “reversible resistance” being here used to describe the portion of the fouling resistance that is eliminated with a decrease in pressure. The SWB method allows analysing the fouling reversibility by comparing the fouling resistance at a same TMP before and after an upper pressure step.

The irreversible resistance that appears for a upper pressure step U_n can be reached by comparing the fouling resistance at step L_n and U_{n-1} (as defined in Figs. 4, 6 and 7) as follow:

$$\frac{r_{if,n}}{R_m} = \frac{R_f}{R_m} \Big|_{L_n} - \frac{R_f}{R_m} \Big|_{U_{n-1}}, \quad (2)$$

where $r_{if,n}$ is the irreversible fouling resistance relative to the step n . When R_f at step L_n equals R_f at step U_{n-1} the resistance observed at U_n step is totally reversible. To calculate the value of the total R_{if} , at a given pressure step, all step resistances r_i are summed. The fouling resistance at n is the sum of the resistance measured at the previous steps:

$$R_{if} = \sum_n r_{if,n}, \quad (3)$$

where R_{if} is the total irreversible resistance. If the fouling is totally irreversible then. $R_f|_{L_n} = R_f|_{U_{n-1}}$. An analysis of the R_f and R_{if} allows then the differentiation of the reversible and irreversible part of fouling in regard to a decrease of pressure all along the permeation flux range.

3.2.2. Calculation of reversible resistance R_{rf}

From the data previously calculated, in comparison with water flux, it is possible to deduce the contribution of osmotic pressure in the total fouling resistance. At each step, one can find the resistance associated with the reversible resistance, named R_{rf} as follows:

$$\frac{R_{rf}}{R_m} = \frac{R_f}{R_m} - \frac{R_{if}}{R_m}. \quad (4)$$

The reversible and irreversible resistances deduced from the SWB experiment (in Fig. 6) are plotted in Fig. 8.

At low flux, the total resistance is low (less than 10% of the membrane resistance) and totally reversible. For a given flux

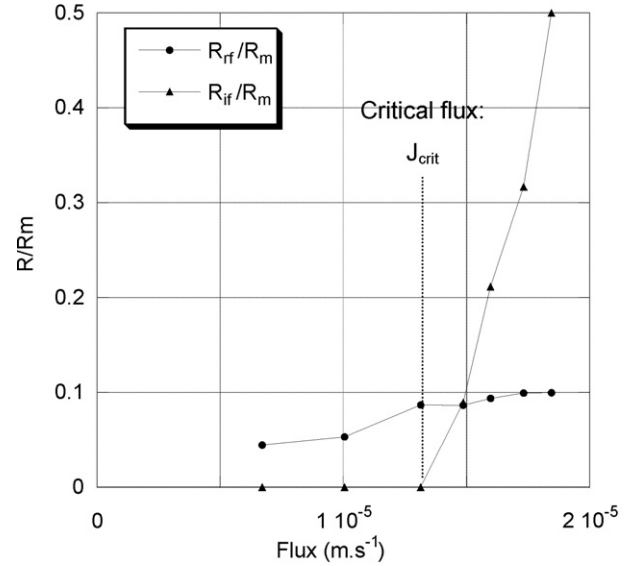


Fig. 8. Evolution of reversible, R_{rf} , and irreversible, R_{if} , fouling resistance vs flux.

(by definition: the critical flux) appears an irreversible resistance which is rapidly growing above this critical value.

3.3. Analysis in terms of irreversible fouling resistance and osmotic pressure

The reversible accumulation of matter, which goes back into suspension after a decrease of pressure, is related to the polarization layer and its induced osmotic pressure, which acts as an opposite force to the applied pressure. In this case, the reversible resistance associated with the polarization concentration layer can be treated like a term of osmotic pressure:

$$J = \frac{\Delta P - \Delta \Pi}{\mu(R_m + R_{if})} = \frac{\Delta P}{\mu(R_m + R_{if} + R_{rf})}. \quad (5)$$

The osmotic pressure at the membrane, Π_m , can be deduced as follows assuming that the membrane is totally retentive:

$$\Delta \Pi = \Pi_m = \Delta P - J\mu R_m \left(1 + \frac{R_{if}}{R_m} \right). \quad (6)$$

Equation (6) allows one to calculate the value of the osmotic pressure along the length of the filtration process with the square wave technique, as shown in Fig. 9. The slight increase in the reversible resistance (as presented in Fig. 8) leads to a more pronounced increase in the osmotic pressure at the membrane with flux (Fig. 9).

The osmotic pressure at the membrane increases due to the evolution of the polarisation layer thickness on the membrane surface (this accumulation is fully reversible if the pressure is decreased). Beyond a given flux ($\sim 1.4 \times 10^{-5} \text{ m s}^{-1}$) is reached, the polarisation layer is thick enough for particles next to the membrane surface aggregate and lead to the creation of an irreversible layer (deposit-like). Beyond this point, the irreversible resistance increase denotes that the deposit layer thickness and/or the surface of the membrane covered by the deposit are increasing. We also observe that the osmotic pressure

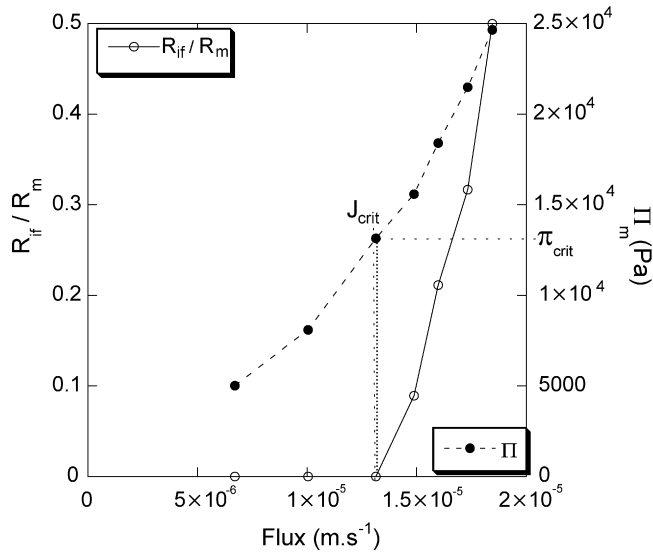


Fig. 9. Evolution of the osmotic pressure at the membrane and the irreversible resistance vs permeation flux.

is increasing above the flux where the first reversible fouling is noticed. That may be due to the growth of the polarization layer in the zone of the membrane where there is no deposit; the deposit layer is probably not completely covering the membrane surface when the critical flux is reached (point discussed in next section). The critical flux, the evolution of the irreversible resistance and the osmotic pressure are function of the particle stability and crossflow velocity as seen in next sections.

3.4. Effect of operating conditions

In this part, the sensitivity of the SWB technique is presented for experiments at different cross-flow velocities. The degree of

particle destabilization is discussed through the evolution of the irreversible resistance, the osmotic pressure and the critical flux.

3.4.1. Effect of crossflow velocity

Results from SWB experiments that are presented in Fig. 10 were performed at crossflow velocities of 0.3 and 0.79 m s⁻¹. These velocities correspond to a laminar flow zone. With these membranes, the transition between laminar and turbulent was measured to be above 0.79 m s⁻¹ [21]. The latex concentration was 0.6 g L⁻¹ and the salt concentration 10⁻³ M KCl.

The first notable point (Fig. 10) is that the irreversible resistance starts to increase at higher permeation flux levels at high crossflow velocities. The second point is that the slope of irreversible resistance R_{if}/R_m decreases with an increase of the crossflow velocity. Those observations are directly related to the evolution of the polarisation layer and have been described in the literature: critical and limiting flux increases with increasing crossflow velocity. Osmotic pressure is almost identical for a small flux but the increases is less for higher crossflow velocities.

When the irreversible resistance appears, the osmotic pressure at the membrane is around 7000–12000 Pa at 0.3 m s⁻¹ and 13000–15000 Pa at 0.79 m s⁻¹. These values are smaller than those determined for the critical transition (16000–23000 Pa) measured with dialysis bags (Fig. 3). This difference may be explained by the fact that the value of the osmotic pressure at the membrane, as determined with the SWB method, corresponds to an averaged value of osmotic pressure along the membrane channel. If locally, at the membrane outlet, the osmotic pressure reaches the value corresponding to the condensation of colloids (critical osmotic pressure), the averaged value of the osmotic pressure along the whole length of the membrane will be less than the critical osmotic pressure. The SWB results show that the averaged osmotic pressure at the membrane increases with

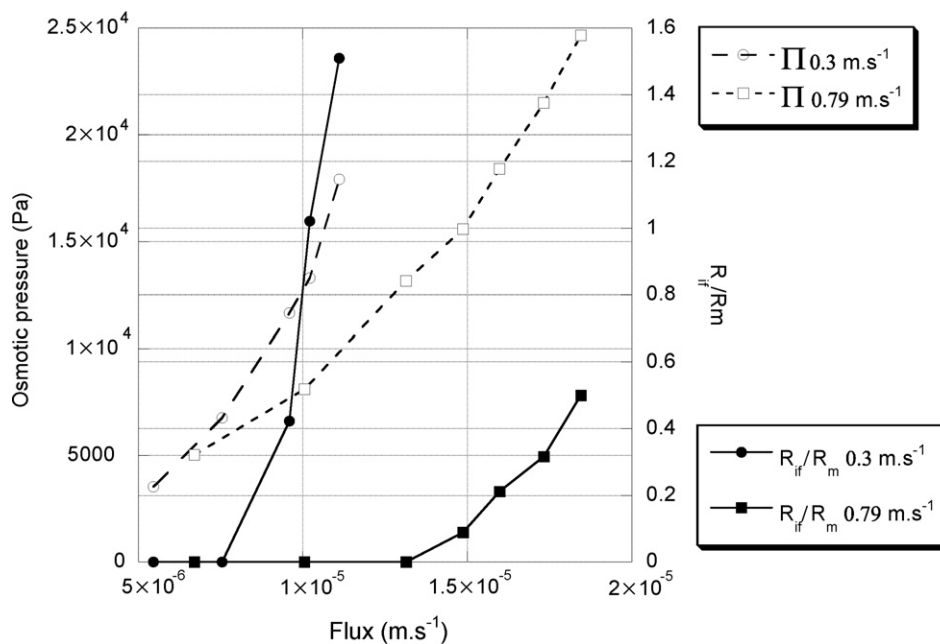


Fig. 10. Evolution of resistances and decomposition in term of osmotic pressure and irreversible resistance for two crossflow velocities; [latex] = 0.6 g L⁻¹, [KCl] = 10⁻³ M.

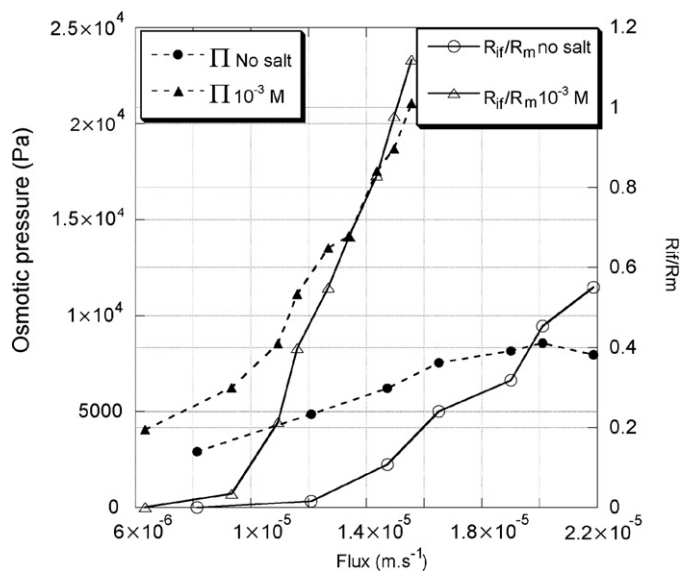


Fig. 11. Evolution of R_f/R_m and R_{if}/R_m and osmotic pressure for two different states of destabilization of particles; different ionic strength, crossflow velocity = 0.6 m s^{-1} , $[\text{latex}] = 0.6 \text{ g L}^{-1}$.

the crossflow velocity. This behaviour can be explained by a different shape of the polarisation layer along the channel for different cross flow velocities. Increasing the cross flow velocity may lead to the formation of a more homogeneous polarisation layer. This could explain that at high crossflow velocities, the average osmotic pressure is close to the critical osmotic pressure measured with dialysis bags. The interpretation of the interdependence between the osmotic pressure and the deposit will require further future investigation with different systems (hollow fibres, flat membrane with different length, etc.) to be able to draw more generic conclusions regarding the development of the polarisation layers and the osmotic pressure.

3.4.2. Effect of the stability of latex dispersion

The evolution of the irreversible resistances and the osmotic pressure as a function of the permeation flux is presented in Fig. 11 at a crossflow velocity of 0.6 m s^{-1} and a latex concentration of 0.6 g L^{-1} . The latex dispersion is more or less destabilized by salt addition: the addition of salt reduces particles interactions and consequently leads to the apparition of a critical flux at lower flux when particles are less stable (i.e. at 10^{-3} M in KCl). The increase in critical flux is accompanied by a decrease in the fouling rate (slopes of R_{if}/R_m vs flux are less important when salt concentration is reduced). These results confirm the importance of the inter-particles interactions in the fouling mechanisms.

The osmotic pressure at the membrane is less important when filtering more stable particles (no salt added). The hypothesis that can be formulated from this is that the volume fraction of particles at the membrane surface is less important with stable particles because they have a better resistance to compression by a more important collective diffusivity. These experimental results confirm theoretical calculations of cross flow filtration that have been discussed in previous article [22].

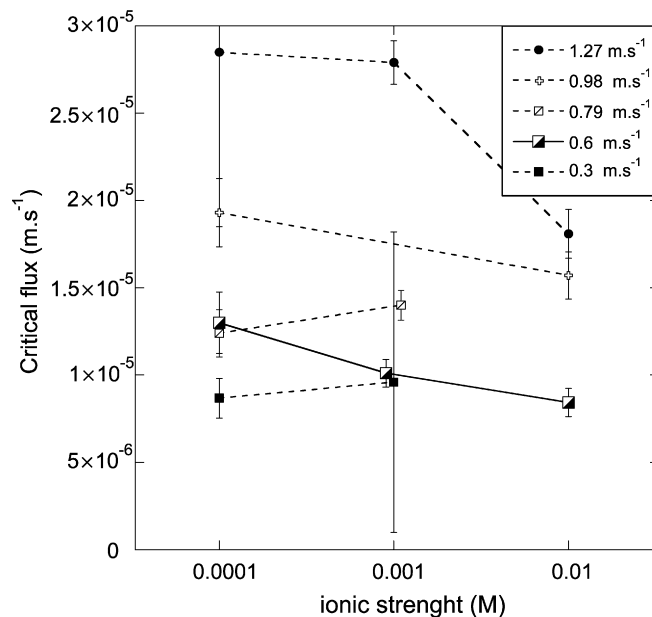


Fig. 12. Evolution of critical flux vs the ionic strength for two crossflow velocities.

The variations of critical flux values with cross flow velocity and salt concentration are summarized in Fig. 12.

As expected, the values of critical flux decrease with the ionic strength, and increases with the increase of the crossflow velocities. The variation of the critical flux at two different crossflow velocities 0.3 and 1.27 m s^{-1} is really important in regard to the observed values in industrial ultrafiltration processes.

4. Conclusions

Using the square wave baroflometry (SWB) technique to analyze membrane fouling data provides a quantitative assessment of the evolution of reversible resistance (osmotic pressure at the membrane) and irreversible resistance with the permeation flux (and the trans-membrane pressure). It allows an accurate determination of the critical flux with regard to the irreversibility of fouling layers. Furthermore, the analysis of fouling in relation to operating conditions allows quantification of the effects of crossflow velocity and particle stability on the growth of the polarized layer and the deposit formation. The measured variation with the operating conditions of the critical flux and the osmotic pressure confirm the importance of the colloidal interactions (or more precisely colloidal stability) on membrane fouling and its reversibility.

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