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Research Article

PERFORMANCE OF FLAT MEMBRANE SILICA PECTIN ON WETLAND SALINE WATER AND ACID MINE DRAINAGE VIA ULTRAFILTRATION PROCESS

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ABSTRACT

Wetland saline water and acid mine drainage are found in South Kalimantan, Indonesia. Ultrafiltration process is potential way applied for desalination wetland saline water, which contains a high salt concentration, and acid mine drainage induced by the mining industry causes environmental and economic issues acid. Membrane fouling deteriorates membrane performances. To resolve this problem, flat membrane was chosen to maintain the water flux and fouling of silica pectin membranes via ultrafiltration process. Membranes were developed through a sol-gel method with silica source deposited from tetraethyl orthosilicate (TEOS), pectin as carbon template from banana peels. The sols were swab coated onto flat alumina (Al_2O_3) support for 4 layers followed by calcination. Performance of ultrafiltration flat membranes were evaluated by feeding wetland saline water and acid mine drainage at room temperature. The performance of ultrafiltration flat membrane exhibited excellent performance with the highest water fluxes 20.52 kg m-2 h-1 with salt rejection 81.05% for wetland saline water and 22.18 kg.m^{-2}.h^{-1} with salt rejection 51.08% for acid mine drainage. Permeate fluxes observed in ultrafiltration were greater than those obtained by pervaporation, making it a promising option for the treatment of wetland saline water and acid mine drainage.

1. INTRODUCTION

Indonesia, especially in South Kalimantan, it is found natural water resources such as wetland and mining activities play important roles in the economy ` . Acid mine drainage is usually produced within and around mining areas (Tong, Fan, Yang, & Li, 2021). Mining activities in South Kalimantan using open pit mining method (F. Mustalifah, Rahma, & Elma, 2021). Mining is a lucrative but also high-risk industry, one of the negative impacts is acid mine drainage (Alghifary & Sihombing, 2022). If acid mine drainage is discharged arbitrarily without treatment, it will lead to a series of environmental problems and cause long-term environmental pollution (Tong et al., 2021). Acid mine drainage is usually produced within and around mining areas (Fornarelli[1], Mullett, & Ralph, 2013). Acid mine drainage is usually produced within and around mining areas (Fornarelli[1] et al., 2013). On the other hand, wetland saline water (WSW) in South Kalimantan has become a serious problem that should be solved (Assyaifi et al., 2021; Muthia Elma, Bilad, et al., 2022; Muthia Elma, Ghani, Rahma, Alyanti, & Dony, 2022; Lestari et al., 2020; Rahma, Elma, Rampun, Situngkir, & Hidayat, 2022). Both long dry season and seawater intrusion caused decreasing of water quality and impacted on water scarcity for tidal wetland area. Decreasing of water quality in tidal wetland is caused by salts tendency to build up in the groundwater due to water evaporation along dry season (Muthia Elma, Rahma, Pratiwi, & Rampun, 2020; Lestari et al., 2020; Rahma, Elma, et al., 2020b). The high number of natural organic matter contain in wetland water may cause its water has brown color and not consumable (Muthia Elma, Rezki, et al., 2020). One of the innovations that can be done to prevent and reduce contaminants contained in wetland saline water and acid mine drainage into surrounding water sources and the ground is the membrane technology with ultrafiltration process to reduce fouling or pollutants (Muthia Elma, Mustalifah, et al., 2020; M Elma et al., 2023; Mahmud et al., 2023; Pratiwi et al., 2023; Zahratunnisa et al., 2024).

Membrane fouling is a complex phenomenon, which needs to be properly characterized to gain better understanding of its behaviors under different filtration conditions (Ngo et al., 2021; Rahma et al., 2023). Membrane fouling is generally defined as pore blocking, deposition and accumulation of foulant and formation of cake layer on the membrane surface that blocks the passage for water permeation pollutants will decrease water flux and performance of membrane (Li, Sun, Lu, Ao, & Li, 2020; Wen, Bo, Jin, & Zhang, 2021). Silica membrane with Tetraethyl orthosilicate (TEOS) as main precursor in the polymerisation silica has excellent mechanical strength and thermal stability but has limitation due to low hydro-stability properties (Muthia Elma, Mustalifah, et al., 2020; Muthia Elma & Riskawati, 2018). Silica membrane can increasing hydro-stability properties by addiction of pectin, it’s make the membrane pore resistance stronger and has high salt rejection because strengthen the pore structure of the membrane (Muthia Elma, Nata, et al., 2022; Muthia Elma, Rezki, et al., 2020). (Gu, Kim, Kim, & Yang, 2011). Sol-gel process is categorized as a well-known, very capable with a high flexibility (Dehghanghadikolaei, Ansary, & Ghoreishi, 2018). Sol-gel process is very cheap, high flexibility on either coating composition or sample geometry (Muthia Elma, Suparsih, & Annahdliyah). Swab coated may not be suitable for all surface types due to loss of adhesive material after multiple uses, the benefits of higher particle collection efficiency are extremely promising, providing a strong substrate-coating bond and reliable coating process and simplicity of the process (Dehghanghadikolaei et al., 2018; Staymates, Grandner, & Gillen, 2011).

The most common approaches for membrane fouling control include modifying membrane surface properties (Rana & Matsuura, 2010; Shang, Xia, Sun, Lipscomb, & Zhang, 2022). In general, the criteria for membrane module configurations include two types: namely, flat sheet and tubular membrane modules (Bopape, Van Geel, Dutta, Van der Bruggen, & Onyango, 2021). Flat membranes offer benefits in simplicity, better flow control on both the permeate and feed side (Wen et al., 2021) ease of sheet replacement, less fouling tendency over tubular configurations (excessive fouling and membrane integrity problems). Ultrafiltration in membrane desalination has preferred technique has advantage as the driving force in the feed water and the permeate (Muthia Elma, Nata, et al., 2022). Ultrafiltration membranes have a good removal performance for suspended particles and microorganisms, including bacteria, viruses, Giardia and Cryptosporidium, which can effectively guarantee the biological safety of drinking water. Moreover, by substituting the traditional treatment processes such as sedimentation or sand filtration, the application of ultrafiltration contributes to reducing the coagulant dosage, saving the land occupation, as well as simplifying the process flow (Li et al., 2020; Yang et al., 2022). This research aims to investigate the performance (water flux and salt rejection) of flat membrane via ultrafiltration process using swab coated method.

2. RESEARCH METHODOLOGY

2.1. Chemical and Materials

Wetland saline water was collected from Muara Halayung village, Indonesia. Acid mine drainage taken from Kintap South Kalimantan, Indonesia. Silica pectin sol was prepared by using tetraethyl orthosilicate (TEOS, 99%, Sigma-Aldrich) as silica precursor, ethanol (EtOH, 70%, Merck), ammonia (0.0003 M NH_3, Merck), dilute nitric acid (0.0008 M HNO_3, Merck), pectin from banana peel, glycerol (85%, Merck), and aquadest. for membrane support, this research using a macroporous α-Al_2O_3 flat support membrane with diameter 0.0393 m.

2.2. Fabrication and Characterization of Flat Membrane

20 mL Ethanol was added to a three-neck flask and cooled using to 0°C and stirred using a magnetic stirrer at 250 rpm for 5 minutes. Then 18.66 g TEOS was added into the three-neck flask slowly and silenced for 5 minutes while stirring 8.0699 g HNO_3 0.00078 N was dropped wisely into solution. The mixtures of Et-OH + TEOS + HNO_3 were refluxed and stirred for 1 hour at 50°C. After 1 hour, the NH_3 solution was added into mixtures and continue refluxed for another 2 hours at the same temperature. At the end, the pH of silica sols wascheck, adjustedto pH ± 6. In other side, preparing carbon template from pectin which extract from banana peels. Prepared glycerol at 50 °C for 90 minutes to mixtures pectin-glycerol (0.5 % pectin concentration), the silica-pectin sols was swab coated onto flat membrane 1 hour for 1 layer, until 4 layers. Membrane was characterization preparing with dried sols into oven for 24 hours to product xerogel. Xerogel was calcined at 300 °C and characterized using FTIR spectra and SEM.

2.3. Membrane Ultrafiltration

Performance and evaluation of flat membrane determine using ultrafiltration process. Wetland saline water and acid mine drainage water is used to determine the resistance and ability of silica-pectin membrane with measure of water flux, and salt rejection. Feed water pressures was variations (0.5 bar, 1 bae and 1.5 bar) were carried out at room temperature. Ultrafiltration was carried out for 5 minutes. Set up ultrafiltration process was design as shown in Figure 1.

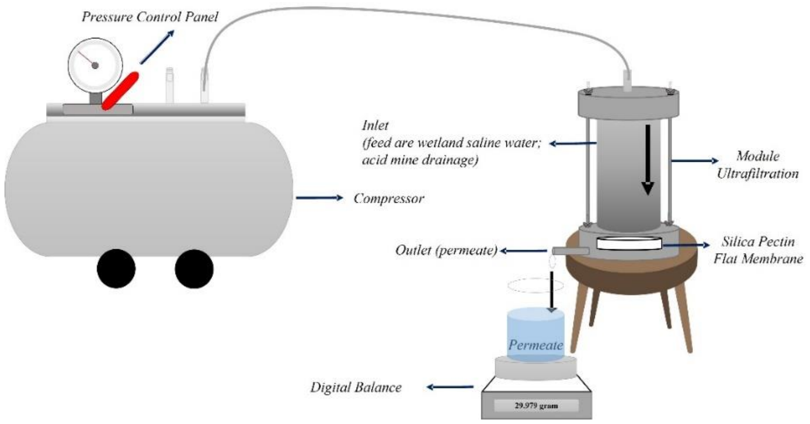

Figure 1. Set Up Ultrafiltration Process

In any membrane filtration, the prediction of permeate flux is critical to calculate the membrane surface required, which is an essential parameter for scaling-up, equipment sizing, and cost determination. The water flux of flat membrane was measured based equation:

$$F = \left(\frac{M}{A\ dt}\right) \qquad (1)$$

Where mass of flux permeate (kg), A is an activated-area on surface silica-pectin membrane (m^2) and operational times was symbol of dt (h). Salt rejection of flat membrane measured with the equations:

$$R = \left(\frac{Cf - Cp}{Cf}\right) \times 100\% \qquad (2)$$

Where Cf is the salt concentration (wt %) in the wetland saline water and acid mine drainage before treatment and Cp is a salt concentration in water permeate (wt %) after treatment. It was determined by a conductivity meter correlated to the conductivity of retentate and permeate.

3. RESULT AND DISCUSSION

3.1. Characteristic of Wetland Saline Water and Acid Mine Drainage

Wetland saline water was collected from Muara Halayung village, Indonesia and acid mine drainage taken from Kintap, South Kalimantan-Indonesia were treated using ultrafiltration by flat membrane silica-pectin. Generally, wetland saline water and acid mine drainage have bad qualities due to its water characteristics. Wetland saline water and acid mine drainage was characterized with parameters which shown successively in Table 1 and integrated with Indonesian regulatory standards for hygiene sanitation:

Table 1. Characteristic of Wetland Saline Water and Acid Mine Drainage in South Kalimantan, Indonesia

Parameter	Values		Unit	Standard Indonesia's Ministry of Health No. 32 2017
	Wetland Saline Water	Acid Mine Drainage		
pH	6.67	4	-	6.5-8.5
TDS	5500	324	mg/L	-
Conductivity	9370	652	µS/cm	-
NaCl	10000	500	ppm	-

3.2 Morphology and Characterization of Flat Membrane

1. Scanning Electro-Microscopy Membrane

Flat membrane silica-pectin morphology was determined by scanning electron microscopy (SEM) image which shows on Figure 2. Analyzed SEM aims to determine the surface morphological structure of flat membrane. The surface area of flat membrane affected the performance of membrane ultrafiltration process.

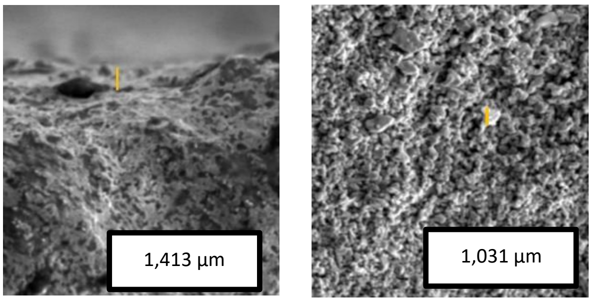

Figure 2. SEM of Flat Membrane Silica Pectin (a) Cross Section (b) Surface Area

From this Figure 2 (a), the cross section of flat membrane silica pectin has thickness for estimated to be ~2 µm. Cross section SEM images of flat membranes exhibited the thickness of the silica-pectin top layer. The membrane with thin film was thinner result higher of water flux (Muthia Elma & Assyaifi, 2018). In this study, the flat membrane using silica pectin with swab coated to reduce and to low cost in production. Therefore, in this work, swab coated of flat membrane silica-pectin was carried out 4 times to result 4 layers. Figure 2 (b) shows the surface area of flat membrane silica-pectin. The calcined temperature also assisted on silica-pectin layer thickness. Silica-pectin membrane sintered over 300 °C becomes thinner. Solvent and water trapped in silica matrices and then evaporated at 400 °C, after that thin layer was created (Muthia Elma, Mustalifah, et al., 2020; Muthia Elma, Nata, et al., 2022; Muthia Elma et al.; Lestari et al., 2020; Rahma, Elma, et al., 2020b).

2. Fourier Transform Infra-Red

Sol gel is a process to synthesize inorganic compound through chemical reaction in solution under low temperature converting sol to gel (Naseer, Ha, Lee, Park, & Song, 2022). The process of sol gel is an easy and cheap method with great coating properties on the substrate, its applications are being increased, relatively easy rather than other coating processes and it provides a good enhancement, the most effective and simplest way to prepare thin membrane layer (Dehghanghadikolaei et al., 2018; Naseer et al., 2022). Xerogel flat membrane silica pectin was analysed using FTIR technique. Figure 3 shown region the FTIR spectra calculated peaks at 1100-700 cm^{-1}. In Figure 3 wavelength at 1061 cm^{-1}, 970 cm^{-1}, and 793 cm^{-1} are wavenumber of siloxane (Si-O-Si), silanol (Si-OH) and silica-carbon (Si-C) respectively. Silanol and siloxane groups are known to affect pore size of the membrane (Muthia Elma, Rakhman, & Hidayati, 2018). The silanol concentration is much smaller than the siloxane concentration, this is because silanol groups tend to form smaller pore size (microporous) (Amalia, Mirwan, & Farid, 2019; Muthia Elma, Rezki, et al., 2020). High concentration of siloxane region with an area of silanol region representing mesoporous or micropores, siloxane can strengthen the membrane structures. The silica-carbon group strengthen Si-OH bonds and contributed to determines the pore on the flat membrane silica-pectin, carbon chain clusters precedence to the formation of small pores (Amalia et al., 2019; Muthia Elma, Mustalifah, et al., 2020; Muthia Elma, Sumardi, et al., 2021; Rahma, Elma, et al., 2020a).

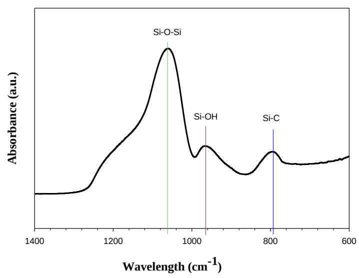

Figure 3. FTIR Spectra (Fourier- transform Infrared Spectroscopy) Xerogel Flat Membrane Silica-Pectin

3.2 Performance of Flat Membrane Silica Pectin

The ultrafiltration process gives many variables for execute desalination performance, flux of water an salt rejection. Permeate water is captured in a condenser and flux (kg. $m^{-2}.h^{-1}$). Performance of flat membrane silica-pectin in ultrafiltration process can be seen from the value of water flux and salt rejection. Figure 4 and Figure 5, both shows the result of membrane performance in terms of water permeate fluxes and salt rejections in various feed application for wetland saline water and acid mine drainage. The performance of flat membrane silica-pectin was shown in Figure 4 and Figure 5, below.

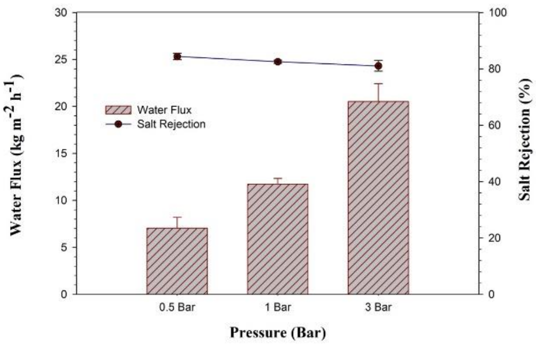

Figure 4. Performance of Flat Membrane Silica-Pectin for Wetland Saline Water (i) bar charts are water fluxes (left axis) and (ii) line scatters are salt rejections (right axis)

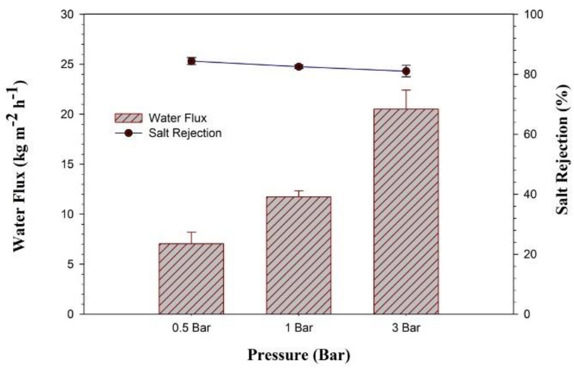

Figure 5. Performance of Flat Membrane Silica-Pectin for Acid Mine Drainage (i) bar charts are water fluxes (left axis) and (ii) line scatters are salt rejections (right axis)

Figure 4 and Figure 5 was shown the performance of flat membrane silica-pectin for wetland saline water and acid mine drainage at room temperature respectively. The result of this work showed performance of flat membrane silica pectin using ultrafiltration process is excellent. The flux is higher than using pervaporation process. The value of flux during the ultrafiltration process 5 minutes is 20.52 kg.$m^{-2}.h^{-1}$ with salt rejection 81.05 % at room temperature for wetland saline water. Flux from flat membrane silica-pectin for acid mine drainage result 22.28 kg.$m^{-2}.h^{-1}$ with salt rejection >50 %. From these results can be concluded that the greater the concentration of feeds, the lower the water flux as well as salt rejection. The membrane matrix may undergo structural changes in the texture and build up of salt molecules that cause clogging of the membrane pores, thus affecting the water flux values. In this research, the flat membrane silica-pectin will be tested at different feed variations, wetland saline water and acid mine drainage. It suggest due to fouling phenomenon similar to wetland water treatment such as previous work done by (Muthia Elma, Lestari, et al., 2021); F. R. Mustalifah, Rahma, Mahmud, Sunardi, and Elma (2021); (Rahma, Elma, Pratiwi, & Rampun, 2020) Desalination performance comparison for silica membranes which shown successively in Table 2, shows comparison among similar research about desalination. Silica membrane employing organic compound for sol gel process has a big contribution in desalination (Muthia Elma, Bilad, et al., 2022; Muthia Elma, Nata, et al., 2022; Muthia Elma, Rahma, Kusumawati, Pratama, & Alyanti, 2022; Mat Nawi et al., 2022; Rahma et al., 2023; Sumardi et al., 2021).

The most common approaches for membrane fouling control include modifying membrane surface properties (Rana & Matsuura, 2010; Shang et al., 2022). Swab coated may not be suitable for all surface types due to loss of adhesive material after multiple uses, the benefits of higher particle collection efficiency are extremely promising, providing a a strong substrate-coating bond and reliable coating process and simplicity of the process (Dehghanghadikolaei et al., 2018; Staymates et al., 2011). Flat membranes offer benefits in simplicity, better flow control on both the permeate and feed side (Wen et al., 2021) ease of sheet replacement, less fouling tendency over tubular configurations (excessive fouling and membrane integrity problems) (Wen et al., 2021; Yang et al., 2022). The application of ultrafiltration contributes to reducing the coagulant dosage, saving the land occupation, as well as simplifying the process flow (Li et al., 2020; Yang et al., 2022).

Table 2. Desalination Performances Comparison for Silica Membranes

Types of Membrane	Desalination Method	Coating Method	Feed Variations	Water Flux	Salt Rejection	References
Tubular	Pervaporation	Dipcoating	Wetland Saline Water	0.93 $kg.m^{-2}.h^{-1}$	99%	(Lestari et al., 2020)
Tubular	Pervaporation	Dipcoating	Acid Mine Drainage	8.4 $kg.m^{-2}.h^{-1}$	99%	(Muthia Elma, Mustalifah, et al., 2020)

Types of Membrane	Desalination Method	Coating Method	Feed Variations	Water Flux	Salt Rejection	References
Tubular	Pervaporation	Dipcoating	Wetland Saline Water	1.25 $kg.m^{-2}.h^{-1}$	96%	(Muthia Elma et al., 2018)
Tubular	Pervaporation	Impregnation	Brackish Water	12.46 $kg.m^{-2}.h^{-1}$	89.49 %	(Huda et al., 2021)
Tubular	Ultrafiltration	Phase Inversion	Peat Wter	92.5 $L.m^{-2}.h^{-}$	-	(Muthia Elma, Pratiwi, et al., 2021)
Tubular	Ultrafiltration	Phase Inversion	Peat Water	39.91 $L.m^{-2}.h^{-}$	-	(Muthia Elma, Rampun, et al., 2020)
Multichannel	Pervaporation	Inner Coating	Brackish Water	15.74 $kg.m^{-2}.h^{-1}$	99.77%	(Muthia Elma, Nata, et al., 2022)
Flat	Ultrafiltration	Swab Coating	Weatland Saline Water and Acid Mine Drainage	20.52 $kg.m^{-2}.h^{-1}$ for wetland saline water 22.28 $kg.m^{-2}.h^{-1}$ for acid mine drainage	81.05 % for wetland saline water >50 % for acid mine drainage	This Work

3. CONCLUSION

3.2 Kesimpulan

Based on the results, flat membrane silica-pectin have siloxane and silanol groups. The flat membrane silica-pectin prepared through the swab coating obtained a shining surface because of the thin film attached on alumina support. The highest water flux achieved from flat membrane silica-pectin for wetland saline water 20.52 kg.m^{-2}.h^{-1} with salt rejection 81.05 % and flux from flat membrane silica-pectin for acid mine drainage result 22.28 kg.m^{-2}.h^{-1} with salt rejection >50 % at 3 bar.

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