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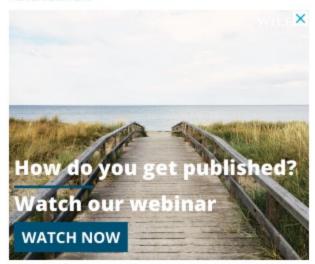
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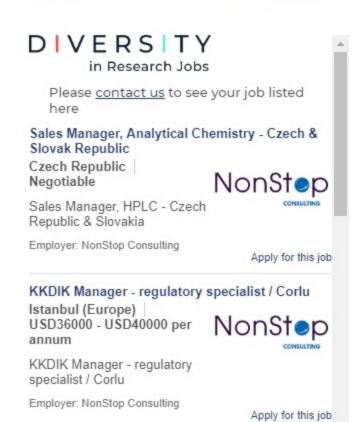
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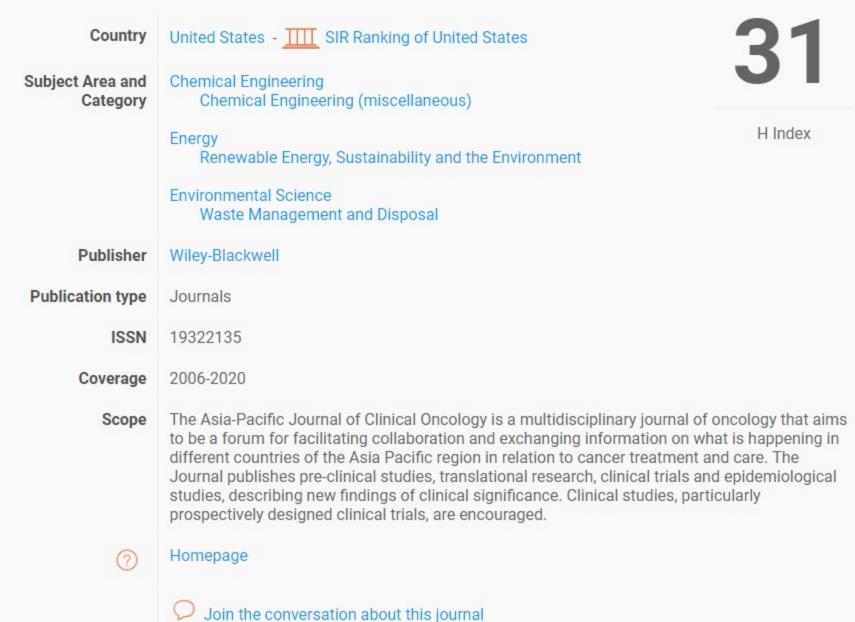
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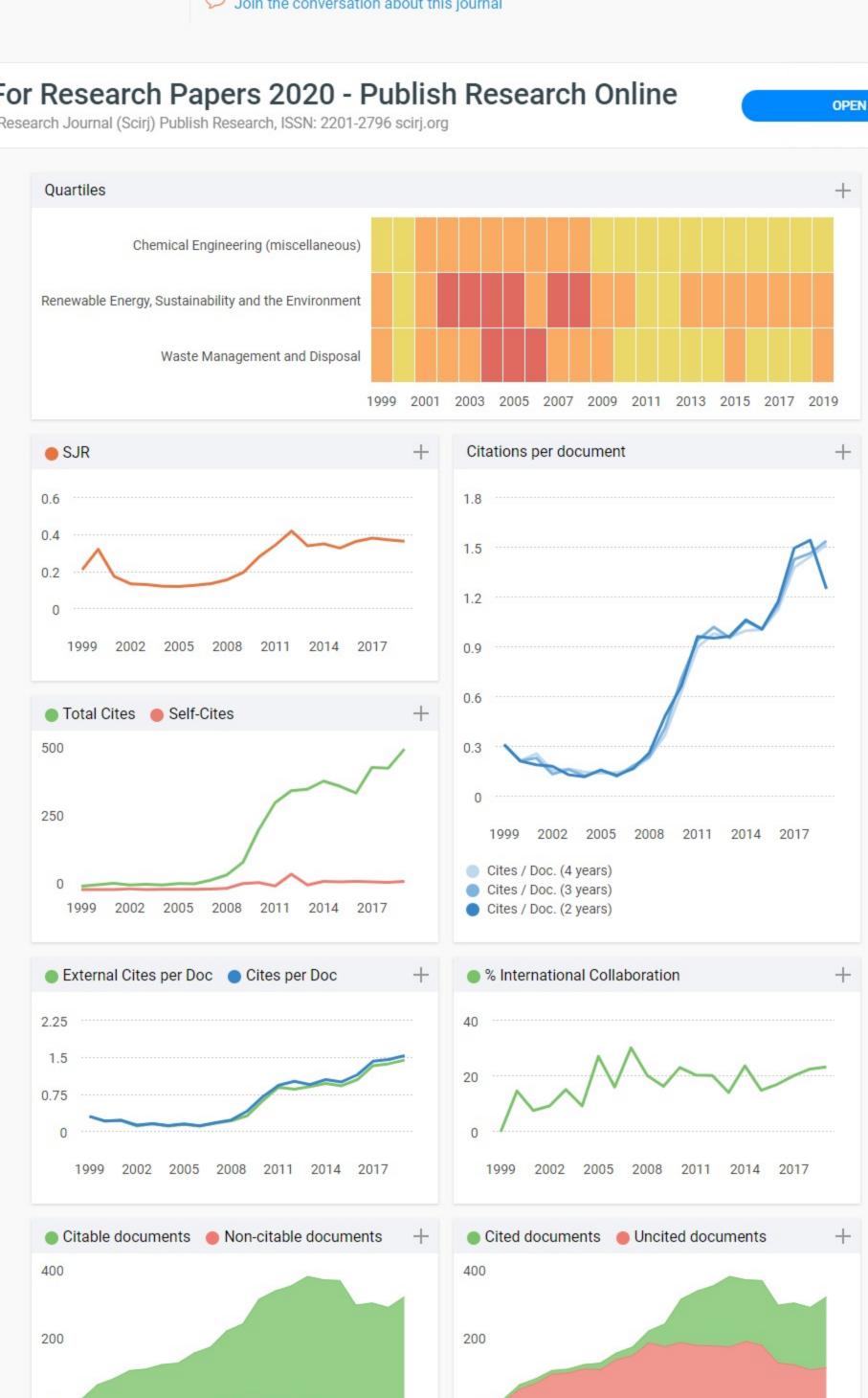
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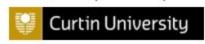
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RESEARCH ARTICLE

# Coagulation as pretreatment for membrane-based wetland saline water desalination

Muthia Elma X, Aulia Rahma, Amalia E. Pratiwi, Erdina L.A. Rampun

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#### Abstract

Wetland saline water is the enormous problem faced by rural people in Kalimantan, Indonesia. During rainy season, seawater intrudes wetland aquifer and turns water to saline. Application of membrane silica-pectin is eligible to solve this issue. Sadly, this water contains high concentration of organic matter that contributes to declining of membrane performance. Therefore, coagulation pretreatment is explored to enhance membrane filtration performance. The objective of this work is to investigate the optimum condition of coagulation pretreatment to enhance silica-pectin membrane performance for wetland saline water desalination. Sol gel process was employed to fabricate silica-pectin membrane and calcined via rapid thermal processing technique. Desalination was operated by pervaporation at 25-60°C. Maximum removal of organic matter UV<sub>254</sub> was obtained at coagulant 30 g L<sup>-1</sup> of 83.5% (pH 7). Combination processes between coagulation pretreatment and silica-pectin membranes offered high water flux at  $60^{\circ}$ C (12.2 kg m<sup>-2</sup> h<sup>-1</sup>). The UV<sub>254</sub> rejection of the feed water with coagulation is 43% higher than without applying coagulation pretreatment. Overall, salt rejection for all processes was extremely good (99.9%). Application of coagulation as pretreatment is promising to enhance performance of silica-pectin membrane for wetland water desalination.

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#### RESEARCH ARTICLE

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#### Coagulation as pretreatment for membrane-based wetland saline water desalination

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#### Abstract

Wetland saline water is the enormous problem faced by rural people in Kalimantan, Indonesia. During rainy season, seawater intrudes wetland aquifer and turns water to saline. Application of membrane silica-pectin is eligible to solve this issue. Sadly, this water contains high concentration of organic matter that contributes to declining of membrane performance. Therefore, coagulation pretreatment is explored to enhance membrane filtration performance. The objective of this work is to investigate the optimum condition of coagulation pretreatment to enhance silica-pectin membrane performance for wetland saline water desalination. Sol gel process was employed to fabricate silica-pectin membrane and calcined via rapid thermal processing technique. Desalination was operated by pervaporation at 25-60°C. Maximum removal of organic matter UV<sub>254</sub> was obtained at coagulant 30 g L<sup>-1</sup> of 83.5% (pH 7). Combination processes between coagulation pretreatment and silica-pectin membranes offered high water flux at 60°C (12.2 kg m<sup>-2</sup> h<sup>-1</sup>). The UV<sub>254</sub> rejection of the feed water with coagulation is 43% higher than without applying coagulation pretreatment. Overall, salt rejection for all processes was extremely good (99.9%). Application of coagulation as pretreatment is promising to enhance performance of silica-pectin membrane for wetland water desalination.

#### KEYWORDS

coagulation pretreatment process, silica-pectin membranes, wetland saline water

#### INTRODUCTION

Water shortage is a massive problem that the world is facing, especially for developing countries like Indonesia. Indonesia has considerable wetland area, which can be considered as water resources. It can be an alternative to resolve water shortage problems. However, wetland water has poor qualities and does not meet drinking water standard. Even more, during rainy season, sea water infiltrates in wetland aguifer and induces water to become saline. Generally, the wetland saline water is brown in color, with low pH, and consists of natural organic matter (NOM). NOM in the water limits the treatment option due to the reaction by products with chlorin are carcinogen.1

On other hand, wetland saline water consists of high salt concentration. Therefore, the desalination process via pervaporation of membrane technology is commonly applied to provide potable water demand. The pervaporation process is a promising desalination technology

to produce fresh water.<sup>2</sup> Generally, the pervaporation process employs inorganic membrane such as silica, which offers advantages, such as stability at high temperature, robustness, and high molecular sieving than zeolite and organic membranes.<sup>3</sup>

Fabrication of silica membrane is conducted by a simple method called sol gel process.<sup>4</sup> Siloxane (Si-O-Si) and silanol (Si-OH) groups were formed by hydrolysis and condensation reactions. Excesses of silanol groups may affect the hydrostability of silica membranes due to hydrophilic properties.<sup>5</sup> Hydrostability of silica membranes can be surmounted by templating carbon into silica matrices.<sup>6</sup> Carbon templating is an affordable method of improving silica matrix structures. Carbon from pectin is a polysaccharide complex compound consisting of multiple neutral sugar chains.<sup>7</sup> Pectin was employed as a natural carbon renewable source, which is costly and easy to obtain from fruit peels and industry waste.<sup>8</sup> Carbon from pectin can potentially be employed as a template agent for fabrication of organic silica membrane.

On the other hand, the performance of the membrane is still limited due to the contaminant clogged on the surface (fouling). Membrane fouling in wetland saline water occurs because of the presence of NOM. Coagulation is attractive to be used for removing NOM in water.9 The coagulation process was effectively a good way to increase NOM removal under an optimum condition.10 Aluminum sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>) is an inorganic coagulant used as long as applicable and affordable. 11 Zhao et al. 12 reported that the Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> coagulant is capable of removing UV<sub>254</sub> as an organic parameter (~ 91.3%). The hybrid process of coagulation and membrane was proven effective for reduction of NOM and thus enhances the quality of surface water13 and wetland water.14 Moreover, coagulation as pretreatment can improve the membrane's performance. 13 The objectives of this work are to investigate the optimum condition of the coagulation process and to evaluate the effect of coagulation toward silicapectin membrane performance for wetland saline water desalination.

#### 2 | MATERIALS AND METHODS

#### 2.1 | Chemical and material

Wetland saline water was taken at Muara Halayung village, South Kalimantan, Indonesia (3.2% NaCl). This water was used as feed water for desalination by the hybrid process of coagulation and silica-pectin membrane pervaporation. The material and chemical employed are aluminum sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, Merck), NaOH, HCl, tetraethyl ortho silicate (TEOS, 99%, Sigma-

Aldrich), apple peel pectin (0.5 wt%, Aldrich), ethanol (EtOH, 99%), glycerol (Aldrich), dilute nitric acid (0.0008 M HNO3, Merck), and ammonia (0.0003 M NH3, Merck) as acid-base catalysts. Macroporous alumina substrate was  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> tubular support (Ceramic Oxide Fabricators, Australia) with average pore size of 100 nm.

## 2.2 | Fabrication and characterization of silica-pectin membrane

Membrane fabrication was divided into three steps: (1) synthesis of silica sol by the sol gel method. The detail of the procedure was described in a previous work.15 (2) Pectin templating to silica sol. In this step, pectin was employed and mixed with silica sol for 45 min at 0°C. Before doing so, pectin was diluted with glycerol for 45 min at 40°C. The final total molar ratio of silica-pectin sol was 1:38:0.0008:5:0.0003:0.00179 (TEOS:EtOH:HNO<sub>3</sub>: H2O:NH3:Pectin). (3) Silica-pectin membrane dip coating. Fabrication of silica–pectin membranes was prepared by dip coating of membrane support (macroporous alumina) into silica-pectin sol. Every layer was calcined in a furnace at 400°C via rapid thermal processing (RTP) method. The silica-pectin membranes' morphology and thickness were examined using scanning electron microscopy (Zeiss EVO LS15).

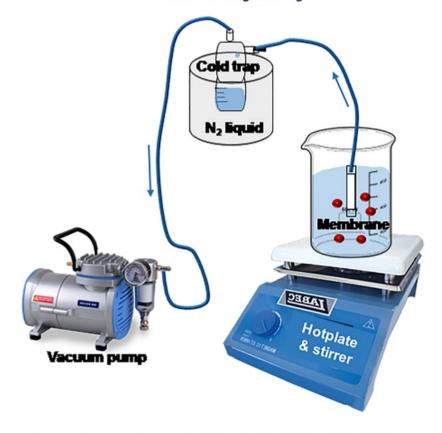
#### 2.3 | Coagulation pretreatment

Coagulation experiments were carried out on wetland saline water samples with pH of 4–8 and  $Al_2(SO_4)_3$  doses of 10–60 mg  $L^{-1}$ . The jar test experiment was performed using a flocculator (Mascotte F-6, Indonesia). Rapid mixing was carried out for 1 min at a speed of 200 rpm followed by slow mixing at 40 rpm for 15 min and then sedimentation for 20 min. The supernatant of the coagulated water was measured by conductivity and the organic content of  $UV_{254}$  by using a conductivity meter (OHAUS Starter 300C) and UV-1600 Spectrophotometer, respectively.

#### 2.4 | Pervaporation experiment

The silica-pectin membrane was installed in a dead-end system immersed in a beaker glass with multiple feeds, that is, (1) wetland saline water without coagulation as pretreatment and (2) water sample after coagulation pretreatment at optimum condition. The schematic of the pervaporation setup is displayed on Figure 1. All experiments were tested at multiple temperatures—25°C, 40°C, and 60°C. The membrane is connected with a vacuum

FIGURE 1 Schematic of pervaporation processes for desalination of wetland saline water

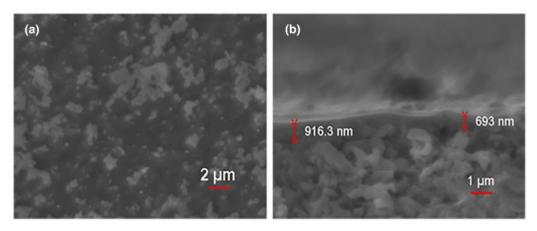


hose, and the permeate flow was collected in a cold trap soaked in a container containing liquid nitrogen. To prevent the polarization of concentration, the feed reactor was connected to a peristaltic pump where the retentate flow was constantly recycled and stirred (Elma et al., 2013) $^{16}$ . Furthermore, the permeate was monitored by the concentration of Total Dissolved Solid (TDS) and conductivity using a conductivity meter and UV<sub>254</sub> with UV–vis, and the flux was also calculated.

#### 3 | RESULTS AND DISCUSSIONS

## 3.1 | Morphology of silica-pectin membrane

The morphology structure of silica-pectin membrane was determined by scanning electron microscopy (SEM) image shown in Figure 2. As shown in Figure 2, the membrane structure is asymmetric. It is caused by



 $\textbf{FIGURE 2} \quad \text{Scanning electron microscopy images of silica-pectin membrane (a) surface with magnification 2000× and (b) cross section with magnification 5000× \\$ 

differences in pore size between silica-pectin top layer and membrane support.<sup>4</sup> This pore difference makes the surface layer of the membrane rough (Figure 2a). After calcination at 400°C, the membrane becomes more compact and solid.

The thickness of silica-pectin was determined by the technique used and the temperature of calcination. Figure 2b shows the thickness of the silica-pectin layer ranging from 693 to 916.3 nm at a calcination temperature of 400°C. The *membranes used in this study* tend to be thicker than the membranes reported by Elma *and* Riskawati<sup>15</sup> due to the different membranes used, namely, pure silica with calcination temperature of 600°C. Generally, silica membranes are calcined at high temperatures up to 600°C. However, the presence of carbon pectin as *a* template *agent* may lead organic *carbon* to easily decompose at high temperatures.

The calcination technique is one of the factors necessary to determine membrane thickness. Silica membrane calcined via conventional thermal processing (CTP) technique had a thickness of 30–50 nm, which is thinner than the one produced from the RTP technique. However, the RTP technique is superior because it can save costs and time for membrane fabrication. Fabrication of silica membranes via CTP generally takes 24 h per layer, whereas making silica–pectin membranes with the RTP technique in this work only takes less than 1 day for four layers.

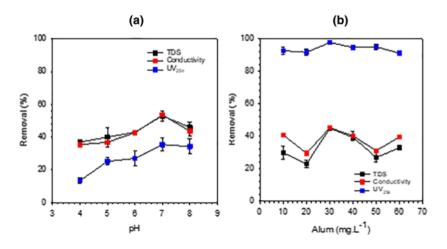
#### 3.2 | Performance of coagulationmembrane processes

Figure 3 shows the effect of pH and alum dose on TDS, conductivity, and  $UV_{254}$  removal. Increasing pH also leads to removal of all parameter ( $UV_{254}$ , TDS, and conductivity). The highest  $UV_{254}$  removal occurs at pH

7 because the pH of the solution greatly affects the solubility of Al (OH)<sub>3</sub> precipitates, which tends to be low. Afterwards, metal ions dissolved in water may accelerate the process of destabilization of colloidal particles to form floc.<sup>17</sup>

The highest percentage of TDS, conductivity, and UV<sub>254</sub> removal was obtained at pH 7 (Figure 3a). These results were lower (5.8%) compared with a previous work by Zhao et al.9 at the same optimum conditions. It is caused by the difference in the characteristics of water sample used in this work. The initial concentrations of TDS, pH, and  $UV_{254}$  in wetland saline water are 75.8 g  $L^{-1}$ , 5.8, and 0.460 cm<sup>-1</sup>, respectively. In accordance with the literature, pH 7 is the optimum pH for removing UV<sub>254</sub> for wetland saline water. The mechanism of decreasing conductivity and TDS might be due to the presence of coagulants in certain doses, which caused several of mineral ions and organic compounds to be dispersed and clump to form floc in the flocculation process, which then settled and separated from the solution. The coagulation process was not good enough to remove conductivity and TDS, so the membrane attempted to remove it after pretreatment.

Addition of coagulant into wetland saline water resulted in fluctuation of TDS, conductivity, and  $UV_{254}$  removal (Figure 3b). All parameters increase under alum doses of  $10\text{--}30~\text{mg L}^{-1}$ . However, doses of  $>30~\text{mg L}^{-1}$  decrease removal efficiency due to load restabilization. <sup>18</sup> The highest percentage of  $UV_{254}$  removal occurs at a dose of 30 mg  $U^{-1}$  ( $\sim$ 97.7%). The performance of coagulation coupled with silica–pectin membrane pervaporation of wetland saline water was carried out using two varied feed, that is, without coagulation and after coagulation pretreatment by optimum dose 30 mg  $U^{-1}$  and pH 7. Figure 4 shows water flux sorted from highest to smallest using feed without and after coagulation at multiple feed temperature. These results are influenced by the presence



**FIGURE 3** Effect of (a) pH by alum loading 30 mg  $\rm L^{-1}$  and (b) alum dose to efficiently remove TDS, conductivity, and UV<sub>254</sub> on coagulation pretreatment

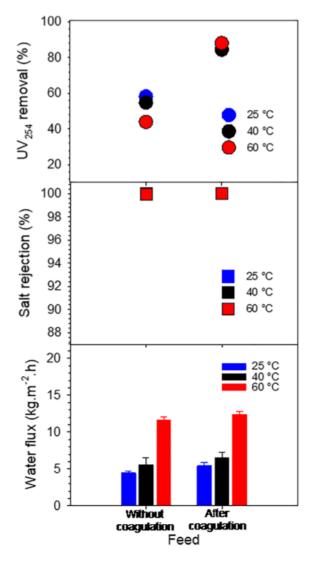


FIGURE 4 Performance of silica-pectin membrane for wetland saline water desalination with varied feed and temperature

of salt and NOM in feed. Wetland saline water without pretreatment has high NOM, which leads to membrane fouling. Fouling plays a role in blocking membrane pore and reduce the membrane performance, so the water flux decreases.19 The highest water flux obtained was after coagulation of the feed of 12.39 kg m<sup>-2</sup> h<sup>-1</sup> (temperature 60°C), whereas the lowest permeate flux was produced from wetland saline water as feed without pretreatment at room temperature ~25°C. Increasing of feed temperature increases water flux as well. These results are in accordance with the literature stating that heating of feed at a certain temperature causes an increase in diffusivity and can reduce the viscosity of the solution.20

Figure 4 shows that the water flux of wetland saline water without coagulation tends to be lower (from 11% to 5.7%) when compared with feed water with coagulation pretreatment on all feed temperatures. These results occur because of differences in the concentration of contaminants in each feed. This was also mentioned in the work of Elma et al.21 which used variations of NaCl concentration (0.3%-7.5% NaCl) as the feed. The high concentration of contaminants in membrane feed water results in concentration polarization, which has a negative effect on reducing membrane performance.4

Coagulation as pretreatment is able to increase membrane water flux in treating water with high NOM content.22 The coagulation process plays a role in reducing NOM in water to ease the work of membranes.23 Interestingly, the results of this study show that water flux of wetland saline water of the feed after coagulation pretreatment at 25°C is almost equivalent to water flux of feed without coagulation at 40°C. We conclude that the coagulation pretreatment greatly improves the performance of silica-pectin membranes for desalination of wetland saline water. Silica-derived membranes are hydrophilic materials, and the water transport can be described by a sorption-diffusion mechanism.4 High hydrophilicity is critical for desalination membranes to minimize membrane fouling by NOM.24 Hence, silicapectin membrane, which has hydrophilic properties, was good for desalinating wetland saline water with or without coagulation pretreatment. Unfortunately, NOM (UV<sub>254</sub>) rejection is still bad for pervaporation of wetland saline water without coagulation as exhibited on Figure 4.

Fascinatingly, UV254 rejection performed high efficiencies of 88.8% (25°C) by coupled coagulation as pretreatment and without pretreatment was only 58.2% (25°C), 55% (40°C), and 43.9% (60°C) (Figure 4). The small amount of UV254 rejection for desalination of these feeds is thought to be due to membrane fouling. The presence of high NOM in water contributed to organic fouling on the membrane.19 We conclude that pretreatment of coagulation acts to ease the work of membranes for removing NOM effectively.

Interestingly, the rejection of salt for all feed water was extremely good at all temperature (25–60°C). Based on Figure 4, salt rejections in all feed are high (>98%). The salt rejection results are inversely proportional to the low UV254 rejection. The rejection of saline salt water without pretreatment is very good even though the silica-pectin membrane has fouling due to the presence of a layer of humic compounds in desalination of wetland saline water without pretreatment. According to Kim et al.25 organic fouling can increase or decrease salt rejection depending on the characteristics of the membrane used.

#### 4 | CONCLUSION

This work shows that wetland saline water had poor characteristics and does not meet drinking water quality standard according to Indonesia health minister regulation No 492/2010 and EPA standard. Coagulation pretreatment was very effective to remove 97.7% of UV<sub>254</sub> at optimum condition (pH 7) and alum dose of 30 mg L<sup>-1</sup>. A hybrid of coagulation process and silica-pectin membranes for wetland saline water desalination is very effective to reduce 43 % UV<sub>254</sub> with the highest water flux of 12.2 kg.m<sup>-2</sup> h<sup>-1</sup> and excellent 99.9% salt rejection (at 60°C feed temperature). In addition, silica-pectin membranes are promising for wetland saline water desalination, considering that templating pectin as a carbon precursor offers relatively high membrane hydrostability to improve and enhance the performance of silica membranes.

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#### RESEARCH ARTICLE



## Asia-Pacific Journal of Chemical Engineering WILEY

# Coagulation as pretreatment for membrane-based wetland saline water desalination

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#### **Abstract**

Wetland saline water is the enormous problem faced by rural people in Kalimantan, Indonesia. During rainy season, seawater intrudes wetland aquifer and turns water to saline. Application of membrane silica-pectin is eligible to solve this issue. Sadly, this water contains high concentration of organic matter that contributes to declining of membrane performance. Therefore, coagulation pretreatment is explored to enhance membrane filtration performance. The objective of this work is to investigate the optimum condition of coagulation pretreatment to enhance silica-pectin membrane performance for wetland saline water desalination. Sol gel process was employed to fabricate silica-pectin membrane and calcined via rapid thermal processing technique. Desalination was operated by pervaporation at 25-60°C. Maximum removal of organic matter  $UV_{254}$  was obtained at coagulant 30 g  $L^{-1}$  of 83.5% (pH 7). Combination processes between coagulation pretreatment and silica-pectin membranes offered high water flux at 60°C (12.2 kg m<sup>-2</sup> h<sup>-1</sup>). The UV<sub>254</sub> rejection of the feed water with coagulation is 43% higher than without applying coagulation pretreatment. Overall, salt rejection for all processes was extremely good (99.9%). Application of coagulation as pretreatment is promising to enhance performance of silica-pectin membrane for wetland water desalination.

#### KEYWORDS

coagulation pretreatment process, silica-pectin membranes, wetland saline water

#### 1 | INTRODUCTION

Water shortage is a massive problem that the world is facing, especially for developing countries like Indonesia. Indonesia has considerable wetland area, which can be considered as water resources. It can be an alternative to resolve water shortage problems. However, wetland water has poor qualities and does not meet drinking water standard. Even more, during rainy season, sea water infiltrates in wetland aquifer and induces water to

become saline. Generally, the wetland saline water is brown in color, with low pH, and consists of natural organic matter (NOM). NOM in the water limits the treatment option due to the reaction by products with chlorin are carcinogen.<sup>1</sup>

On other hand, wetland saline water consists of high salt concentration. Therefore, the desalination process via pervaporation of membrane technology is commonly applied to provide potable water demand. The pervaporation process is a promising desalination technology to produce fresh water.<sup>2</sup> Generally, the pervaporation process employs inorganic membrane such as silica, which offers advantages, such as stability at high temperature, robustness, and high molecular sieving than zeolite and organic membranes.<sup>3</sup>

Fabrication of silica membrane is conducted by a simple method called sol gel process.<sup>4</sup> Siloxane (Si-O-Si) and silanol (Si-OH) groups were formed by hydrolysis and condensation reactions. Excesses of silanol groups may affect the hydrostability of silica membranes due to hydrophilic properties.<sup>5</sup> Hydrostability of silica membranes can be surmounted by templating carbon into silica matrices.<sup>6</sup> Carbon templating is an affordable method of improving silica matrix structures. Carbon from pectin is a polysaccharide complex compound consisting of multiple neutral sugar chains.<sup>7</sup> Pectin was employed as a natural carbon renewable source, which is costly and easy to obtain from fruit peels and industry waste.<sup>8</sup> Carbon from pectin can potentially be employed as a template agent for fabrication of organic silica membrane.

On the other hand, the performance of the membrane is still limited due to the contaminant clogged on the surface (fouling). Membrane fouling in wetland saline water occurs because of the presence of NOM. Coagulation is attractive to be used for removing NOM in water.9 The coagulation process was effectively a good way to increase NOM removal under an optimum condition.<sup>10</sup> Aluminum sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>) is an inorganic coagulant used as long as applicable and affordable. 11 Zhao et al. 12 reported that the Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> coagulant is capable of removing UV<sub>254</sub> as an organic parameter (~ 91.3%). The hybrid process of coagulation and membrane was proven effective for reduction of NOM and thus enhances the quality of surface water<sup>13</sup> and wetland water.<sup>14</sup> Moreover, coagulation as pretreatment can improve the membrane's performance.<sup>13</sup> The objectives of this work are to investigate the optimum condition of the coagulation process and to evaluate the effect of coagulation toward silicapectin membrane performance for wetland saline water desalination.

#### 2 | MATERIALS AND METHODS

#### 2.1 | Chemical and material

Wetland saline water was taken at Muara Halayung village, South Kalimantan, Indonesia (3.2% NaCl). This water was used as feed water for desalination by the hybrid process of coagulation and silica-pectin membrane pervaporation. The material and chemical employed are aluminum sulfate (Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub>, Merck), NaOH, HCl, tetraethyl ortho silicate (TEOS, 99%, Sigma-

Aldrich), apple peel pectin (0.5 wt%, Aldrich), ethanol (EtOH, 99%), glycerol (Aldrich), dilute nitric acid (0.0008 M HNO3, Merck), and ammonia (0.0003 M NH3, Merck) as acid-base catalysts. Macroporous alumina substrate was  $\alpha$ -Al $_2$ O $_3$  tubular support (Ceramic Oxide Fabricators, Australia) with average pore size of 100 nm.

# 2.2 | Fabrication and characterization of silica-pectin membrane

Membrane fabrication was divided into three steps: (1) synthesis of silica sol by the sol gel method. The detail of the procedure was described in a previous work.<sup>15</sup> (2) Pectin templating to silica sol. In this step, pectin was employed and mixed with silica sol for 45 min at 0°C. Before doing so, pectin was diluted with glycerol for 45 min at 40°C. The final total molar ratio of silica-pectin sol was 1:38:0.0008:5:0.0003:0.00179 (TEOS:EtOH:HNO3: H<sub>2</sub>O:NH<sub>3</sub>:Pectin). (3) Silica-pectin membrane dip coating. Fabrication of silica-pectin membranes was prepared by dip coating of membrane support (macroporous alumina) into silica-pectin sol. Every layer was calcined in a furnace at 400°C via rapid thermal processing (RTP) method. The silica-pectin membranes' morphology and thickness were examined using scanning electron microscopy (Zeiss EVO LS15).

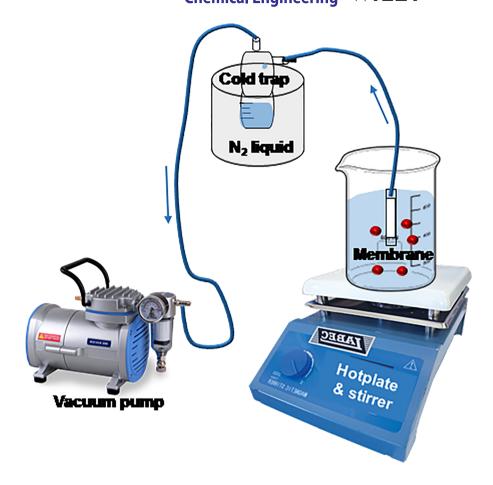
#### 2.3 | Coagulation pretreatment

Coagulation experiments were carried out on wetland saline water samples with pH of 4–8 and  $Al_2(SO_4)_3$  doses of 10–60 mg  $L^{-1}$ . The jar test experiment was performed using a flocculator (Mascotte F-6, Indonesia). Rapid mixing was carried out for 1 min at a speed of 200 rpm followed by slow mixing at 40 rpm for 15 min and then sedimentation for 20 min. The supernatant of the coagulated water was measured by conductivity and the organic content of  $UV_{254}$  by using a conductivity meter (OHAUS Starter 300C) and UV-1600 Spectrophotometer, respectively.

#### 2.4 | Pervaporation experiment

The silica–pectin membrane was installed in a dead-end system immersed in a beaker glass with multiple feeds, that is, (1) wetland saline water without coagulation as pretreatment and (2) water sample after coagulation pretreatment at optimum condition. The schematic of the pervaporation setup is displayed on Figure 1. All experiments were tested at multiple temperatures—25°C, 40°C, and 60°C. The membrane is connected with a vacuum

**FIGURE 1** Schematic of pervaporation processes for desalination of wetland saline water



hose, and the permeate flow was collected in a cold trap soaked in a container containing liquid nitrogen. To prevent the polarization of concentration, the feed reactor was connected to a peristaltic pump where the retentate flow was constantly recycled and stirred (Elma et al., 2013)<sup>16</sup>. Furthermore, the permeate was monitored by the concentration of Total Dissolved Solid (TDS) and conductivity using a conductivity meter and UV<sub>254</sub> with UV–vis, and the flux was also calculated.

#### 3 | RESULTS AND DISCUSSIONS

## 3.1 | Morphology of silica-pectin membrane

The morphology structure of silica-pectin membrane was determined by scanning electron microscopy (SEM) image shown in Figure 2. As shown in Figure 2, the membrane structure is asymmetric. It is caused by

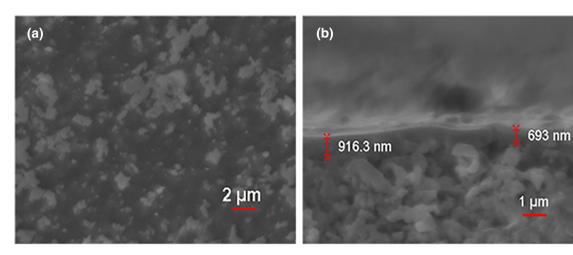


FIGURE 2 Scanning electron microscopy images of silica–pectin membrane (a) surface with magnification  $2000\times$  and (b) cross section with magnification  $5000\times$ 

differences in pore size between silica–pectin top layer and membrane support.<sup>4</sup> This pore difference makes the surface layer of the membrane rough (Figure 2a). After calcination at 400°C, the membrane becomes more compact and solid.

The thickness of silica-pectin was determined by the technique used and the temperature of calcination. Figure 2b shows the thickness of the silica-pectin layer ranging from 693 to 916.3 nm at a calcination temperature of 400°C. The *membranes used in this study* tend to be thicker than the membranes reported by Elma *and* Riskawati<sup>15</sup> due to the different membranes used, namely, pure silica with calcination temperature of 600°C. Generally, silica membranes are calcined at high temperatures up to 600°C. However, the presence of carbon pectin as *a* template *agent* may lead organic *carbon* to easily decompose at high temperatures.

The calcination technique is one of the factors necessary to determine membrane thickness. Silica membrane calcined via conventional thermal processing (CTP) technique had a thickness of 30–50 nm, which is thinner than the one produced from the RTP technique. However, the RTP technique is superior because it can save costs and time for membrane fabrication. Fabrication of silica membranes via CTP generally takes 24 h per layer, whereas making silica–pectin membranes with the RTP technique in this work only takes less than 1 day for four layers.

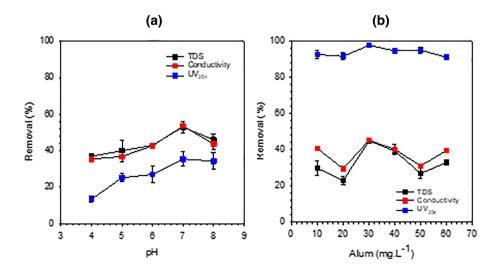
#### 3.2 | Performance of coagulationmembrane processes

Figure 3 shows the effect of pH and alum dose on TDS, conductivity, and  $UV_{254}$  removal. Increasing pH also leads to removal of all parameter ( $UV_{254}$ , TDS, and conductivity). The highest  $UV_{254}$  removal occurs at pH

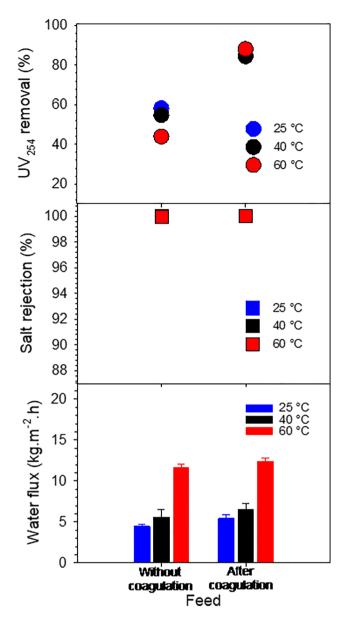
7 because the pH of the solution greatly affects the solubility of Al (OH)<sub>3</sub> precipitates, which tends to be low. Afterwards, metal ions dissolved in water may accelerate the process of destabilization of colloidal particles to form floc.<sup>17</sup>

The highest percentage of TDS, conductivity, and UV<sub>254</sub> removal was obtained at pH 7 (Figure 3a). These results were lower (5.8%) compared with a previous work by Zhao et al.<sup>9</sup> at the same optimum conditions. It is caused by the difference in the characteristics of water sample used in this work. The initial concentrations of TDS, pH, and  $UV_{254}$  in wetland saline water are 75.8 g  $L^{-1}$ , 5.8, and 0.460 cm<sup>-1</sup>, respectively. In accordance with the literature, pH 7 is the optimum pH for removing UV<sub>254</sub> for wetland saline water. The mechanism of decreasing conductivity and TDS might be due to the presence of coagulants in certain doses, which caused several of mineral ions and organic compounds to be dispersed and clump to form floc in the flocculation process, which then settled and separated from the solution. The coagulation process was not good enough to remove conductivity and TDS, so the membrane attempted to remove it after pretreatment.

Addition of coagulant into wetland saline water resulted in fluctuation of TDS, conductivity, and UV<sub>254</sub> removal (Figure 3b). All parameters increase under alum doses of 10–30 mg L<sup>-1</sup>. However, doses of >30 mg L<sup>-1</sup> decrease removal efficiency due to load restabilization. <sup>18</sup> The highest percentage of UV<sub>254</sub> removal occurs at a dose of 30 mg L<sup>-1</sup> (~97.7%). The performance of coagulation coupled with silica–pectin membrane pervaporation of wetland saline water was carried out using two varied feed, that is, without coagulation and after coagulation pretreatment by optimum dose 30 mg L<sup>-1</sup> and pH 7. Figure 4 shows water flux sorted from highest to smallest using feed without and after coagulation at multiple feed temperature. These results are influenced by the presence



**FIGURE 3** Effect of (a) pH by alum loading 30 mg  $L^{-1}$  and (b) alum dose to efficiently remove TDS, conductivity, and  $UV_{254}$  on coagulation pretreatment



**FIGURE 4** Performance of silica–pectin membrane for wetland saline water desalination with varied feed and temperature

of salt and NOM in feed. Wetland saline water without pretreatment has high NOM, which leads to membrane fouling. Fouling plays a role in blocking membrane pore and reduce the membrane performance, so the water flux decreases. The highest water flux obtained was after coagulation of the feed of 12.39 kg m<sup>-2</sup> h<sup>-1</sup> (temperature 60°C), whereas the lowest permeate flux was produced from wetland saline water as feed without pretreatment at room temperature ~25°C. Increasing of feed temperature increases water flux as well. These results are in accordance with the literature stating that heating of feed at a certain temperature causes an increase in diffusivity and can reduce the viscosity of the solution.

Figure 4 shows that the water flux of wetland saline water without coagulation tends to be lower (from 11% to 5.7%) when compared with feed water with coagulation pretreatment on all feed temperatures. These results occur because of differences in the concentration of contaminants in each feed. This was also mentioned in the work of Elma et al.<sup>21</sup> which used variations of NaCl concentration (0.3%–7.5% NaCl) as the feed. The high concentration of contaminants in membrane feed water results in concentration polarization, which has a negative effect on reducing membrane performance.<sup>4</sup>

Coagulation as pretreatment is able to increase membrane water flux in treating water with high NOM content.<sup>22</sup> The coagulation process plays a role in reducing NOM in water to ease the work of membranes.<sup>23</sup> Interestingly, the results of this study show that water flux of wetland saline water of the feed after coagulation pretreatment at 25°C is almost equivalent to water flux of feed without coagulation at 40°C. We conclude that the coagulation pretreatment greatly improves the performance of silica-pectin membranes for desalination of wetland saline water. Silica-derived membranes are hydrophilic materials, and the water transport can be described by a sorption-diffusion mechanism.4 High hydrophilicity is critical for desalination membranes to minimize membrane fouling by NOM.24 Hence, silicapectin membrane, which has hydrophilic properties, was good for desalinating wetland saline water with or without coagulation pretreatment. Unfortunately, NOM (UV<sub>254</sub>) rejection is still bad for pervaporation of wetland saline water without coagulation as exhibited on

Fascinatingly,  $UV_{254}$  rejection performed high efficiencies of 88.8% (25°C) by coupled coagulation as pretreatment and without pretreatment was only 58.2% (25°C), 55% (40°C), and 43.9% (60°C) (Figure 4). The small amount of  $UV_{254}$  rejection for desalination of these feeds is thought to be due to membrane fouling. The presence of high NOM in water contributed to organic fouling on the membrane.<sup>19</sup> We conclude that pretreatment of coagulation acts to ease the work of membranes for removing NOM effectively.

Interestingly, the rejection of salt for all feed water was extremely good at all temperature (25–60°C). Based on Figure 4, salt rejections in all feed are high (>98%). The salt rejection results are inversely proportional to the low  $UV_{254}$  rejection. The rejection of saline salt water without pretreatment is very good even though the silica–pectin membrane has fouling due to the presence of a layer of humic compounds in desalination of wetland saline water without pretreatment. According to Kim et al.<sup>25</sup> organic fouling can increase or decrease salt

rejection depending on the characteristics of the membrane used.

#### 4 | CONCLUSION

This work shows that wetland saline water had poor characteristics and does not meet drinking water quality standard according to Indonesia health minister regulation No 492/2010 and EPA standard. Coagulation pretreatment was very effective to remove 97.7% of UV<sub>254</sub> at optimum condition (pH 7) and alum dose of 30 mg  $L^{-1}$ . A hybrid of coagulation process and silica-pectin membranes for wetland saline water desalination is very effective to reduce 43 % UV<sub>254</sub> with the highest water flux of 12.2 kg.m<sup>-2</sup> h<sup>-1</sup> and excellent 99.9% salt rejection (at 60°C feed temperature). In addition, silica-pectin membranes are promising for wetland saline water desalination, considering that templating pectin as a carbon precursor offers relatively high membrane hydrostability to improve and enhance the performance of silica membranes.

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