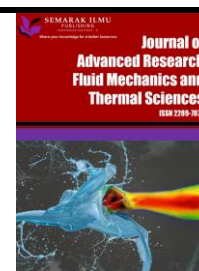




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Banana Peels Pectin Templated Silica Ultrafiltration Membrane in Disk Plate Configuration Applied for Wetland Water Treatment

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ABSTRACT

South Kalimantan has natural surface water such as wetland water which easily found to prevent water scarcity. Two types of wetland water provided in tropical area i.e. (1) peat water which located in land area; and (2) wetland saline water that area is near to coastal zone. Both are unsuitable for consumption and required to be treated. Ultrafiltration (UF) is one of methods that can be applied for treating natural wetland water using silica membrane. However, it needs improved by carbon due to lacking its low hydrostability properties. This works investigated performance of banana pectin templated silica ultrafiltration membrane in varied operation pressure and feed water treatment. Preparation of ultrafiltration membrane was used banana peels pectin as carbon source that templated into silica sol through sol gel process. Silica pectin xerogel was characterized by Fourier Transformed Infra-red (FTIR) and morphology of silica pectin membrane was characterized by Scanning Electron Microscopy (SEM). The sol is carried out by a coating process to the alumina membrane support on 4 layers. The calcination process is using the Conventional Thermal Processing (CTP) method. Furthermore, the ultrafiltration process run on the dead-end ultrafiltration silica pectin membrane has three functional groups, namely siloxane (Si-O-Si) at a wavelength of 1061 cm^{-1} , silanol (Si-OH) at a wavelength of 966 cm^{-1} and silica carbon (Si-C) at a wavelength of 795 cm^{-1} . The performance of silica pectin membranes at pressures of 0.5 bar, 1 bar and 1.5 bar in peat water resulted in fluxes of 5.07 $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, 11.31 $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and 18.90 $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ with rejection of organic matter of 85.56%, 85.49% and 84.68%. Meanwhile in wetland saline water it was 7.04 $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$, 11.73 $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ and 20.52 $\text{kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ with rejection of organic matter of 82.50%, 79.46% and 78.11%. Salt rejection for wetland saline water for 0.5 bar until 1.5 bar is 84.39%, 82.54% and 81.04%. Ultrafiltration It can be concluded that the increase in pressure affects the performance of the silica pectin membrane where the flux value increases with each increase in pressure, but the rejection value of organic matter and salt rejection show a decreasing.

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1. Introduction

Water scarcity is a serious global issue that is getting critical by increasing population of people, industry, and agricultural which bring out the freshwater source could be disappearing [1]. Water is known as essential for the survival of mankind [2]. 2.7 billion people are suffering from water scarcity problems and still growing up [3]. The World Health Organization (WHO) also said there is more than 15% of the world's population does not have access to drinking water [4]. Drinking water problems has been encountered many regions in Indonesia due to different geographical and demographic characteristic [5].

South Kalimantan is located in the souths part of Kalimantan island-Indonesia that considerable found of wetland with height of soil at 0.16 m under sea level [6]. It is important in hydrology and big potential as water reservoir [7]. South Kalimantan has an abundant source of water especially wetland water [8]. However, Wetland water has poor qualities and not suitable for any consumption. Generally, the wetland water has brown in colour, low pH, and contain of natural organic matter (NOM) [9]. So, this wetland water is big source of clean water requires for contaminant removal process [7].

Membrane technology has long been a separation process in the industrial world because of its fast and efficient process [10-28]. In general, there are three types of membrane process that are usually applied, it is called reverse osmosis (RO), membrane distillation (MD), pervaporation and ultrafiltration [28]. Among various water treatment technologies, ultrafiltration (UF) is an alternative technology to improve water quality [26]. UF is a promising and the most widely applied technology for producing water consumption from surface water with a lower cost [29]. Besides, membranes UF has ease of fabrication and simple operation system [30]. Compared to reverse osmosis (RO) membrane, a UF membrane is operated by using lower operating pressure to retain solutes. The range of 1-10 bars is typical operating pressure for UF [31].

Membrane fabrication frequently applied mesopores silica in water separation. Silica has good mechanical strength and thermal stability [1]. In order to increase hydro stability and silica matrices endurance for mechanical and chemical strength, it is required a modification process on pure silica matrix by adding and templating carbon silica [26,32-42]. In addition, embedding carbon molecules into silica matrix lead well hydro stability of silica [43]. This carbon source may employ pectin [36]. Pectin can be extracted from banana peels (around 24,8 wt.%) [23,44].

As a great potential, pectin as carbon templated has promised excellent membrane performance. Previous work has been demonstrated silica pectin membrane has the great performance than silica P-123 membrane by pervaporation [45]. Elma *et al.*, [44] also reported 0.5 wt.% pectin concentration has better performance than 0.1 wt.% concentration when applied for desalination of wetland saline water and acid mine drainage. Therefore, this work investigates banana peels templated silica pectin performance on wetland water by ultrafiltration.

2. Methodology

2.1 Chemical and Materials

The natural feed water is implemented for the UF experiment in this work included peatland water and wetland saline water. Peatland water is taken from Sukamaju Village at Banjarbaru (3°23'57.6"S 114°43'09.0"E), whereas, the wetland saline water is taken from Muara Halayung Village at Banjar Region, South Kalimantan-Indonesia (3°29'57.7"S 114°34'20.9"E). Membrane support derived from α -alumina disc membrane with pore size 100 μm (Ceramics Oxide Fabricators, Australia). Silica pectin sols is generally fabricated from Tetraethyl Orthosilicate (TEOS, 99%, Sigma-

Aldrich), ethanol (96%), ammonia (0.00003N, Merck), nitric acid (0.0008N, Merck), pectin from banana peels (0.5%), and glycerol (85%, Merck).

2.2 Membrane Fabrication and Characterization

Silica pectin sols is synthesized by following our previous work [18,19]. TEOS mixed with Ethanol for 5 minutes at 0 °C. Then, HNO₃ is added into solution for an hour at 50 °C. Afterwards, NH₃ is added by dropwise under stirring for 2 hours at the same condition. Next, check the solution pH of pure silica sols become 6. The pectin is dissolved into 5 ml glycerol and then stir well for 95 minutes at 50 °C. Finally, 0.5 wt.% banana peels pectin template are added into sols and stir it still for 45 minutes at 0 °C. Banana peels pectin template silica sol gel was dried for 24 h at 60 °C and then grounded as xerogels for characterization. The homogenous xerogel powder are calcinated for 2 h at 300 °C using Conventional thermal processing (CTP) technique. Then, the Fourier Transform Infra-Red (FTIR) spectra is carried out to measure the functional groups in silica pectin sol. For the last process, Alumina disk plate is coated with silica pectin sol by swab technique. Then, the coated membrane is calcinated for 2 hours at 300 °C by CTP technique. Coating processes are done as much as 4 layers. The morphology and thickness of membrane are characterized by Scanning Electron Microscopy (SEM).

2.3 Membrane Ultrafiltration

The membrane is installed to the ultrafiltration set-up by dead-end method as shown in Figure 1. Wetland water as a feed was filled into reactor before ultrafiltration process runs and then close all valves except compressor valves. Next, ultrafiltration process run with various pressure (0.5 bar, 1 bar and 1.5 bar). The water flux (kg.m⁻².h⁻¹) and rejection are calculated using equations below.

$$F = \frac{m}{(A\Delta t)} \quad (1)$$

$$R = \frac{C_f - C_p}{C_f} \times 100\% \quad (2)$$

m is flux mass (kg), A is the surface-active area of membrane (m²), Δt is operation time (hour), R is removal /rejection efficiency (F), C_f and C_p are the feed and permeate concentration and flux concentration. The NOM removal is measured by UV254 parameter (Spectrophotometer Genesys 10S UV-Vis). Salt rejection is estimated from calibration curve that suit with conductivity data measured (OHAUS ST300C).

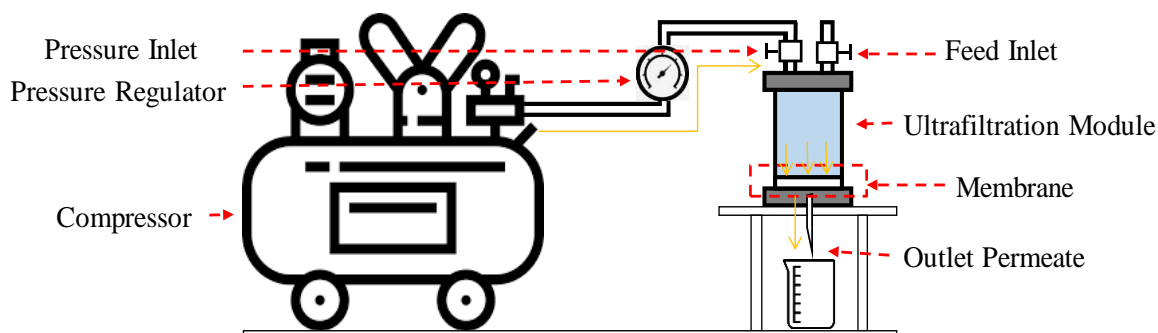


Fig. 1. Illustration of ultrafiltration experiment set up

3. Results

3.1 Membrane Characterization

3.1.1 Xerogel characterization

The FTIR spectra is carried out to measure the functional groups in silica pectin sol [7]. The FTIR spectra of silica pectin xerogel are shown in Figure 2. The result show xerogels has siloxane (Si-O-Si), silanol (Si-OH) and silica carbon (Si-C) functional groups. Siloxane functional groups are detected at wavelength 1061 cm^{-1} . Then at wavelength 966 cm^{-1} indicates the presence of silanol and the other peak in the figure is silica carbon which detected at wavelength 795 cm^{-1} . This results practically similar to our previous research reported by Mustalifah *et al.*, [18]. It explains siloxane at wavelength 1080 cm^{-1} , silanol at wavelength 976 cm^{-1} and silica carbon at wavelength 790 cm^{-1} . Siloxane functional groups is shaping membrane pores into mesopore. But more silanol effect the smaller form size (micropores). The combination between siloxane and silanol increase the membrane performance. These results are water flux becomes lower and transportation resistance gets higher. It means that a better rejection [4]. The presence of silica carbon groups is due to the addition banana peels pectin as carbon resource that increase strength and membrane hydro stability. Moreover, silica carbon support siloxane establishment [39].

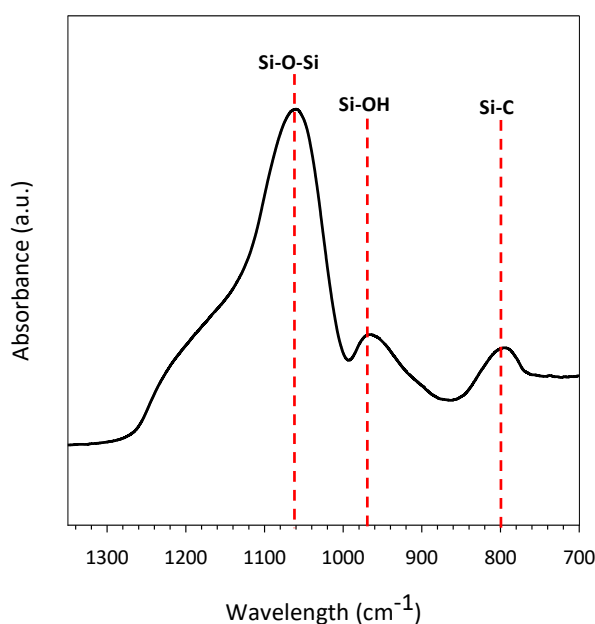


Fig. 2. FTIR spectra for the xerogel silica pectin

3.1.2 Membrane morphology

The structure of the silica-pectin membrane was analysed using Scanning Electron Microscopy (SEM) which shows on Figure 3. SEM analysis aims to determine the surface morphological structure of the membrane. The surface form of the membrane can affect the performance of membrane [14,18]. Figure 3 and Figure 4 show the SEM image of surface and cross-section silica pectin membrane by swab coating technique.

In Figure 3 silica pectin membrane in this work shows rough on the surface, where alumina particles look clearly. In fact, silica pectin sol infiltration into alumina membrane support similar to Elma *et al.*, [46] previous research. This is related to the dry porous surface contact and sol, which induces wetting forces like capillary forces and modulated by surface liquid tension [47]. Figure 4 show cross-section image of silica pectin membrane. Cross-section identify the formation of the coating layer attached [14]. Based on Figure 4 shows top part of the membrane does not show thin film formation. The silica pectin sol gets infiltrated into the pores of the alumina membrane support. It has been proved that the top part of the membrane is denser than the lower level in Figure 4. Swab technique in this work makes less silica pectin sol used than other coating methods and so silica pectin thin film membrane does not form. Refer to our previous work reported thin film sol can fill the pores of the membrane support which has big pores size (100 μm) [28].

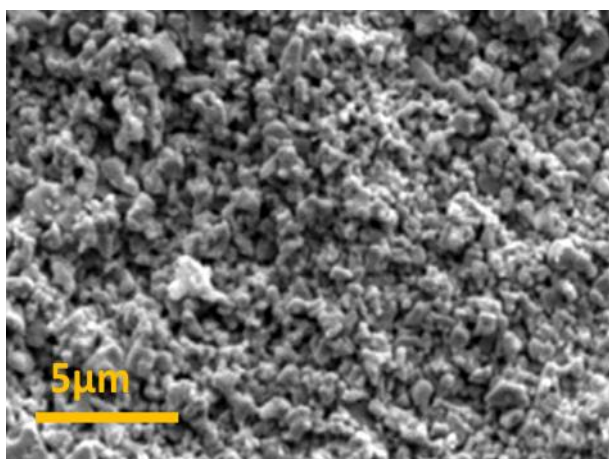


Fig. 3. SEM image of silica pectin membrane for top surface

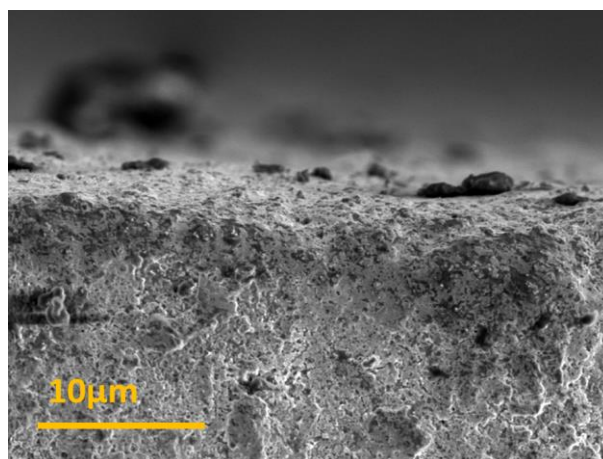


Fig. 4. SEM image of silica pectin membrane for cross-section

3.2 Characteristic of Wetland Water

The type of wetland water used is peat water and wetland saline water. Peat water is obtained from Suka maju village and wetland saline water are from Muara Halayung village, South Kalimantan-Indonesia. Generally, these wetland water have low qualities due to its characters. The characteristic of these water is written in Table 1. pH value of these wetland water is below of the drinking water standard quality. These pH results are not much different than previous research by Rahma *et al.*, [48] and Elma *et al.*, [44] with the same type of water. UV 254 is representing aromatic NOM [26]. UV254 value in peat water and wetland saline water show high content of NOM. Same results are obtained from previous research by Rahma *et al.*, [48] which classify the wetland water has high UV254 value.

Table 1
 Characteristics of wetland water

Parameters	Feeds		Unit	Standard
	Peat Water	Wetland Saline Water		
pH	5.14	6.35	-	6.5-8.5
UV ₂₅₄	0.947	0.396	cm ⁻¹	-
Conductivity	58.03	10500	μS.cm ⁻¹	-

3.3 Membrane Performance
3.3.1 Membrane water flux

Banana peels pectin template silica ultrafiltration membrane performance is rated by water flux, UV254 and salt rejection in the various pressure operation. Higher flux and removal value signify high performance of silica pectin membrane in treating water [22]. This research conducted with 3 various ultrafiltration operating pressure (0.5 bar, 1 bar and 1.5 bar). Figure 5 shows the influence of pressure on the silica pectin membrane performance. Based on the result from 0.5- 1.5 bar, peat water flux obtains 5.07 – 18.90 kg.m⁻².h⁻¹. Besides, wetland saline water has preponderant water flux. They are 7.04 – 20.52 kg.m⁻².h⁻¹. This result exhibits higher pressure obtains higher water flux [26,49]. Similar result is obtained from other research by Elma *et al.*, [50] that higher filtration pressure affects membrane operation driving force so the water flux enhanced. Also, the graph define wet saline water flux are higher than peat water. This different result of water flux ultrafiltration is caused by particulate concentration contained in water. Water fluxes are decreasing along increasing NOM concentration which represented by UV254. NOM particulate are attached in the pores and affect the permeability [51]. Shown in Table 1 that peat water UV254 value is higher than wetland saline water thus proving lower water flux.

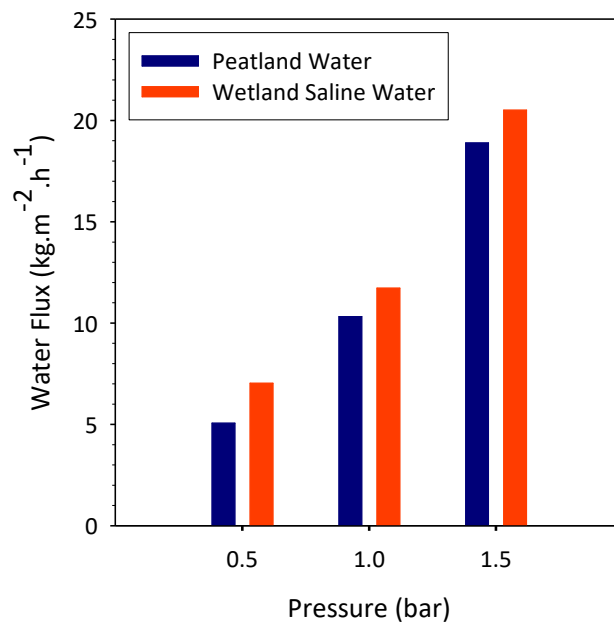


Fig. 5. Water flux performance of wetland water ultrafiltration on silica pectin membrane

3.3.2 NOM removal

Figure 6 shows the NOM removal efficiency ultrafiltration performance. NOM removal are determined by UV254, indicating aromatics/hydrophobics compounds can be preferentially removed over membrane [26,52]. Peat water removal obtain 85.56, 85.49 and 84.68 wt.% and wetland saline water is 82.50, 79.46 and 78.11 wt.% sequentially by pressure. Generally, the removal efficiency of NOM decreased along increasing pressure. The result is obtained by previous studies that the magnitude of NOM removal is inversely proportional to the applied pressure [50]. The deformation of the membrane due to high pressure, which causes membrane compaction that constrict the pore size and thicker foulant layer that became the secondary filter on top of the silica-pectin membrane. These results demonstrated that no remarkable differences when varying pressure and feeds which differences NOM concentration values. It is just NOM removal efficiency of peat water are higher than wetland saline water. The differences NOM removal efficiency are due to NOM concentration feed [52]. Increasing NOM will increase NOM removal efficiency. During ultrafiltration runs, NOM particulate are deposited on the membrane surface and pores so increase removal efficiency and decrease water flux [53].

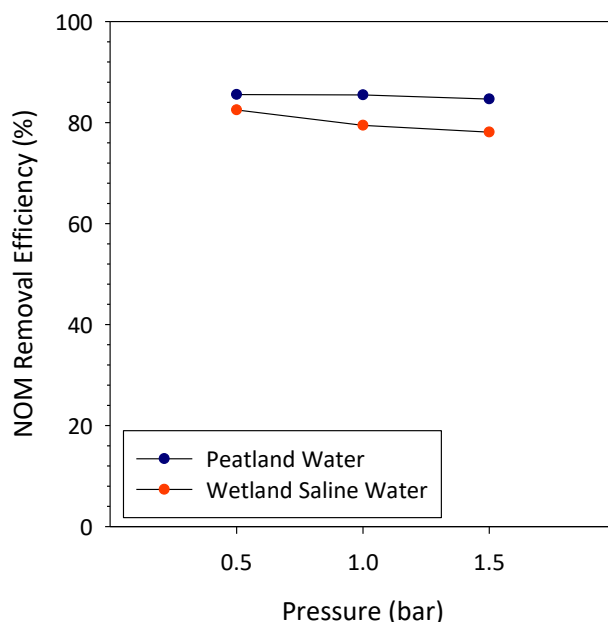


Fig. 6. NOM removal efficiency represented by UV 254 absorbances

3.3.3 Salt rejection

Salt rejection shows the ratio of the amount of salt concentration into the feed towards water permeate [41]. Salt concentration are correlated to conductivities of the water determined by a conductivity metre [42]. This experiment pointing on wetland saline water because it is classified as sea water typical with higher salt concentration (1-3.5%) [54].

Figure 7 shows the influence of pressure on salt rejection for silica pectin membrane ultrafiltration. Experimental runs have allowed to detect a decreasing salt rejection as a function of pressure operation on silica pectin membrane. Results showed decreasing salt rejection from 84.39% to 81.04% along increasing pressure. This is due to the increase in flux with increasing operating pressure which enhances the degree of concentration polarization and thus reduces the salt rejection

due to higher concentration of salt in the solution deposited onto the membrane and undermines the contribution of diffusion relative to convective salt transport [1,55]. Besides, salt rejection occurs by silica-pectin coated on membrane which is in the diameter range between water molecule and hydrated salt ions [56].

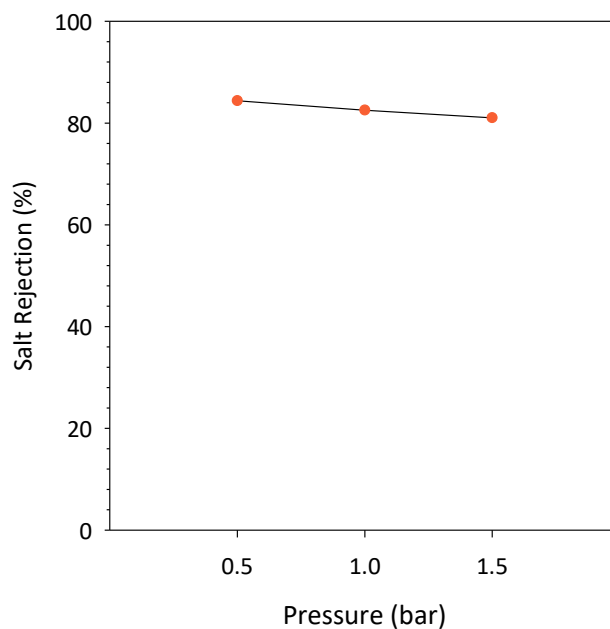


Fig. 7. Salt rejection performance

4. Conclusions

This work silica-pectin membrane has been successfully fabricated silica pectin membrane by CTP calcination process. The FTIR spectra of silica pectin membrane indicates siloxane (Si-O-Si), silanol (Si-OH) and silica carbon (Si-C) functional groups. Ultrafiltration performance on various pressure show water flux reach $18.90 \text{ kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ on peat water and $20.52 \text{ kg}\cdot\text{m}^{-2}\cdot\text{h}^{-1}$ on wetland saline water in 1.5 bar pressure operation. The highest value of NOM removal efficiency in peatland water reaches 85.56% and wetland saline water is 82.50% in 0.5 bar. Besides, salt rejection ability in wetland saline water shows 84.39% rejection in 0.5 bar and decreasing along increasing pressure operation. These results define increasing water flux by increasing pressure operation affected decreasing NOM removal efficiency and salt rejection.

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