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# Influence of Water Compositions and Conditioning on Flux Enhancement in an Immersed Membrane System

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**Abstract:** The objective was to quantify the importance of operational conditions, aeration, and physico-chemical conditioning on membrane fouling intensity. The suspension filterability was also analysed by using frontal filtration and a cake filtration model. Results pointed out the moderated role of aeration to reduce compound accumulation on the membrane surface. It did not appear as a determining criterion to prevent membrane fouling. In contrast, the physico-chemical conditioning appeared as a determining criterion to increase critical flux. According to the experimental conditions $200\,l/m^2/h/bar$ membrane permeability could be maintained transmembrane pressure (TMP) when filtering stored rainwater. This permeability value was 2–3 times higher than the values obtained without conditioning. Moreover, according to the low turbidity of such stored rainwater and because of the high selectivity of the membrane, the coagulation step, a very low amount of 10 mg/l $FeCl_3$, was sufficient to intensify the filtration step. This conditioning interest appeared less significant when filtering salted water in immersed membrane systems, but a 20 mg/l $FeCl_3$ addition appeared sufficient to double the value of critical flux. Nevertheless filtration

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in frontal mode pointed out the significant impact of physico-chemical conditioning in reducing the cake deposit hydraulic resistance.

**Keywords:** Clarification, coagulation, immersed membrane, membrane permeability, salted water



## INTRODUCTION

Nowadays pressure driven membrane processes are widely applied for producing high water quality, whatever the water resource (1–2). Micro-ultrafiltration clarifies water technologically and replaces conventional water clarification by providing not only very good quality water, but also because it removes specific infectious contaminants including Giardia or Cryptosporidium. It is also strongly recommended as a pre-treatment step prior to Nanofiltration and Reverse Osmosis operations, highly sensitive to colloids and suspended solids causing bundle clogging. Micro-ultrafiltration may be carried on under highly flexible methodology including the frontal filtration mode or tangential mode when the suspensions contain a large range of compounds (3–6) obliging specific conditioning to minimize fouling (7). In water treatment most fouling is caused by suspended matter and colloid fractions deposit on the membrane surface or by pore blocking. Suspension conditioning such as coagulation-flocculation, adsorption, and oxidation steps are then favorable. Nevertheless according to raw water characteristics, it appears important to minimize the operating cost by optimizing conditioning, membrane module configuration, and hydrodynamics. Immersed membrane systems present several advantages such as selectivity and compactness due to the possibility of developing all the unitary steps of treatment (physico-chemical conditioning, PAC adsorption, separation) in the same tank. In addition, external membrane fouling can be controlled at low energy costs by practicing air injection close to the membrane bundles acting as a turbulence supplier and maintaining a high permeate flux level over relatively long operational periods, without any necessity of frequent chemical membrane cleaning (5, 6, 8). The objective of this work was to point out the role of aeration and physico-chemical conditioning when filtering stocked rainwater and salted water.

## MATERIALS AND METHODS

### Experimental Setup and Feed Water

The experiments were carried out on a lab-scale pilot (Fig. 1) where a hollow fiber bundle was directly immersed in a 35 liter tank. The immersed

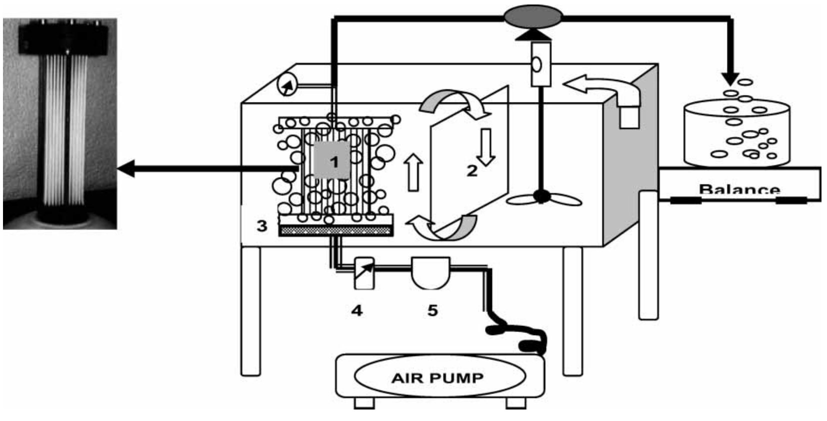


*Figure 1.* Schematic of a lab scale pilot Immersed Membrane System: (1) Capillary membrane module (2) Partition sheet (3) Air distributor (4) Air flow regulator and (5) Air filter connect with air pump.



membrane bundle was packed with polysulfone capillary fibers with a pore size of 0.1 μm and surface area of 0.1 $m^2$. The characteristics of the membrane bundle used are given in Table 1. The aeration supply was provided by a distributor placed under the membrane module. The generated bubbles raised up throughout the fibre network and the suspension were also stirred by an impeller to avoid any particle settling. Transmembrane pressure (TMP) was monitored by a negative manometer sensor placed on the permeate pipe and connected with a computer data logger.

Raw stored rainwater and synthetic salted water, mixed with 0.05 and 5.0 g/l of clay particles, were used as feed suspensions. They contain part inorganic and organic suspended solids and also some natural soluble organic matter which correspond to the characteristics given in Table 2.

*Table 1.* Membrane characteristics

| Specification | Values |
|---|---|
| Model | UFL 3 |
| External diameter of fiber (mm) | 2 |
| Number of fibers | 72 |
| Fiber length (mm) | 200 |
| Filtration surface ($m^2$) | 0.1 |
| Specific surface area ($m^2/m^3$) | 258 |
| Pore size (μm) | 0.1 |
| Initial membrane resistance (Rm, $m^{-1}$, 20°C) | 1.0E + 12 |

***Table 2.*** Feed water characteristics

| | Parameters | | | | |
|---|---|---|---|---|---|
| Type | pH | Salinity (ppt) | Turbidity (NTU) | Color (Pt-Col) | UV-254 (Abs) |
| Stored rainwater (Srw) | 7–8 | 0 | 2–5 | 10–20 | 0.11–0.14 |
| Salted clay susp. (Scs): 0.05 g/l-clay particles | 7–8 | 29–31 | 8–10 | 12 | 0.04–0.06 |
| Salted clay susp. (Scs): 5 g/l-clay particles | 7–8 | 29–31 | 1,000–1,100 | 1,250 | Not analysed |

## Operating Conditions and Membrane Fouling Prediction

The operating conditions were set up to determine the effect of aeration and coagulation on critical flux values to optimize continuous membrane separation and to evaluate fouling potential by examining raw suspensions filterability.

The pH was adjusted by adding caustic or acid solutions to be maintained in the neutral range. The temperature of feed suspension was maintained at room temperature (25–28°C). The initial permeability of the membrane module was also evaluated. The membrane performance and notably the critical flux were investigated according to the critical flux step method (9). During the continuous experimental runs, permeate recirculation was maintained with the objective of keeping the concentration of suspensions constant all along the experimental run. Different types of coagulants $FeCl_3$ Alum and PACl (Polyaluminiumchloride), were used and compared in terms of membrane fouling intensity. To optimize the coagulant addition some runs were carried on in a Jar Test procedure (150 rpm rapid mixing for 1 minute, 40 rpm slow mixing for 30 minutes, and then 30 minutes of settling). Table 3 gives the optimal dose found for each reagent.

***Table 3.*** Optimal dose of coagulant in Jar test runs

| | Optimal doses obtained (mg/l) | | |
|---|---|---|---|
| Feed suspensions tested | $FeCl_3$ | Alum | PACl |
| Stored rainwater (Srw) | 20 | 50 | 10 |
| Salted clay suspension (Scs): 0.05 g/l clay particles | 20 | 100 | not analysed |
| Salted clay suspension (Scs): 5.0 g/l clay particles | 20 | 100 | not analysed |

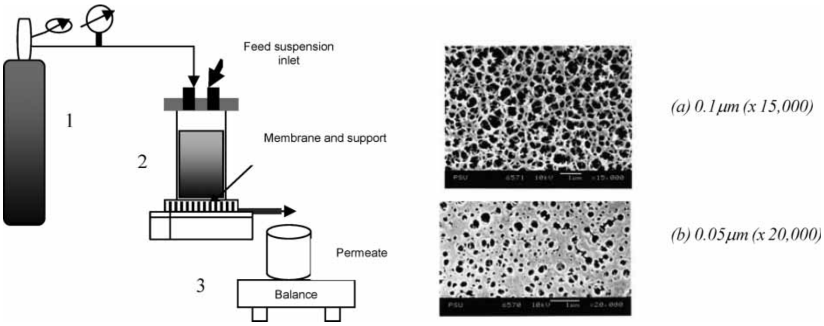


***Figure 2.*** Schematic diagram of frontal filtration unit and membrane surface by SEM: (1) = $N_2$ tank equipped with pressure regulator (2) = Pressurized filtration cell and (3) = Balance and permeate receiver.



A specific membrane cleaning procedure was practiced with different cleaning periods: rinsing with water, back washing with a 1 wt.% citric acid solution at low $15\,l/m^2/h$ rates for 60 minutes. Membranes were then directly immersed in a 1 wt.% sodium hydroxide solution for 120 minutes. The membrane permeability was measured after each cleaning step by filtering tap water to evaluate the cleaning step performance.

The filterability and fouling potential of raw suspensions were analyzed by using a frontal filtration mode in a specific filtration cell described in Fig. 2. The unit consisted of a 150 ml pressurized filtration cell equipped with plane organic membranes. The characteristics of membranes are given in Table 4 and two photos of used membranes are given in Fig. 2.

***Table 4.*** Plane membrane characteristics

| | Type | |
|---|---|---|
| Membrane material | VMWP 04700 Millipore plane nitrocellulose | VCWP 04700 Millipore plane mixed cellulose ester |
| Dimension (mm, diameter) | 47 | 47 |
| Filtration area ($cm^2$) | 11.3 | 11.3 |
| Pore size (μm) | 0.05 | 0.1 |
| Filtration layer | | |
| - Porosity (%) | 72 | 74 |
| - Thickness (μm) | 105 | 105 |
| Water permeability (20°C, 1 bar) ($l^{-1}\,h^{-1}m^2$) | 400 | 800 |
| Membrane resistance $R_m$ ($m^{-1}$) | $1.0 \times 10^{12}$ | $0.5 \times 10^{12}$ |

The cake filtration theory was used to examine and explain the effect of the various components tested in suspension. The methodology consisted of following up the cumulated volume of filtrate during filtration time for given TMP at 0.25 bars without any applied turbulence. The changes in the filtered volume can be described according to the cake filtration law, in which the ratio t/V is a linear function of V:

$$\frac{t}{V} = \frac{\mu \times \alpha \times W}{2 \times \Delta P \times \Omega^2} \times V + \frac{\mu \times R_m}{\Delta P \times \Omega} \tag{1}$$

Where W macromolecule or particle deposit mass per cumulated volume of filtrate $kg \cdot m^{-3}$; ΔP transmembrane pressure Pa; $R_m$ initial membrane resistance $m^{-1}$; V cumulated volume of filtrate $m^3$; t time s; $\alpha$ specific resistance $m \cdot kg^{-1}$; $\mu$ dynamic viscosity $Pa \cdot s$; Ω membrane area $m^2$.

## Sample Analysis

To quantify the performances of separation and characterizing the feed suspension and permeate, different parameters were evaluated by using methods given in Table 5.

## RESULTS

To point out the role of suspended solid concentration and salinity in membrane fouling intensity, the experiments were successively carried on with natural stored rainwater and synthetic salted water (representative of sea water). Stored rainwater (Srw) came from Prince of Songkla University reservoir. The critical filtration condition was identified by increasing the permeate flux step by step, and measuring the associated TMP value obtained for each step. Results obtained are shown in Figs. 3–5. Figure 3 presents the variation of mean TMP with permeate flux according to different air flow-rates. It can be observed that a critical flux value was

***Table 5.*** Parameters and analytical methods

| Parameters | Methods |
|---|---|
| Turbidity | Turbidity meter-HACC |
| Color | Spectrophotometric method (10) |
| pH | pH meter |
| NOM (humic acid) | Spectrophotometric method using humic acid as the standard solution at a wavelength of 254 nm |
| Particle size | Particle size analyzer-MALVERN |

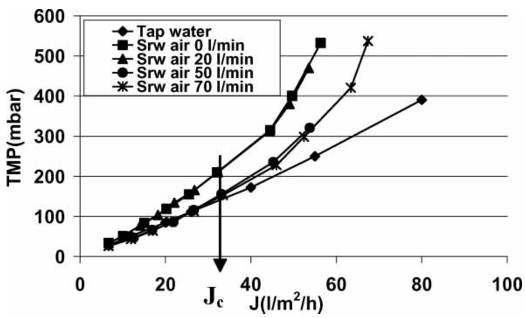


*Figure 3.* Influence of air flow rate on critical flux values of stored rain water (Srw: without coagulation).

maintained in the range of 30–35 $l/m^2/h$ whatever the air-flow rates. When stored rainwater was conditioned by the adding of different types of coagulants, the results clearly showed the positive effect of this conditioning (Fig. 4).

The average permeability remained close to the obtained value when filtering tap water, i.e. close to 200 $l/m^2/h/bar$ corresponding to an intensification of filtration by a factor of 2 or even 3. Results also point out the benefit of using $FeCl_3$ to control fouling evolution. The optimal dose determining to minimize the sludge production appeared very low, 20 mg/l in comparison with the values obtained with Alum and PACl. Furthermore, the optimal 10 mg/l dose of $FeCl_3$ observed in the membrane system appeared two times lower than the dose obtained in the Jar Test that pointed out the role of membrane selectivity in comparison with gravitational settling. The former only necessitates particle coagulation and the formation of small flocs, while settling necessitates the formation of larger flocs able to settle. The flocculation step appears also necessary. In addition, air injection did

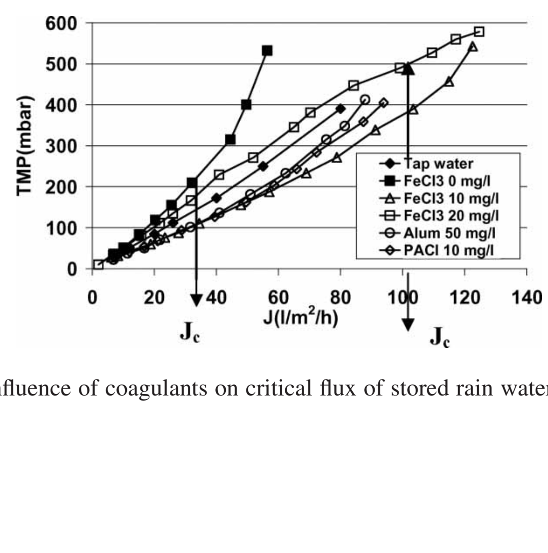


*Figure 4.* Influence of coagulants on critical flux of stored rain water (Srw: without aeration).

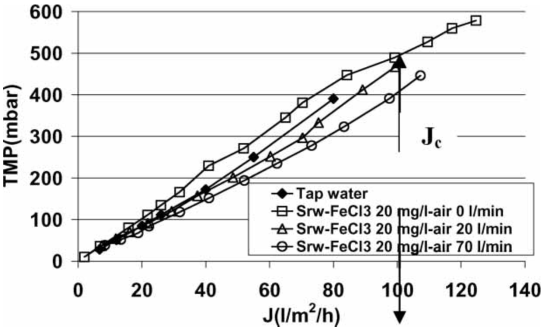


***Figure 5.*** Influence of air flow rate on critical flux of stored rain water conditioned with optimal dose of $FeCl_3$.



not actually modify the intensification of permeate flux as shown in Fig. 5, illustrating results obtained with $FeCl_3$ conditioning.

Results obtained when filtering salted clay suspensions (Scs) are presented in Figs. 6 and 7. The salinity of salted suspension was close to the value of sea water (30 to 35 g/l) and the role of suspended particles was analyzed by adding clay powder at two concentrations—a low 0.05 g/l concentration to compare with results obtained with stored rainwater and a high 5 g/l concentration to point out the role of suspended solids. Without any conditioning, the critical flux procedure shows a lower critical flux value (20 $l/m^2/h$) and lower membrane permeability in comparison with the value obtained with stored rainwater (Fig. 3). The suspended solids concentration in salted water does not influence the intensity of filtration. In contrast to results obtained with stored rainwater, the addition of coagulants only allowed a small increase of 20% in membrane permeability but it remained significantly favourable to the increased critical permeate flux that reached 40 $l/m^2/h$ at a high rate of

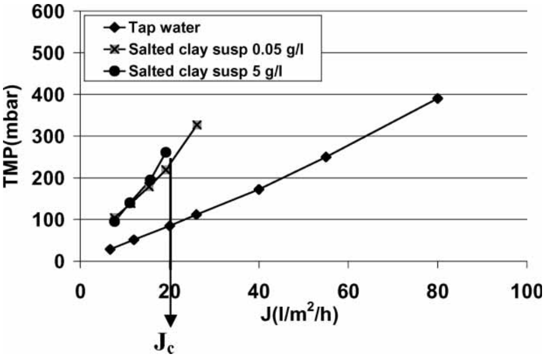


***Figure 6.*** Critical flux determination of salted clay suspension (Scs: without coagulation).

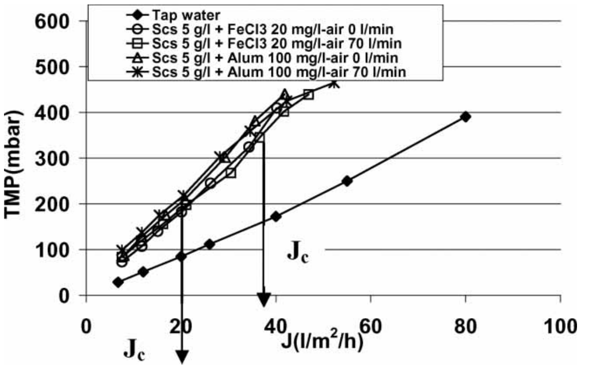


*Figure 7.* Influence of coagulants and air flow rate on critical flux values when filtering salted clay suspension.

aeration supplied at 70 l/min (Fig. 7). This weak impact of conditioning is probably due to the ionic strength of the salted suspension modifying the interaction between suspended solids and coagulants and the destabilizing compounds to favor less intensive particle encountering.

The filtered water showed a very high quality of water clarification with turbidity values lower than 0.5 NTU and colour values lower than 12 Pt-Co for all suspension tested.

Figures 8 and 9 show results obtained when filtering salted suspensions in frontal mode inside the specific filtration cell described in Fig. 2. To evaluate the filterability of the suspension, we suppose that the determinant step reducing membrane permeability was cake formation due to particle retention on the membrane surface. Then a specific cake hydraulic resistance ($\alpha$W) may be evaluated by using cake filtration relation. The evolution of t/V vs V, in Figs. 8 and 9, allows slope calculation and ($\alpha$W) deduction (t appears as the time and V the permeate volume obtained at time t).

Table 6 presents the ($\alpha$W) values obtained on both porous membranes (described in Table 4) for different experimental tested conditions (clay addition and specific conditioning). As expected, the specific hydraulic resistance criteria ($\alpha \cdot$ W) appeared as directly proportional to suspended particle concentration when filtration was carried out without conditioning ($\alpha$ being proportional to the particle concentration). Then, results pointed out the great benefit of conditioning that allowed the transformation of small particles in large flocs inducing a significant reduction of the specific resistance criteria ($\alpha \cdot$ W) by a factor of 20 to 100 or more, when working with high concentrated suspensions. The relatively small values of ($\alpha \cdot$ W) criteria are significant to the presence of large cake porosity and weak deposit compactness in the chosen working conditions.

When suspension conditioning occurred no significant differences appeared for ($\alpha \cdot$ W) values, whatever the conditions. The presence of such

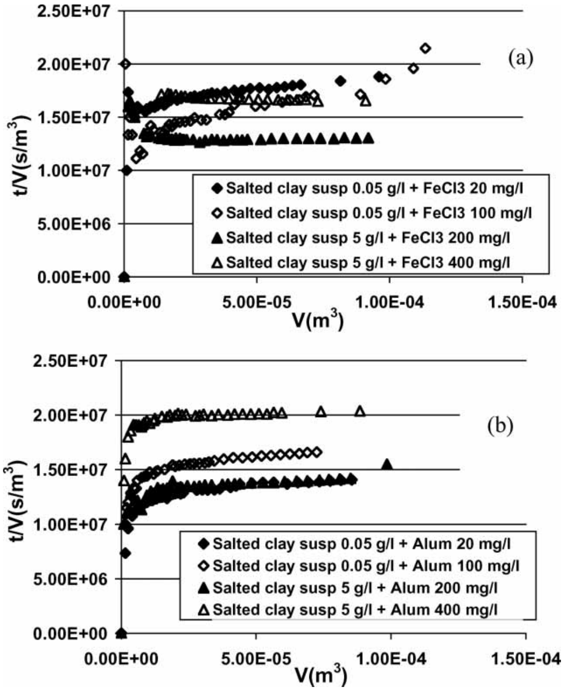


***Figure 8.*** Evolution of filterability (t/V vs V) of salted clay suspension conditioned with $FeCl_3$ (a) and with Alum (b): membrane 0.05 micron-TMP 0.25 bar.



a cake deposit on the membrane surface may represent a dynamic layer which avoids any entry of small particles into the membrane pores, thus having a positive impact to reduce irreversible membrane fouling. In any case the membrane in cellulose ester presents lower hydraulic resistance values than nitrocellulose that can translate

(1) the deposit cake resistance which is negligible in comparison with initial membrane resistance, two times higher for nitrocellulose, (see Table 4), and;
(2) the importance of membrane material in fouling intensity.

These results obtained in frontal mode, point out the determining role of coagulants on specific hydraulic resistance of cake deposits. These appear in contrast to results previously obtained in immersed systems. This shows in fact that the determining fouling phenomenon in the immersed system was not an external deposit, which explains the non-influence of aeration, but more specifically, the irreversible interactions between non-flocculated soluble compounds with membrane materials.

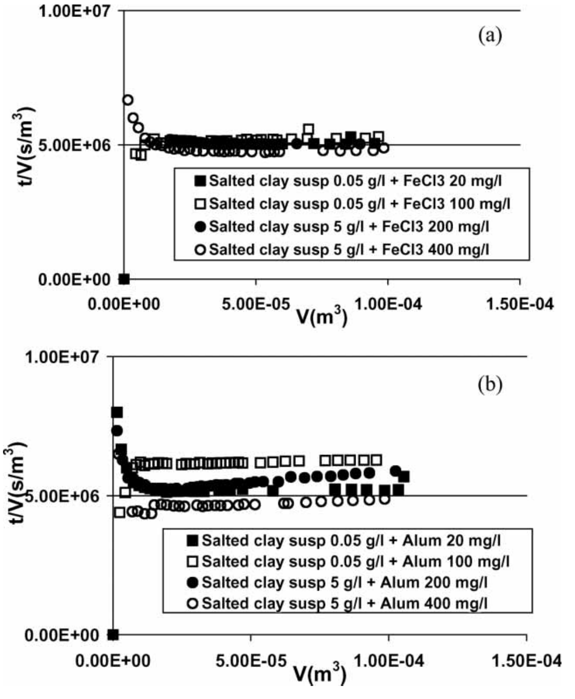


***Figure 9.*** Evolution of filterability (t/V vs V) of salted clay suspension conditioned with $FeCl_3$ (a) and with alum (b): membrane 0.1 micron-TMP 0.25 bar.



***Table 6.*** $\alpha \cdot W$ values obtained from dead end filtration of conditioned stored rainwater and salted clay suspension tested at TMP 0.25 bar

| | $\alpha \cdot W$ ($\times 10^{12}$ m$^{-2}$) | | | | | |
|---|---|---|---|---|---|---|
| | Stored rainwater: (Srw) | | Salted clay suspension: (Scs) 0.05 g/l | | Salted clay suspension: (Scs) 5.0 g/l | |
| Type of coagulants | 0.05 μm | 0.1 μm | 0.05 μm | 0.1 μm | 0.05 μm | 0.1 μm |
| Without coagulants | 3.4 | 1.7 | 10 | 1.1 | 110 | 110 |
| $FeCl_3$ = 20 mg/l | 1.1 | 0.28 | 4.25 | 1.42 | — | — |
| $FeCl_3$ = 100 mg/l | 1.1 | 0.34 | 4.96 | 0.71 | — | — |
| Alum = 20 mg/l | 3.4 | 0.5 | 3.54 | 0.71 | — | — |
| Alum = 100 mg/l | 1.1 | 2.5 | 5.66 | 0.71 | — | — |
| $FeCl_3$ = 200 mg/l | — | — | — | — | 0.71 | 1.42 |
| $FeCl_3$ = 400 mg/l | — | — | — | — | 3.54 | 0.02 |
| Alum = 200 mg/l | — | — | — | — | 2.83 | 0.64 |
| Alum = 400 mg/l | — | — | — | — | 4.25 | 0.64 |

## CONCLUSIONS

The Immersed Membrane System appeared as an appropriate operation in order to obtain very high water clarification quality, whatever the raw water quality, even salted water, which justifies its development for water clarification and its benefits of pre-treatment upstream, a desalination step by reverse osmosis for example. In the operational conditions tested, water conditioning by $FeCl_3$ appeared very efficient for clarifying stored rainwater that pointed out the determining role of cake deposits when no conditioning occurred. When treating flocculated salted water, significant differences were observed when comparing frontal and tangential filtration modes. The determining role of free soluble compounds or initial membrane resistance has an effect on membrane fouling.

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