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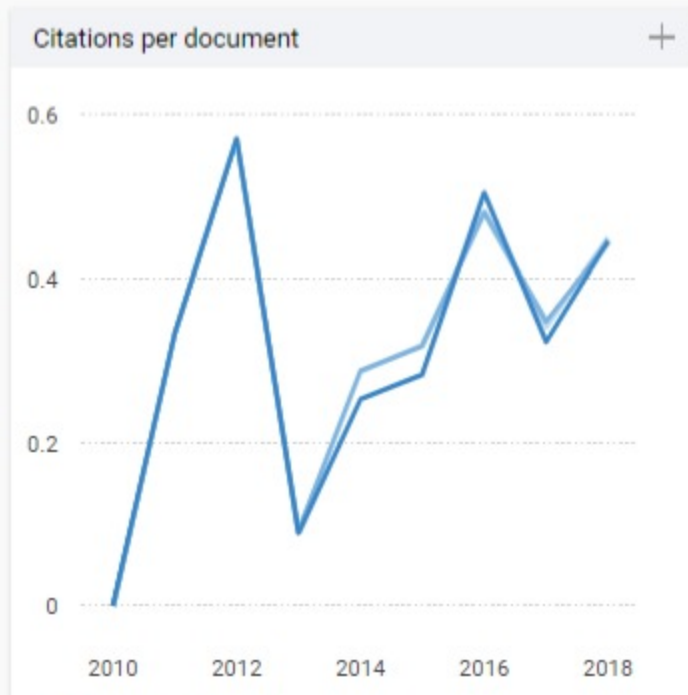

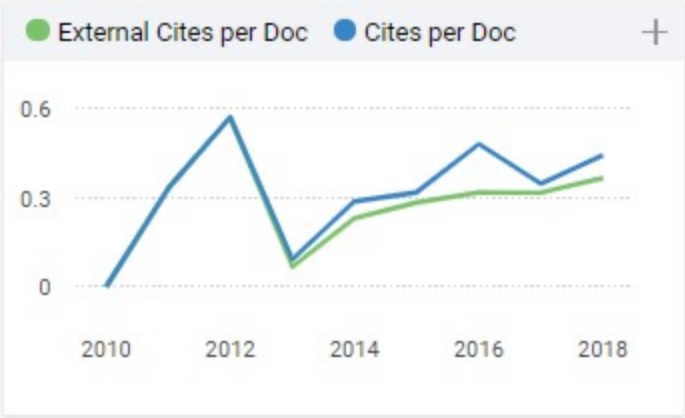

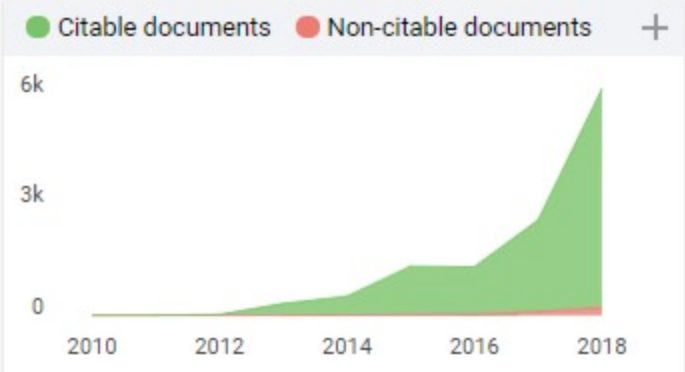

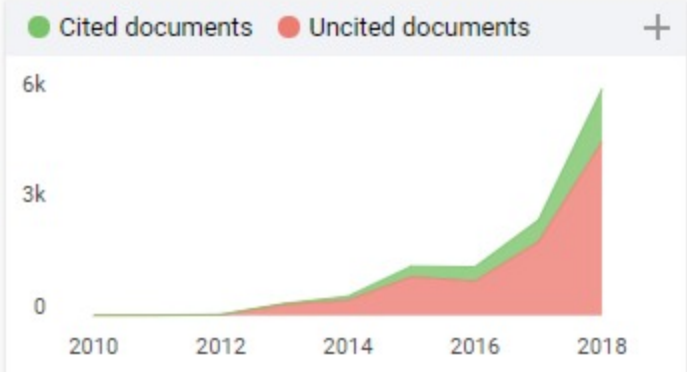

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Organo-silica membrane for brine water pervaporation

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Abstract

Pervaporation is currently used for brine water desalination. This high water salt content may impact the environment if it may not be treated. So, in this paper demonstrates the performance of organo-silca membrane to produce potable water from brine water. Organo-silica membrane was employed by sol gel process for 3 hours using tetra orthosilicate (TEOS) as precursor as well as dual catalyzed (organo catalyst/citric acid) and base (ammonia) on reflux temperature of 0°C. The molar ratios were 1:38:0.0007:5:003 for TEOS:EtOH:Citric Acid:H2O:NH3. Organo-silica membrane was applied in various artificial brine water (7.5-15 wt % NaCl) operated at 40 and 60°C as feed temperatures. The citric acid was chosen as carbon source to increase the carbon chains in the silica matrices. It was found that the water flux was increased as the feed temperature also increased from 40°C (1.27 $kg.m^2.h^{-1}$) to 60°C (1.55 $kg.m^2.h^{-1}$). Organo–silica membranes can produce water with a salt rejection of >90%.

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Organo-silica membrane for brine water pervaporation

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Abstract. Pervaporation is currently used for brine water desalination. This high water salt content may impact the environment if it may not be treated. So, in this paper demonstrates the performance of organo-silca membrane to produce potable water from brine water. Organo-silica membrane was employed by sol gel process for 3 hours using tetra orthosilicate (TEOS) as precursor as well as dual catalyzed (organo catalyst/citric acid) and base (ammonia) on reflux temperature of 0 ℃. The molar ratios were 1:38:0.0007:5:003 for TEOS:EtOH:Citric Acid:H_2O:NH_3. Organo-silica membrane was applied in various artificial brine water (7.5-15 wt % NaCl) operated at 40 and 60 ℃ as feed temperatures. The citric acid was chosen as carbon source to increase the carbon chains in the silica matrices. It was found that the water flux was increased as the feed temperature also increased from 40 ℃ (1.27 kg.m^2.h^{-1}) to 60℃ (1.55 kg.m^2.h^{-1}). Organo–silica membranes can produce water with a salt rejection of >90%.

1. Introduction

Brine water from industrial effluent is need to undergoes treatment before release to environment. Since industrial activities are increase in last decade, the world meet the challenge to keep water clean. To be specific, brine water can be produced from ballast water, desalination, ion-exchange regeneration, etc. Also, brine water has adverse effect because it contain high salt concentration and other pollutant [1, 2]. Pollutant is depends on the source. For example, desalination has been contaminated by chemical utilized. Ion-exchange regeneration, contaminated with resins and other organic pollutant [3]. Whereas, microorganisms in ballast water is harmful and may affect the environment when it discharged [4].

Pervaporation (PV) in particular is a separation process worked by evaporates the feed to form permeate vapor and followed by condensed it downstream. Indeed, this process offer rejection close to 100%. PV in operation need a vacuum at permeate side for start-up [5]. Unlike Reverse Osmosis (RO), PV can treat brine water without high pressure pump [6]. In PV, chemical potential gradient between feed and permeate is the membrane driving force [7]. PV denote several benefits such as high rejection, low operating cost and low pressure required [8].

Inorganic membranes are suitable for high temperature and normally prepared by thin film deposition on macro porous substrates. It also shows high chemical resistant [9]. Silica have been proposed in pervaporation application [10-12]. However, when silica membrane expose to water, the

performance might be demote due to low hydro stability [12]. Some improvement has been made to overcome this problem such as carbon template silica [12-14], hybrid organic-inorganic [15-17] and incorporation of metal oxide silica membranes [18-21]. These approach will prevent silica collapsed.

Another important requirement in membrane fabrication is catalyst. Not only choose appropriate material is needed, catalyst playing role in pore formation. Gas separation is normally used smaller pore in the presence of acid catalyst (*e.g.* HNO_3 or HCl), because they need high rejection, but this type of membrane does not good for desalination [22, 23]. Meanwhile, base catalyst (*e.g.* NH_3) provides macropore and larger water flux. However rejection is generally still low [11]. Regarding this, dual acid-base catalyst is often used. Citric acid can replace the use of carbon precursor in silica matrix. This membrane called organo-silica. Previous research by Elma, et al. [24] successfully fabricated organo-silica membrane but at lower temperature 250 °C. By applied citric acid, it would reduce cost of membrane fabrication. Therefore, in this study we demonstrate the performance of organo-silica membrane to produce potable water from brine water using dual acid base catalyst.

2. Materials and methods

2.1. Materials

Tetraethyl orthosilicate (TEOS, 99%, Sigma-Aldrich), Ethanol (EtOH, 99%) and aquades, dilute citric acid (0.0007 Merck), NH_3, (25%, Merck), membrane support (-Al_2O_3), NaCl, conductivity meter, and furnace.

2.2. Organo-silica membrane preparation

Organo-silica membrane was prepared using sol-gel method. TEOS was dissolved into ethanol (EtOH, 99%) under stirring for 5 min at 0°C to avoid the partial hydrolysis reaction. After that, the reflux was started by the addition of diluted citict acid (0.0007 N $C_6H_8O_7$) for 1 h at 50°C. Then, the second reflux of ammonia solution (NH_3, 25%, Merck) diluted in ethanol was added drop wise into sol mixture for 2 h. silica sols was dried in a temperature controlled oven at 60°C for 24 h to obtain the dried gel. The final molar ratios of the TEOS:EtOH:$C_6H_8O_7$:H_2O:NH_3 sol were calculated to be 1:38:0.0007:5:*x*, where *x* was 0.0003 as NH_3. This organo-silica sols was placed in the petri dish and dried in the oven at temperature 60°C for 24 hours. Then, organo-silica xerogel was grounded into powder and calcined in air at 600°C.

The organo-silica thin films were coated on macro porous alumina substrate (α-Al_2O_3 tubular support (Ø≈100 nm), Ceramic Oxide Fabricates, Australia) via dip-coating process with a dwelling time of 2 min, dipping rate of 10 cm min^{-1} and withdrawal rate of 5 cm min^{-1}. Then, the deposited membrane layer was dried and calcined in air at 600 °C. via RTP techniques for 1 h without applying ramping/cooling rates. A process of dip-coating, drying and calcination was repeated for 4 times to get organo-silica membrane 4 layers for desalination application. All membranes were characterized by Scanning Electron Microscopy (SEM JOEL ZEISS) to investigate its morphology and thickness.

2.3. Pervaporation

The membrane was set into a pervaporation set-up to perform in desalination as shown in Figure 1. One of sided membrane was blocked and another one was connected to a cold trap (immersed in liquid nitrogen) and the vacuum pump was operated at 1 bar. The artificial brine water were prepared using (NaCl, Sigma Aldrich) in variance of 7.5, 10 and 15 wt %. Pervaporation was operated at variance temperature of feed in ranging of 40 °C and 60°C which was controlled by thermocouple and hotplate. Homogenous of feed solutions was controlled in constant stirring to prevent the polarization. The water flux, F ($kg.m^{-2}.h^{-1}$), was determined based on the Equation $F = m / (A \cdot \Delta t)$, where m is the mass of permeate (kg) retained in the cold trap, A is the surface-active area (m^2) and Δ is the time measurement (h). The salt rejection, R (%), was calculated as $R = (Cf\text{-}Cp) / Cf \times 100\%$, where C_f and C_p are the feed and permeate concentrations of salt (wt %).

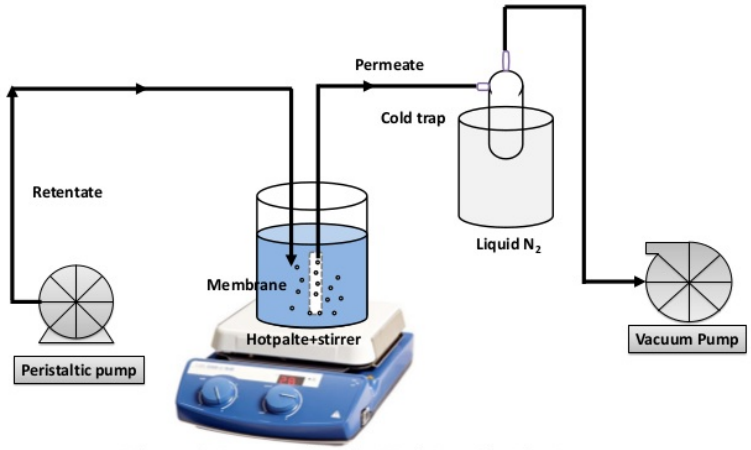

Figure 1. Pervaporation Set Up in Desalination Process.

3. Results and discussion

3.1. SEM characteristics

Before pervaporation test, membrane was characterized by SEM to show interlayer-free membrane thickness and surface (Figure 2 and Figure 3). Organo-silica membrane in this work, shows rough in surface (Figure 2). This is because organo-silica sol infiltration occurs when membrane support dipped into it. Nevertheless, calcination would offer thermal induced. Organo-silica membrane thickness (1 μm) prepared are thicker than previous study around ~470 nm [11] (Figure 3). RTP calcination process is done shorter than the Conventional Thermal Process (CTP) makes imperfect solvent evaporation. Different with CTP, RTP process does not use the heating rate and cooling rate. Despite this, RTP take longer time (more than one week) instead of CTP (1 day) [25].

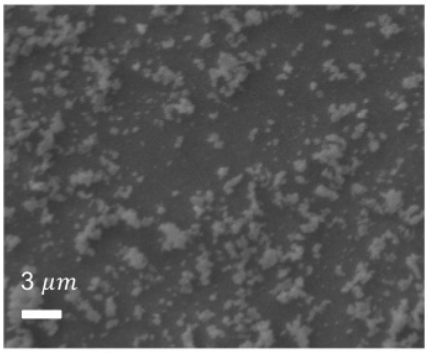

Figure 2. Surface area of interlayer-free organo-silica membrane during calcination in air.

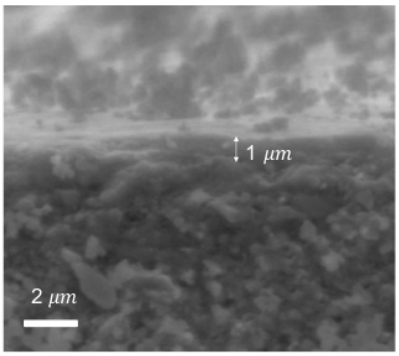

Figure 3. Cross-section area of interlayer-free organo-silica membrane during calcination in air.

3.2. Pervaporation of brine water using organo-silica membranes

The effect of feed temperature on membrane performance relates to water flux. Figure 4 shows water fluxes enhance as feed temperatures increase from 40 to 60 °C. It raise vapour pressure and membrane driving force [26]. Highest temperature delivers high water flux as well. For organo membrane silica with brine water as feed, the highest is 1.55 $kg.m^2.h^{-1}$ Brine water. Rather than using RO,

pervaporation were able to treat brine water without extra osmotic and hydraulic pressure [27]. It reduces energy consumption.

Compare to polymeric membranes, this organo-silica membrane performed better (Table 1). It produce larger water flux of 1.55 $kg.m^2.h^{-1}$ than single acid catalyzed only. It suggests citric acid can maximize the performance. Recent studies show higher water flux compare to our study because organo silica membrane prepared in our work, it was calcined in high temperature (600 °C). In this temperature, carbon has been lost and reduce membrane performance (Table 1). Temperature should be considered first. In sol gel method, catalyst give impact to silica matrix, which alkoxysilane come through hydrolysis and condensation reactions [28]. Mesoporous structure will be formed in the presence of acid base dual catalyst. Apply acid catalyst only would make membrane pore smaller. Small pore which is known as micropore mostly used in gas separation and it does not suit for pervaporation due to lower water flux. Whereas base catalyst addition give larger water flux, but poor rejection. Bottle neck pore appear with combination of acid and base. This type of pore, would allow H_2O (2.6 Å) easily diffuse and bigger molecules such as Na^+ (7.16 Å) and Cl^- (6.64 Å) will be hindered on bottle neck pore. But still provide high water fluxes [11].

In addition, in this work, carbon chain from citric acid resist pore collapsed in silica matrix. It creates stronger membrane structure and higher hydrostabilty [29], Rangelova, et al. [30]. Citric acid reportedly provides mesoporous structure. This work supports the benefit of carbon effect which agree in similar carbon templated researches for pectin [10, 31] and P123, where give less effect of hydrated ion retention on membrane surface.

Despite increasing water fluxes, salt rejection slightly reduces until 90% after organo-silica membrane treat on higher salt concentration (15 wt%) for both temperatures. Salt polarization occurs in this case. It happen when salt particles accumulated on membrane surface. This phenomena will block the membrane pores in period of time. As a result, membrane will produce lower water fluxes because a way [11]. As explain before some salt particles might be detained on bottle neck pore. Beside it, raising temperature also reduces salt rejection 38.5% for all brine concentration.

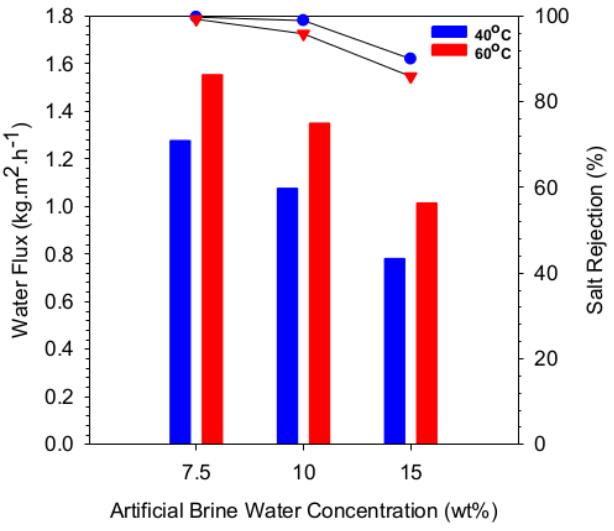

Figure 4. Performance of interlayer-free organo-silica membrane for brine water pervaporation.

Table 1. Comparison of membrane performance for pervaporation in different catalyst, feed temperature, and feed.

Membrane type	Catalyst	Feed temperature (°C)	Feed	Water fluxes ($kg.m^2.h^{-1}$)	Salt rejection (%)	Ref
Organo-silica membrane	Acid-base	40-60	NaCl 3.5%	1.55	99	This work
Polyelectrolyte	Acid	58.5	Ethanol/water	0.5	-	[32]
Polyetheramide	-	68-70	NaCl 32 g/L	0.56	99.99	[33]
Silica-Pectin	Acid-base	25	Wetland saline water	3.4	99.99	[10]
Silica-P123	Acid-base	25	NaCl 7.5wt%	6.6	99.5	[13]

4. Conclusion

From the experimental results, the operating parameter in pervaporation which is feed temperature can increasing water flux from 40 °C (1.27 $kg.m^2.h^{-1}$) to 60 °C (1.55 $kg.m^2.h^{-1}$). Organo –silica membranes give salt rejection of >90%. It proves this membrane is suitable for treat high concentration salt. Pervaporation may reduce operation cost. In addition, membrane prepared with citric acid would make membrane performance more robust when it applied in pervaporation processes.

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Abstract. Pervaporation is currently used for brine water desalination. This high water salt content may impact the environment if it may not be treated. So, in this paper demonstrates the performance of organo-silca membrane to produce potable water from brine water. Organo-silica membrane was employed by sol gel process for 3 hours using tetra orthosilicate (TEOS) as precursor as well as dual catalyzed (organo catalyst/citric acid) and base (ammonia) on reflux temperature of 0 ℃. The molar ratios were 1:38:0.0007:5:003 for TEOS:EtOH:Citric Acid:H_2O:NH_3. Organo-silica membrane was applied in various artificial brine water (7.5-15 wt % NaCl) operated at 40 and 60 ℃ as feed temperatures. The citric acid was chosen as carbon source to increase the carbon chains in the silica matrices. It was found that the water flux was increased as the feed temperature also increased from 40 ℃ (1.27 kg.m^2.h^{-1}) to 60℃ (1.55 kg.m^2.h^{-1}). Organo–silica membranes can produce water with a salt rejection of >90%.

1. Introduction

Brine water from industrial effluent is need to undergoes treatment before release to environment. Since industrial activities are increase in last decade, the world meet the challenge to keep water clean. To be specific, brine water can be produced from ballast water, desalination, ion-exchange regeneration, etc. Also, brine water has adverse effect because it contain high salt concentration and other pollutant [1, 2]. Pollutant is depends on the source. For example, desalination has been contaminated by chemical utilized. Ion-exchange regeneration, contaminated with resins and other organic pollutant [3]. Whereas, microorganisms in ballast water is harmful and may affect the environment when it discharged [4].

Pervaporation (PV) in particular is a separation process worked by evaporates the feed to form permeate vapor and followed by condensed it downstream. Indeed, this process offer rejection close to 100%. PV in operation need a vacuum at permeate side for start-up [5]. Unlike Reverse Osmosis (RO), PV can treat brine water without high pressure pump [6]. In PV, chemical potential gradient between feed and permeate is the membrane driving force [7]. PV denote several benefits such as high rejection, low operating cost and low pressure required [8].

Inorganic membranes are suitable for high temperature and normally prepared by thin film deposition on macro porous substrates. It also shows high chemical resistant [9]. Silica have been proposed in pervaporation application [10-12]. However, when silica membrane expose to water, the

performance might be demote due to low hydro stability [12]. Some improvement has been made to overcome this problem such as carbon template silica [12-14], hybrid organic-inorganic [15-17] and incorporation of metal oxide silica membranes [18-21]. These approach will prevent silica collapsed.

Another important requirement in membrane fabrication is catalyst. Not only choose appropriate material is needed, catalyst playing role in pore formation. Gas separation is normally used smaller pore in the presence of acid catalyst (*e.g.* HNO_3 or HCl), because they need high rejection, but this type of membrane does not good for desalination [22, 23]. Meanwhile, base catalyst (*e.g.* NH_3) provides macropore and larger water flux. However rejection is generally still low [11]. Regarding this, dual acid-base catalyst is often used. Citric acid can replace the use of carbon precursor in silica matrix. This membrane called organo-silica. Previous research by Elma, et al. [24] successfully fabricated organo-silica membrane but at lower temperature 250 °C. By applied citric acid, it would reduce cost of membrane fabrication. Therefore, in this study we demonstrate the performance of organo-silica membrane to produce potable water from brine water using dual acid base catalyst.

2. Materials and methods

2.1. Materials

Tetraethyl orthosilicate (TEOS, 99%, Sigma-Aldrich), Ethanol (EtOH, 99%) and aquades, dilute citric acid (0.0007 Merck), NH_3, (25%, Merck), membrane support (-Al_2O_3), NaCl, conductivity meter, and furnace.

2.2. Organo-silica membrane preparation

Organo-silica membrane was prepared using sol-gel method. TEOS was dissolved into ethanol (EtOH, 99%) under stirring for 5 min at 0°C to avoid the partial hydrolysis reaction. After that, the reflux was started by the addition of diluted citict acid (0.0007 N $C_6H_8O_7$) for 1 h at 50°C. Then, the second reflux of ammonia solution (NH_3, 25%, Merck) diluted in ethanol was added drop wise into sol mixture for 2 h. silica sols was dried in a temperature controlled oven at 60°C for 24 h to obtain the dried gel. The final molar ratios of the TEOS:EtOH:$C_6H_8O_7$:H_2O:NH_3 sol were calculated to be 1:38:0.0007:5:*x*, where *x* was 0.0003 as NH_3. This organo-silica sols was placed in the petri dish and dried in the oven at temperature 60°C for 24 hours. Then, organo-silica xerogel was grounded into powder and calcined in air at 600°C.

The organo-silica thin films were coated on macro porous alumina substrate (α-Al_2O_3 tubular support (Ø≈100 nm), Ceramic Oxide Fabricates, Australia) via dip-coating process with a dwelling time of 2 min, dipping rate of 10 cm min^{-1} and withdrawal rate of 5 cm min^{-1}. Then, the deposited membrane layer was dried and calcined in air at 600 °C. via RTP techniques for 1 h without applying ramping/cooling rates. A process of dip-coating, drying and calcination was repeated for 4 times to get organo-silica membrane 4 layers for desalination application. All membranes were characterized by Scanning Electron Microscopy (SEM JOEL ZEISS) to investigate its morphology and thickness.

2.3. Pervaporation

The membrane was set into a pervaporation set-up to perform in desalination as shown in Figure 1. One of sided membrane was blocked and another one was connected to a cold trap (immersed in liquid nitrogen) and the vacuum pump was operated at 1 bar. The artificial brine water were prepared using (NaCl, Sigma Aldrich) in variance of 7.5, 10 and 15 wt %. Pervaporation was operated at variance temperature of feed in ranging of 40 °C and 60°C which was controlled by thermocouple and hotplate. Homogenous of feed solutions was controlled in constant stirring to prevent the polarization. The water flux, F ($kg.m^{-2}.h^{-1}$), was determined based on the Equation $F = m / (A \cdot \Delta t)$, where m is the mass of permeate (kg) retained in the cold trap, A is the surface-active area (m^2) and Δ is the time measurement (h). The salt rejection, R (%), was calculated as $R = (Cf-Cp) / Cf \times 100\%$, where C_f and C_p are the feed and permeate concentrations of salt (wt %).

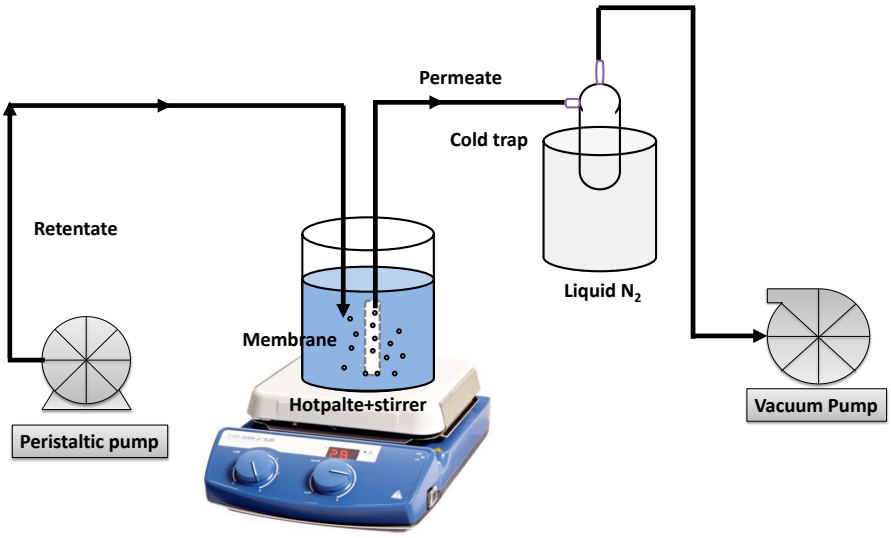

Figure 1. Pervaporation Set Up in Desalination Process.

3. Results and discussion

3.1. SEM characteristics

Before pervaporation test, membrane was characterized by SEM to show interlayer-free membrane thickness and surface (Figure 2 and Figure 3). Organo-silica membrane in this work, shows rough in surface (Figure 2). This is because organo-silica sol infiltration occurs when membrane support dipped into it. Nevertheless, calcination would offer thermal induced. Organo-silica membrane thickness (1 μm) prepared are thicker than previous study around ~470 nm [11] (Figure 3). RTP calcination process is done shorter than the Conventional Thermal Process (CTP) makes imperfect solvent evaporation. Different with CTP, RTP process does not use the heating rate and cooling rate. Despite this, RTP take longer time (more than one week) instead of CTP (1 day) [25].

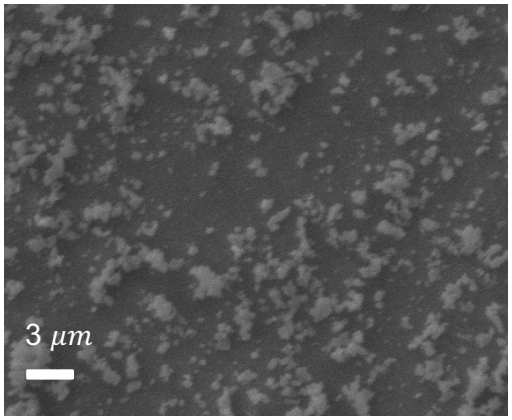

Figure 2. Surface area of interlayer-free organo-silica membrane during calcination in air.

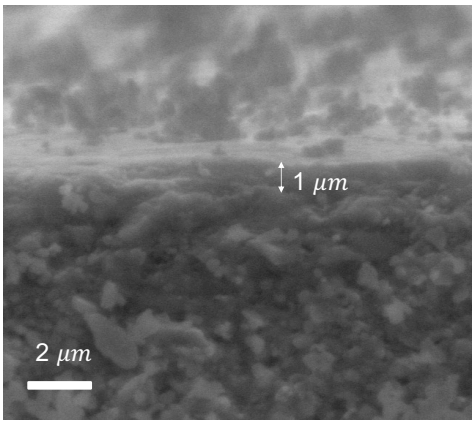

Figure 3. Cross-section area of interlayer-free organo-silica membrane during calcination in air.

3.2. Pervaporation of brine water using organo-silica membranes

The effect of feed temperature on membrane performance relates to water flux. Figure 4 shows water fluxes enhance as feed temperatures increase from 40 to 60 °C. It raise vapour pressure and membrane driving force [26]. Highest temperature delivers high water flux as well. For organo membrane silica with brine water as feed, the highest is 1.55 $kg.m^2.h^{-1}$ Brine water. Rather than using RO,

pervaporation were able to treat brine water without extra osmotic and hydraulic pressure [27]. It reduces energy consumption.

Compare to polymeric membranes, this organo-silica membrane performed better (Table 1). It produce larger water flux of 1.55 $kg.m^2.h^{-1}$ than single acid catalyzed only. It suggests citric acid can maximize the performance. Recent studies show higher water flux compare to our study because organo silica membrane prepared in our work, it was calcined in high temperature (600 °C). In this temperature, carbon has been lost and reduce membrane performance (Table 1). Temperature should be considered first. In sol gel method, catalyst give impact to silica matrix, which alkoxysilane come through hydrolysis and condensation reactions [28]. Mesoporous structure will be formed in the presence of acid base dual catalyst. Apply acid catalyst only would make membrane pore smaller. Small pore which is known as micropore mostly used in gas separation and it does not suit for pervaporation due to lower water flux. Whereas base catalyst addition give larger water flux, but poor rejection. Bottle neck pore appear with combination of acid and base. This type of pore, would allow H_2O (2.6 Å) easily diffuse and bigger molecules such as Na^+ (7.16 Å) and Cl^- (6.64 Å) will be hindered on bottle neck pore. But still provide high water fluxes [11].

In addition, in this work, carbon chain from citric acid resist pore collapsed in silica matrix. It creates stronger membrane structure and higher hydrostabilty [29], Rangelova, et al. [30]. Citric acid reportedly provides mesoporous structure. This work supports the benefit of carbon effect which agree in similar carbon templated researches for pectin [10, 31] and P123, where give less effect of hydrated ion retention on membrane surface.

Despite increasing water fluxes, salt rejection slightly reduces until 90% after organo-silica membrane treat on higher salt concentration (15 wt%) for both temperatures. Salt polarization occurs in this case. It happen when salt particles accumulated on membrane surface. This phenomena will block the membrane pores in period of time. As a result, membrane will produce lower water fluxes because a way [11]. As explain before some salt particles might be detained on bottle neck pore. Beside it, raising temperature also reduces salt rejection 38.5% for all brine concentration.

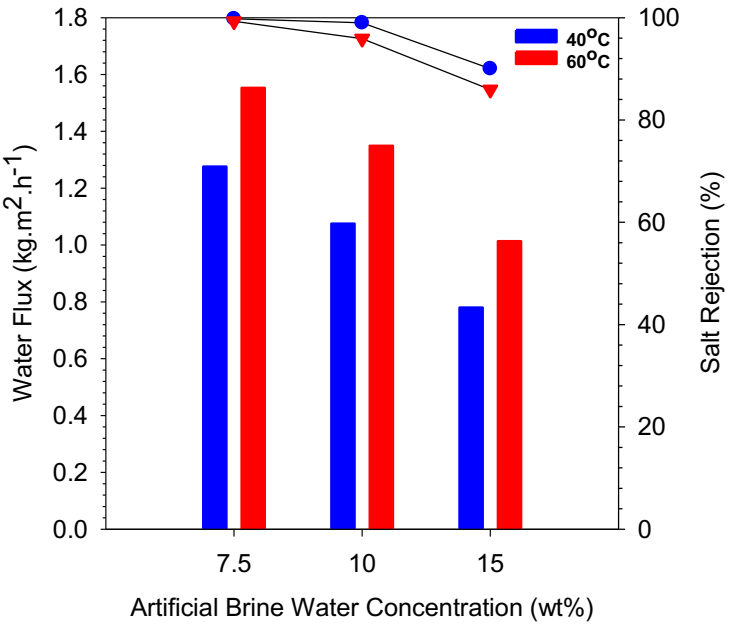

Figure 4. Performance of interlayer-free organo-silica membrane for brine water pervaporation.

Table 1. Comparison of membrane performance for pervaporation in different catalyst, feed temperature, and feed.

Membrane type	Catalyst	Feed temperature (℃)	Feed	Water fluxes ($kg.m^2.h^{-1}$)	Salt rejection (%)	Ref
Organo-silica membrane	Acid-base	40-60	NaCl 3.5%	1.55	99	This work
Polyelectrolyte	Acid	58.5	Ethanol/water	0.5	-	[32]
Polyetheramide	-	68-70	NaCl 32 g/L	0.56	99.99	[33]
Silica-Pectin	Acid-base	25	Wetland saline water	3.4	99.99	[10]
Silica-P123	Acid-base	25	NaCl 7.5wt%	6.6	99.5	[13]

4. Conclusion

From the experimental results, the operating parameter in pervaporation which is feed temperature can increasing water flux from 40 ℃ (1.27 $kg.m^2.h^{-1}$) to 60 ℃ (1.55 $kg.m^2.h^{-1}$). Organo –silica membranes give salt rejection of >90%. It proves this membrane is suitable for treat high concentration salt. Pervaporation may reduce operation cost. In addition, membrane prepared with citric acid would make membrane performance more robust when it applied in pervaporation processes.

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