

Removal of Total Suspended Solid and Polysaccharide in Seawater using Polysulfone Ultrafiltration Membrane

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Abstract. The aim of this research measured the total removal of suspended solids and organic material in seawater using a polysulfone (PSf) ultrafiltration (UF) membrane. The results indicate that the salt concentration was significantly affected membrane flux. The lower flux of the membrane was obtained by a high content of the salt concentration. The fouling potential in decreasing the flux value was more dominant in the polysaccharide feed than the TSS feed. However, the fouling potential occurs more obviously in the mixed feed (TSS + polysaccharide), resulting in lower flux and higher rejection. PSf UF membranes successfully remove 93% of polysaccharides in a single feed, 95% in mixed feed, and 100% of suspended solids in a single feed and mixture.

Keywords: Polysaccharide; Polysulfone; Seawater; Total Suspended Solid

1. Introduction

Nowadays, many countries worldwide suffer from the scarcity of freshwater resources for industrial and agricultural purposes. Furthermore, several illnesses are associated with contaminated drinking water. To overcome this problem, seawater desalination is an alternative technology to produce freshwater (Khawaji et al., 2008). However, seawater cannot be consumed directly due to seawater has high inorganic and organic compounds (Ghaffour et al., 2013). Mostly, natural organic matter (Natural Organic Matter / NOM) in seawater can react easily with chlorine. Byproducts of the reaction are trihalomethanes (THMs), haloacetic acids (HAAs), and another halogenated organic that is carcinogenic. For this reason, seawater is necessary for treatment using some separation methods such as coagulation, flocculation, media filtration, and membrane process (microfiltration/ultrafiltration) (Vial et al., 2003).

Several studies have recently reported the high performance of MF/UF membranes as seawater pre-treatment (Ebrahim et al., 2001; Woo et al., 2015; Xu et al., 2010). Membrane technology is an efficient and cost-effective method to conventional separation processes. The pressure-driven membrane process was classified according to retain particle size into four categories such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). MF and UF membrane has high performance in removing total suspended solids and organic material. However, the main problem of membrane usage is fouling. Therefore, fouling control is a crucial step in membrane used.

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In this study, the feed consist of a single feed of suspended solids (kaolin), a single feed of polysaccharides (sodium alginate), and a mixed feed of both (kaolin + sodium alginate). The specific studies in high salinity environments such as seawater, particularly with variations in the range of high salt concentrations (10,000 mg NaCl / L, 20,000 mg NaCl / L, and 30,000 mg NaCl / L) and varying salinity distributions have not been widely carried out. This research was measured the total removal of suspended solids (TSS) and polysaccharides in seawater using MF and UF membrane.

2. Methods

2.1. Materials

The polyethersulfone MF (MicroPES) and polysulfone UF (GR61PP) flat sheet membrane were used in this study. The flat sheet membrane had an MWCO of 0.04 – 0.12 μm and 20 kDa for MF and UF, respectively. NaCl, NaC6H7O6 (sodium alginate as a polysaccharide), C₆H₅OH, H₂SO₄ were purchased from Merck Inc., Germany. Kaolin as TSS was obtained from a local chemical store in Semarang.

2.2. Filtration system

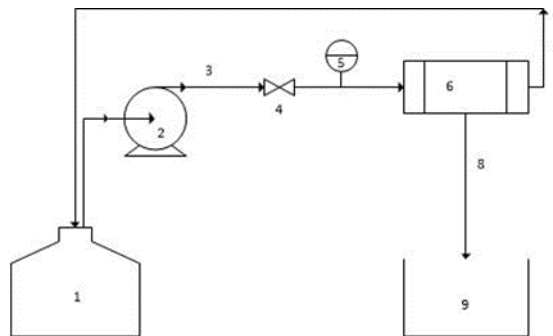


Figure 1 Schematic diagram of the membrane filtration system. (1: Feed tank, 2: Pressure pump, 3: Feed flow, 4: Valve, 5: Pressure gauge, 6: Membrane housing, 7: Concentrate flow, 8: Permeate flow, 9: Permeate)

A crossflow filtration set-up was used in the present study, as shown schematically in Fig. 1. The filtration experiment was conducted at a constant pressure of 0.5 bar and 1 bar for MF and UF membrane, respectively. Sodium alginate and kaolin (100 mg/l for MF and 10 mg/l for UF) were used as feed solution. Before the filtration, the membrane was compacted for 30 min. The permeate sample was gravimetrically 4 min interval. A small amount of samples were taken from the feed, permeate, and final concentrate for analysis.

2.3. Analytical methods

Sample from the feed, permeate, and concentrate was taken for the determined concentration of TSS and polysaccharide using spectrophotometry UV-Vis (Thermo Fisher Scientific, Genesys 20). Meanwhile, the presence of TSS and polysaccharide concentration by applied membrane process was showed in terms of % rejection and flux. The rejection of the main component was calculated using Equation (1):

$$R = 1 - \frac{C_p}{C_f} \times 100\% \quad (1)$$

Where C_p represents the concentration of a component in the permeate stream and C_f is the concentration of components in the concentrate stream. The permeate flux (J) is the volume of permeate collected per unit area membrane (A) and per unit time (t) as presented in Equation (2):

$$J = \frac{V}{Axt} \quad (2)$$

On the other hand, SEM (JEOL JSM-6510 LA) was used to observe the morphology of the fresh and fouled membrane surface. FTIR (Spectrum Two FTIR Spectrometer PerkinElmer, USA) measurements were used to analyze membrane top surface, and the recorded wavelength ranged between 400 and 4000 cm^{-1} .

3. Results and Discussion

3.1. Effect of Salt Concentration in Feed Solution on Membrane Performance

3.1.1. Salt Solution containing total suspended solids

Profiles flux (J_w/J_o) resulted with feed 100 mg/L kaolin for membranes Microfiltration Polyethersulfone (PES) filtration and 10 mg/L kaolin for Ultrafiltration membranes Polysulfone (PSf) filtration can be seen in Figure 2.

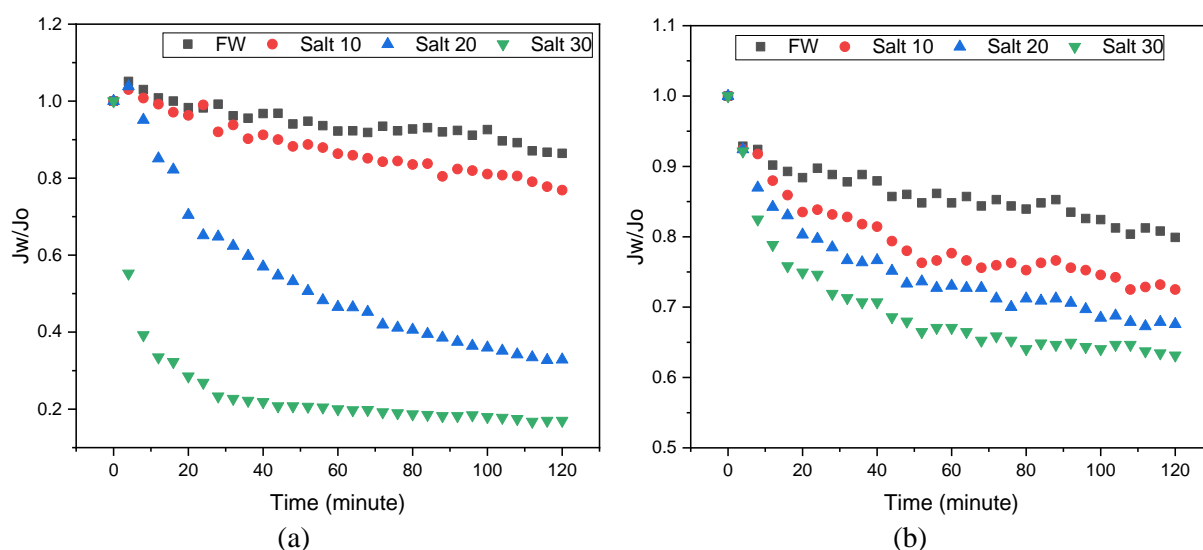


Figure 2 Profile flux of (a) 100 mg/l kaolin feed solution at various salt concentration using PES microfiltration membrane (b) 10 mg/l kaolin feed solution at various salt concentration using PSf ultrafiltration membrane

Figure 2 showed that, in general, the microfiltration and ultrafiltration membranes flux profile decreased during the operating time of 120 minutes. The lowest flux profile was on Salt 30 (30,000 mg salt solution of NaCl / L).

On Fresh Water (FW), kaolin likely to caused concentration polarization where kaolin covering the surface of the membrane. Concentration polarization was more easily swept away by the flow of crossflow. PSf ultrafiltration membranes flux profile decreased significantly by increasing salt concentration in the feed solution. From the pictures above, it appears that over time, a more dominant potential fouling occurred at higher salt concentrations. The higher the salt content, the more aggregates were formed and attached to the membrane's surface to lower the value of the flux profile. This founding was also consistent with [Song and Singh \(2005\)](#).

Under standard conditions with a plain water solution, kaolin always has a negative charge. The electrical double layer on the surface of the kaolin molecule initially led the negative charge into neutrality. That caused the decline of electrostatic repulsion between the kaolin and the membrane surface and supported kaolin's aggregation of the membrane's surface to form fouling.

TSS rejection in a single feed can be seen in Table 1.

Table 1 Number of receptors in each container

Permeate	TSS Rejection (%)	
	MF PES	UF PSf
Fresh Water	92.08	100
Salt 10	92.08	100
Salt 20	92.20	100
Salt 30	92.74	100

From Table 1, it can be seen that the membrane microfiltration PES can set aside TSS more than 92%, while for both ultrafiltration membrane can set aside 100%. The ability of membrane performance can explain this phenomenon. Ultrafiltration can remove the material with a smaller size.

Membrane pore size also affected the amount of rejection as Lau (2013) stated that the larger the pores, the lower the rejection rate. It caused due to the separation proceeds in the membrane was a sieving mechanism. It means that the particles in the feed with a smaller size than the membrane pores will escape, while particles larger than the membrane pores will be retained. Kaolin has a particle size of about 350 nm with a 100 kDa \approx 10 nm (Jermann et al., 2007). This separation mechanism occurred either at the membrane surface or inside the membrane.

3.1.2. Salt Solution containing polysaccharide

Figure 3 shows the results for profile flux (J_w/J_0) with a feed of 50 mg/L sodium alginate membranes Microfiltration Polyethersulfone (PES), and Ultrafiltration membranes Polysulfone (PSf).

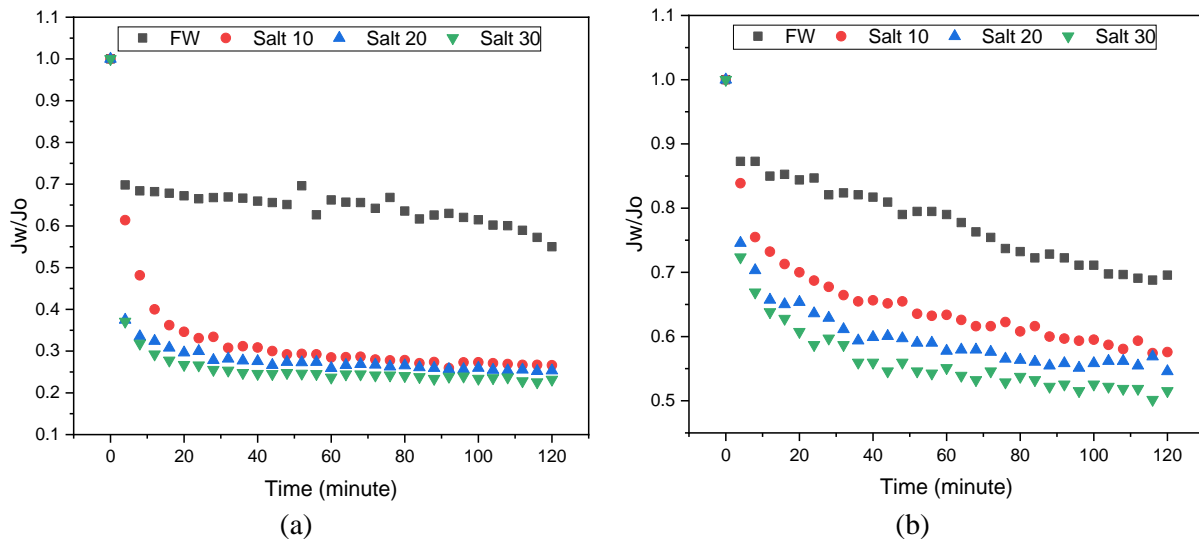


Figure 3 Profile flux of 50 mg/l sodium alginate feed solution at various salt concentration using (a) PES microfiltration membrane (b) PSf ultrafiltration membrane

Figure 3 showed that during the operating time of 120 minutes on each type of membrane, the flux profile decreased over time. The influence of collected particles on the surface of the membrane at the first minute was quite significant. As time went by, the foulant movement on the membrane surface was diminished. So its effect on reducing the flux profile was also reduced. The higher the salt content in the feed also gave a stronger effect on alginate fouling development, as a resulted flux profile also become lower. While in the microfiltration membrane, an increase in the salt concentration was not caused a significant decreased in the flux profile.

Resosudarmo et al. (2013) previously stated that a high salt concentration in sea water could significantly increase the potential fouling by organic material. Jermann et al. (2007) pointed out that sodium alginate membranes' interaction can produce strong electrostatic repulsion. That is

because both of them are negatively charged (Ven et al., 2008). The presence of ions, on the other hand, will greatly reduce the impact of the charge on the membrane, enhancing the contact between the membrane and the alginate molecules. It caused the electrostatic repulsion between molecules alginate and between molecule alginate-membrane surface to decrease (Lee et al., 2006; Ven et al., 2008 and Resosudarmo et al., 2012). According to Listiarin et al. (2011), salt's addition to a solution containing sodium alginate cause a gel formation that will increase the aggregate size of the sodium alginate. It explains how the effect of high saline conditions caused alginate fouling development during the filtration process.

Polysaccharide rejection in a single feed can be seen in Table 2.

Table 2 Polysaccharide rejection in a single feed

Permeate	Polysaccharide Rejection (%)	
	MF PES	UF PSf
Fresh Water	53.67	93.16
Salt 10	47.83	93.68
Salt 20	57.12	94.99
Salt 30	60.45	97.35

Table 2 shows that the PES membrane microfiltration can set aside approximately 50-60% sodium alginate, while both ultrafiltration membranes can set aside more than 85 percent sodium alginate. The sieving mechanism occurred during filtration using a membrane unit when the separation system was based on the membrane's pore size.

Sodium alginate has a molecular weight between 12-80 kDa (Jermann et al., 2009). Since the particle size of sodium alginate was similar to the ultrafiltration membrane's pore size, it was more effective to use ultrafiltration membranes to extract it. But it also contributed to a higher fouling potential as sodium alginate particles that accumulated near the surface of the membrane and surrounded the membrane pore able to degraded the membrane performance. On the other hand, the microfiltration membranes tend to pass sodium alginate molecules. However, the alginate's molecular weight can also be reached 0.20 μm . That enabled the MF membrane to restrain alginate molecules partially. Another problem that causes severe fouling was the adsorption of particles into the membrane, which downsizing the membrane pores.

3.1.3. Salt Solution containing total suspended solid and polysaccharide

Profiles flux (J/J_0) were obtained using a feed of 100 mg / L kaolin + 50 mg / L sodium alginate membranes. Microfiltration is a term used to describe the method Ultrafiltration membranes with polyethersulfone (PES) and 10 mg/L kaolin + 50 mg/L sodium alginate.

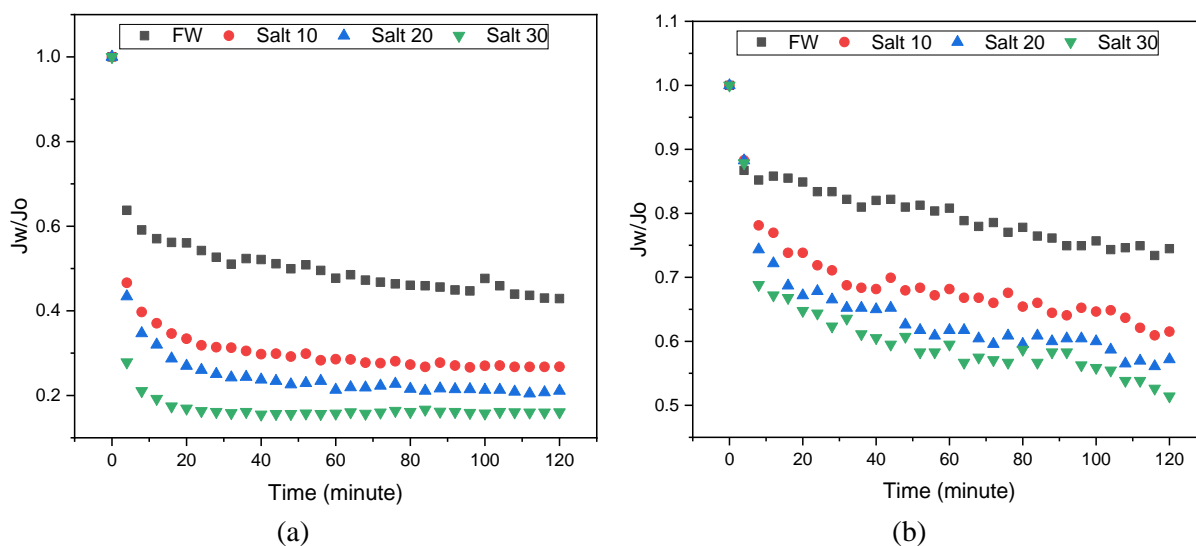


Figure 4 Profile flux resulted in 100 mg/l kaolin and 50 mg/l feed solution containing sodium alginate at various salt concentration using (a) PES microfiltration membrane (b) PSf ultrafiltration membrane

Figure 4 illustrates how the higher the salt content in the feed solution, the more robust the fouling effect on each membrane, resulting in a lower flux profile over time. The presence of sodium alginate and kaolin also raised the potential for greater fouling. The membrane with the mixed feed of kaolin and sodium alginate compared to the feed kaolin or sodium alginate only, the flux profile's tendency was similar to sodium alginate.

As already explained in the previous section how the interaction between the kaolin and sodium alginate with a high salt content resulted in a significantly decrease flux profile. It can be concluded fouling potential increased with the increasing salt concentration. Kaolin cake layer and alginate gel formation have resulted from the dense fouling from increasing salt concentration.

The feed stream with high ionic strength can compress the electrical double layer so that the electrostatic forces between particles were largely suppressed and non-electrostatic interactions become dominant (Motsa et al., 2015). The presence of sodium alginate and kaolin simultaneously produced thicker fouling on the membrane surface so that its rejection could be increasing. The deposition process of particles on the membrane surface will form a gel layer commonly referred to as a secondary membrane (Lopes et al., 2005). However, other things were also able to affect the rejection, such as molecules and membrane pore size.

Based on the results of the study, it showed rejection kaolin and polysaccharides in salt solution containing that mix feed can be seen in Table 3 and Table 4.

Table 3 Rejection kaolin in salt solution containing that mix feed

Permeate	TSS Rejection (%)	
	MF PES	UF PSf
Fresh Water	91.86	100
Salt 10	91.86	100
Salt 20	91.86	100
Salt 30	91.86	100

Table 4 Rejection polysaccharides in salt solution containing that mix feed

Permeate	Polysaccharide Rejection (%)	
	MF PES	UF PSf
Fresh Water	70.54	95.05
Salt 10	67.67	95.52
Salt 20	73.67	96.15
Salt 30	86.39	97.39

From Table 3 and Table 4 it can be seen that the membrane microfiltration PES can set TSS aside around 92%, while both of the ultrafiltration membranes can eliminate 100% of TSS. PES membrane microfiltration can be set aside around 70% for polysaccharides, while the average rejection for PSf ultrafiltration membranes was about 96%.

Based on Table 4 as compared to Tables 1 and 2, the rejection of sodium alginate on all membranes is higher in the feed mixture of kaolin with sodium alginate. The fouling formed on the membrane surface is greater, built up as a secondary layer. Ion concentration in the feed can also affect rejection, as described in the previous section, where the higher concentration of ions then will increase fouling development.

3.2. Effect of TSS for Ultrafiltration Membrane Performance

The addition of kaolin, as in the previous discussion, may lead to concentration polarization or aggregates' formation. Based on the results of the comparison to the MF filtration, it can be

seen that the flux profile with feed kaolin (mixture) decreased lower than feed without kaolin (only sodium alginate). However, filtration UF showed opposite results. Profile flux both coincide; even with flux profile of kaolin mixture solution tends to be above the solution without kaolin. It showed that a small amount of kaolin concentration in UF filtration (10 mg/l) did not significantly influence. To prove this assumption, test flux with the addition of kaolin mixed solution of 50 mg/l in the salt solution (30,000 ppm NaCl) and FW (Freshwater / Water Flute). The flux profile results were compared to the addition of kaolin mixed solution 0 mg / l and 10 mg / l.

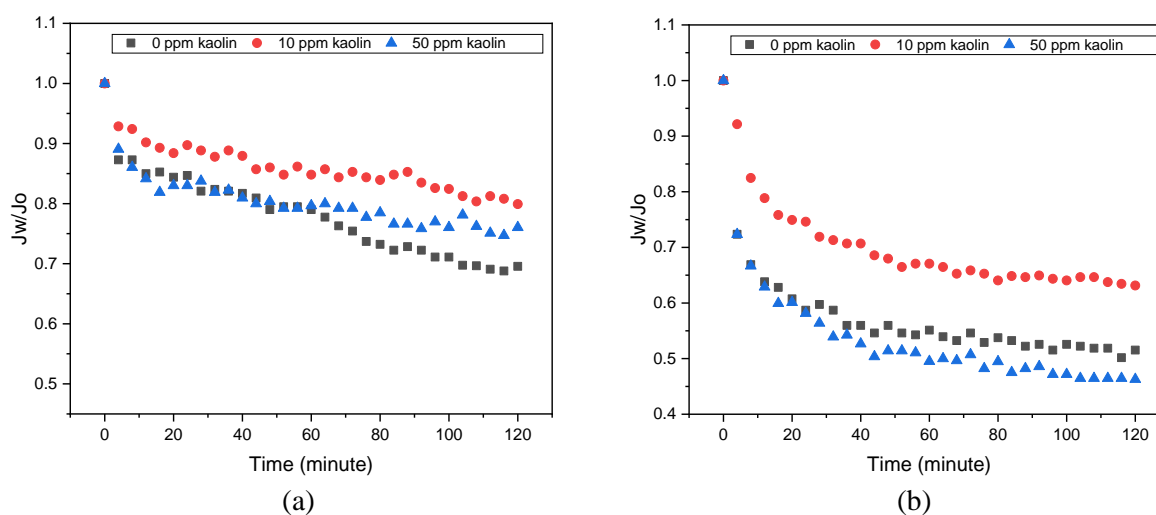


Figure 5 Profile flux with feed solution; Sodium Alginate, Kaolin, and Mixed in Fresh Water using (a) PES microfiltration membrane (b) PSf ultrafiltration membrane

According to the flux profile test results on both UF membranes in fresh water (see Figure 5), all feed solutions with 0 ppm kaolin (sodium alginate feed only), 10 ppm kaolin, or 50 ppm kaolin had a higher flux profile than the salt solution. Unlike in the salt solution, the decline in flux profile becomes a bit lower as the higher kaolin concentration was added.

A decreased in the flux profiles at both UF membranes between the concentration of a single feed to a mixture of 10 ppm kaolin in salt solution was so small that there's no significant difference in the results. It can occur because the kaolin feed concentration was very small, so it was more easily swept away by the crossflow and did not contribute to fouling. But after kaolin was added to 50 ppm in salt solution feed, the flux profile on the membrane filtration was significantly lower. It was consistent with [Jermann et al. \(2007\)](#), which states that the flux profiles with a feed mix will contribute to greater fouling than a single feed. These results were also consistent with previous research, which states that the reduction in profile flux significantly demonstrated by the fouling combination between colloidal silica and organic material in connection with the increase in osmotic pressure at the membrane surface ([Lee et al., 2005](#); [Li and M. Elimelech., 2006](#)).

Table 5 Rejection TSS and polysaccharides in freshwater containing that mix feed

Kaolin (mg/L)	Rejection (%)	
	TSS	Polysaccharide
0	100	93.16
10	100	-
50	100	95.42

Table 6 Rejection TSS and polysaccharides in salt solution containing that mix feed

Kaolin	Rejection (%)
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(mg/L)	TSS	Polysaccharide
0	100	97.35
10	100	-
50	100	97.64

Based on Table 5 and Table 6 for PSf ultrafiltration membrane, the rejection of sodium alginate increased with the addition of kaolin in the feed solution, followed by a decline in flux profile. On filtration in a salt solution, adding kaolin with higher concentrations did not increase the rejection of sodium alginate through flux profile also decreased, indicating increased fouling potential—besides, the rejection in freshwater also greater than the salt solution. In contrast, a decrease in the freshwater flux profile is lower than the saline solution.

When their alginate molecules are trapped in a kaolinite structure, they can not diffuse back (Jermann et al., 2007). According to Jermann, it will intensify the alginate-pore blocking and adsorption on the membrane. It also allowed the alginate molecules smaller than the pore membrane to permeate to pass together. It explained the phenomenon of lower rejection in sharper declined flux profile in the salt solution. As for the phenomenon that occurred in freshwater, this may be caused by the cross-flow as described earlier. So it was possible to maintain high profile flux and greater rejection value in the freshwater feed solution.

3.3. Fouling characterization on the ultrafiltration membrane surface

3.3.1. Fourier Transform Infrared Spectroscopy (FTIR)

FTIR test was to find out the fouling composition on the membrane surface. Figure 6 shows the FTIR test results for polysulfone ultrafiltration membranes with sodium alginate feedback on Fresh Water and 30,000 mg NaCl / L Salt Solution.

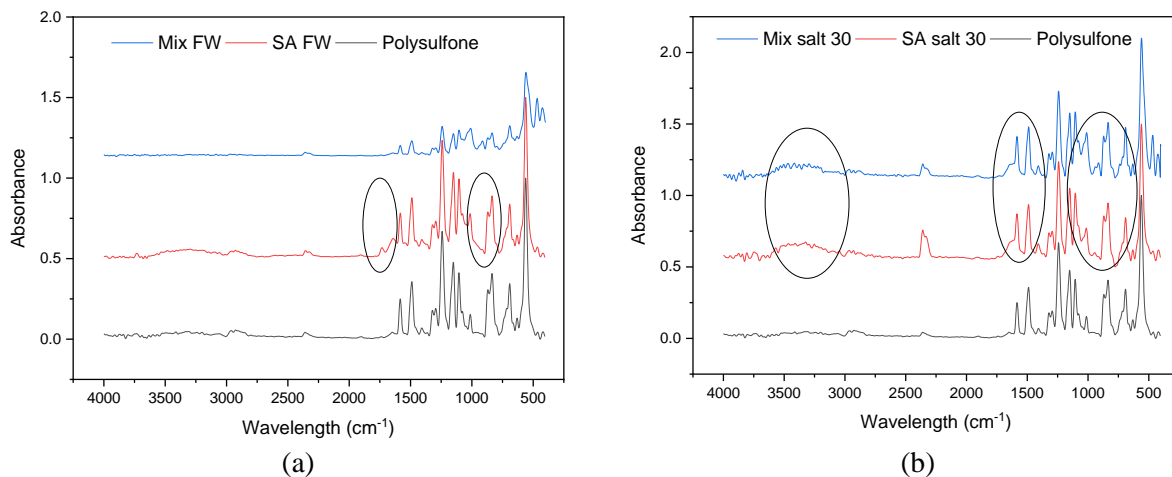


Figure 6 The FTIR result of polysulfone ultrafiltration membranes with sodium alginate feedback (a) Fresh Water (b) Salt Solution.

The presence of peaks of unique wavelengths can be seen in the four figures above, with the most important peak occurring in salt solution as compared to freshwater, which is the area about $3500\text{--}3200\text{ cm}^{-1}$, which indicates the presence of hydroxyl groups (OH) bonded to hydrogen, and at $3100\text{--}3000\text{ cm}^{-1}$, which indicates the presence of aromatic groups (CH). Wavenumber $1760\text{--}1665\text{ cm}^{-1}$ indicate the presence of a carbonyl group (C = O) as the aromatic group, $1680\text{--}1640$ shows the alkene group (-C = C-), $1600\text{--}1585\text{ cm}^{-1}$ shows an aromatic group (CC), $1320\text{--}1000\text{ cm}^{-1}$ indicates the presence of carboxyl group (CO). The alginate, according to Mury et al. (2005) in Mutia et al. (2011), is a natural polymer with aromatic groups (ROR) containing -OH, -COOH and -CH, -C = C- and -C = O. Isomer of sodium alginate lies in the absorption peak in 1614 cm^{-1} and 1431 cm^{-1} . According to Stuart (2004), the presence of an OH group in the $3800\text{--}3400\text{ cm}^{-1}$ range for kaolin and Si-O groups in the $1300\text{--}400\text{ cm}^{-1}$ range for kaolin indicate the presence of

OH stretching and bending. The content of Al (III) in the kaolin will form a strong bond at 1120-1000 cm^{-1} region. Thus, the results obtained FTIR spectra showed that the fouling caused by sodium alginate is more dominant in the feed salt solution was 30,000 mg NaCl / L compared with Fresh Water.

3.3.2. Scanning Electron Microscopy (SEM)

SEM analysis showed the form and change in surface morphology of the samples analyzed. In polysulfone ultrafiltration membrane filtration, the fouling behavior of each foulant was revealed by SEM results. The results of filtration by PSf ultrafiltration membrane are presented in the figures below.

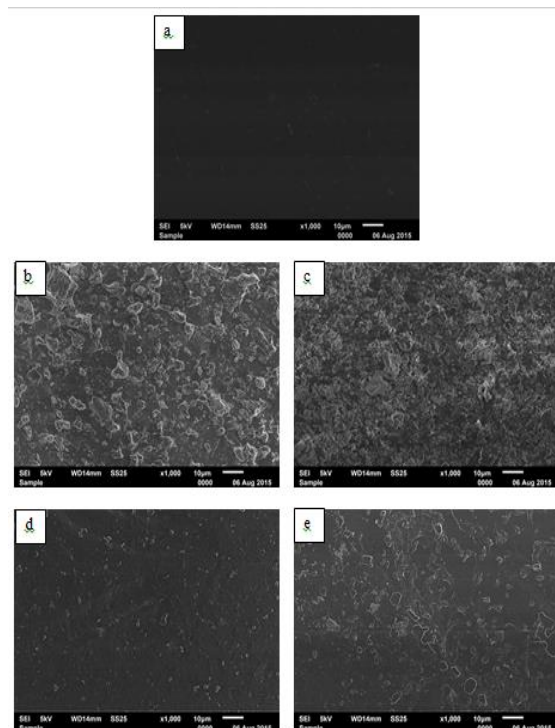


Figure 7 UF Membrane SEM results in Polysulfone Filtration; a] New Membrane; b] Mix (sodium alginate 50 ppm and 10 ppm kaolin) in Salt 30; c] 50 ppm Sodium Alginate in Salt 30; d] Mix (sodium alginate 50 ppm and 10 ppm kaolin) in FW; e] 50 ppm Sodium Alginate in FW

By comparing all the figures above, it showed the difference between the results of SEM before filtration membrane with the membrane after filtration using a solution of a mixture of kaolin and sodium alginate and a single sodium alginate solution. Different phenomena showed on the condition of Fresh Water. With a solution of salt 30,000 mg NaCl/L, the presence of high salt content caused the foulant to bind to each other and formed aggregates that produced more large effect fouling compared to normal conditions (Fresh Water).

In the mixed feed, it appeared that the accumulated fouling on the membrane surface was a more dominant place in the kaolin mixture with sodium alginate compared with a single sodium alginate solution. In the salt solution, a fouling layer formed more likely to have a larger structure. It can be caused due to the condition of ionic, kaolin, and sodium alginate more easily bind to form aggregates (enlarge molecular structure). With a high ion condition, kaolin can adsorb sodium alginate, absorbed by the membrane surface or into the membrane's pores. The presence of kaolin adsorption with sodium alginate was the main caused of the formation of the cake layer. It was more dominant on the surface of the membrane with a feed salt solution. This assumption was by [Zularisam et al. \(2011\)](#), which states that the organic material will form the structure of the fouling layer that serves as the "glue" for inorganic constituents.

For single sodium alginate solution feed on Fresh Water showed that sodium alginate was spread evenly on the surface of the membrane and is not mutually bonded to one another in the absence of the influence of ions affects the charge, respectively. The addition of salt causes more complex alginate molecules formation because it reduced the charge's effect in the alginate molecules. It also caused more entanglement of the polymer chains that were more complex and increased the alginate molecules' density. The interaction between the membrane and alginate molecules caused pore blocking or narrowing pore, but based on its main properties can also form a gel layer on the membrane surface. Sodium alginate gel structure was formed on the membrane's surface depending on the ionic environment (Ven et al., 2008).

Fouling behavior by kaolin and sodium alginate in different environments can be described in the figure below.

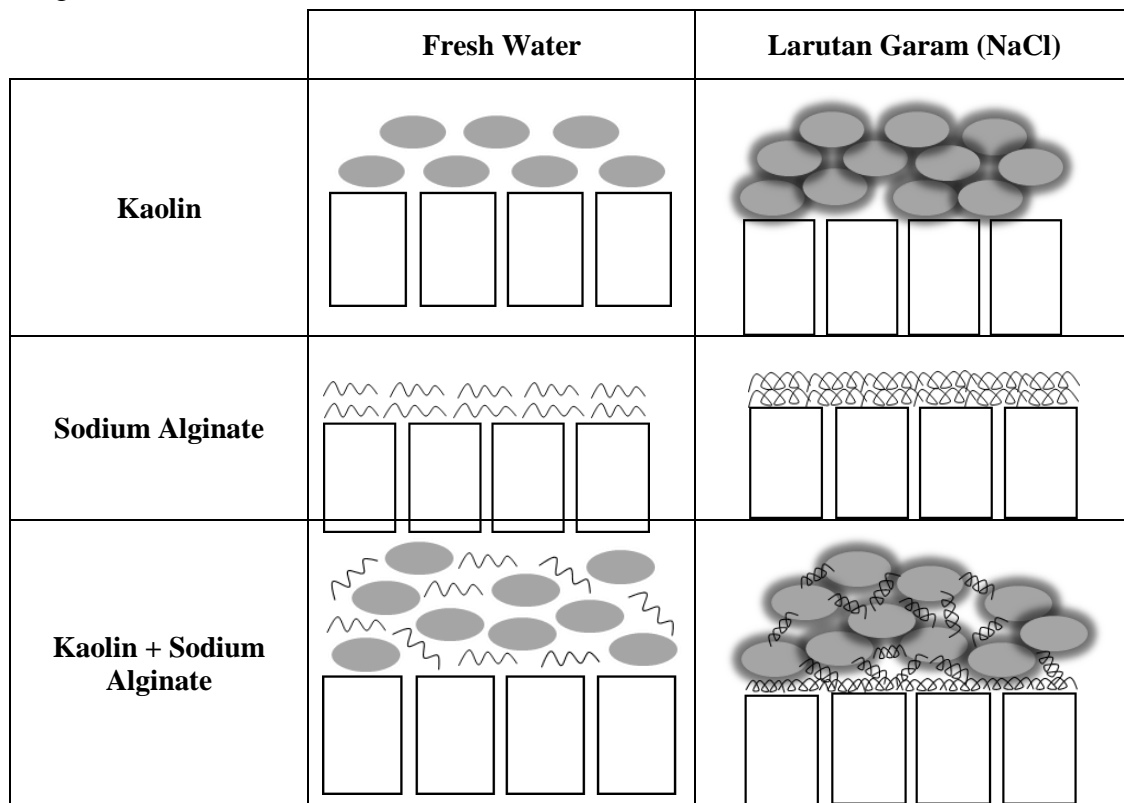


Figure 8 The Fouling phenomenon is illustrated by Sodium Alginate and Kaolin in different environments

4. Conclusions

Polysulfone ultrafiltration membrane removes total suspended solid and polysaccharide in seawater effectively. It is evidenced by the high resulting polysaccharide rejection in the synthetic seawater. PSf UF membranes successfully remove 93% of polysaccharides in a single feed, 95% in mixed feed, and 100% of suspended solids in a single feed and mixture. However, the fouling potential occurs more obviously in the mixed feed (TSS + polysaccharide), resulting in lower flux and higher rejection.

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