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CHEMICAL CLEANING OF MICROFILTRATION CERAMIC MEMBRANE FOULED BY NOM

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Abstract

Microfiltration membrane made from Sayong ball clay by using uniaxial dry compaction method was used to treat natural organic matter (NOM) source water. A sintering temperature of 900 °C to 1000 °C were applied. The effect of sintering temperature on membrane porosity, strength and water flux were identified. The porosity of the membrane decreased with increasing sintering temperature and the strength and flux increased with temperature. The membrane was subjected to NOM filtration experiments. The results showed an improvement to the quality of permeate water, where there is a reduction in COD, TSS, BOD_5, turbidity, hardness and salinity; and an increased pH value. The effect of chemical cleaning on the fouled membrane also was studied. After cleaning with NaOH solution, a high flux recovery was achieved (up to 50% from the initial pure water flux). The degree of cleanliness of fouled membranes after chemical cleaning was further observed with SEM and EDX analysis.

Keywords: Ceramic membrane, microfiltration, NOM, flux recovery, sintering temperature

Abstrak

Membran penapisan mikro diperbuat daripada bebola tanah liat Sayong menggunakan kaedah penekanan kering telah digunakan untuk merawat air yang mengandungi bahan organik semulajadi. Suhu pembakaran 900 °C sehingga 1100 °C di kenakan. Kesan suhu pembakaran terhadap keliangan membran, kekuatan dan fluk air dikenal pasti. Jumlah keliangan membran berkurangan dengan peningkatan suhu pembakaran dan kekuatan dan fluks bertambah selari dengan peningkatan suhu. Ujikaji penapisan membran mengunakan air mengandungi bahan organik semulajadi telah dijalankan. Hasil ujikaji menunjukkan bahawa kualiti air yang ditapis semakin meningkat, di mana terdapat pengurangan nilai bacaan permintaan oksigen kimia (COD), pepejal terampai (TSS), permintaan oksigen biokimia (BOD_5), kekeruhan, kekerasan dan kemasinan air; dan peningkatan nilai pH. Selain itu, kesan pembersihan kimia terhadap membran yang tercemar dikaji. Setelah dicuci menggunakan cecair NaOH, pemulihan fluk yang tinggi diperolehi (sehingga 50% daripada nilai awal fluks air suling). Kadar kebersihan membran yang tercemar selepas pembersihan kimia diperhati lebih lanjut menggunakan SEM dan EDX.

Kata kunci: Membran seramik, penapisan mikro, NOM, pemulihan fluks, suhu pembakaran

1.0 INTRODUCTION

Recently, many researches on ceramic membranes were done, owing to its excellent properties, such as good mechanical and thermal stability [1]. Ceramic membranes can be applied in diverse applications, including food, beverage, biotechnological, pharmaceutical and electronic industry. Ceramic materials commonly used in membrane fabrication are alumina, silica, zirconia, kaolin, clay, fly ash and etc. Although alumina, silica and zirconia have superior properties as a membrane material, their expensive raw materials and processes are the main disadvantages. To overcome the drawback, many researches were done with cheaper materials, especially the naturally abundant ceramic clay. Sayong ball clay is a kaolinitic sedimentary clay of natural origin and also known as secondary clay. Sayong ball clay used in this work is originated from Sayong, in Perak. It is commonly used in labu sayong production, a black gourd-shaped clay used to contain drinking water.

Microfiltration (MF) is a low pressure membrane process that is commonly used in water treatment for the past two decades [2]. MF is suitable in suspended solids removal, especially bacteria and algae. However, dissolved contaminants like natural organic matter (NOM) are hardly removed by microfiltration. According to Rook [3], NOM is a well-established cause of colour in water and is also linked to disinfection by-products formation of with potential health implications. NOM is the main contributor to membrane fouling for surface waters and some ground-waters [4].

One of the main issues affecting the membrane performance is fouling. Fouling is a deposition of microorganisms, colloids, solutes, and cell debris within or on the membranes [5]. This fouling will deteriorate the membrane performance by reducing flux. Membrane fouling caused by NOM is affected by several factors, including feed constituents properties (size, hydrophobicity, charge density and isoelectric point), membrane properties (hydrophobicity, charge density, surface roughness and porosity), solution properties (pH, ionic strength and concentrations of metal) and the hydrodynamics of the membrane system [4, 6]. NOM components were found to be composed of hydrophobic acids, hydrophobic neutrals and hydrophilic neutral or bases [7].

Membrane fouling by NOM can be minimised by pre-treatment of feed water, hydrodynamic cleaning, and optimisation of chemical and operational conditions, modification of the membrane surface and backwashing and flushing [8-11]. Among these methods, chemical cleaning are commonly used for cleaning and regenerating to restore membrane performance. Chemical cleaning able to restore the reversible flux decline cause by NOM fouling [12]. Caroll *et al*., [13] found that coagulation pre-treatment before filtration of raw water improves NOM removal and reduces membrane fouling. Chemical cleaning can be done with various types of cleaning agent, such as acid (H_3PO_4, citric acid), alkali (NaOH), detergents (alkaline, non-ionic) and others. Alkaline cleaning agent is usually used for organic foulant removal. Zondervan & Roffel [14] studied the cleaning model of different cleaning agent on ultrafiltration (UF) membrane. They found that the membrane is fouled by water that have high organic content and was effectively cleaned by using caustic and oxidising cleaning agents. Lee *et al*. [7] found that membrane fouled with hydrophobic NOM water cleaned with NaOH has high flux recovery (64.8 %) than NaCl (55 %), citric acid (49.7 %) and SDS (29.3 %). However, for hydrophilic NOM water, only 18.9 % flux was recovered with NaOH as compared to NaCl (43.3%).

Several methods can be used to measure membrane degree of cleanliness after undergoing a cleaning process. These methods include flux measurement, AFM, FTIR, zeta potential, EDX and magnetic resonance imaging (MRI) [11, 15-18]. Efficiency of membrane cleanliness after chemical cleaning is usually evaluated by flux measurement [7, 13, 19-21]. Lee *et al*., [19] reported that the water flux increases after back flushing and that toluene is the most effective chemical cleaning agent for removing waste oils with flux recovery of 84% as compared to the initial flux. However, water flux measurement cannot accurately guarantee the degree of cleanliness because there may be residual foulant deposits between the pores or inside the coarse support of asymmetric microporous membranes without disturbing the flux. Besides, there is a possibility of bacterial growth on the membrane, which is not concerned by flux measurement [15, 21].

The objective of this work is to study the performance of ball clay membrane during filtration of lake water at different transmembrane pressure. In addition, the effectiveness of chemical cleaning by NaOH is also analysed through flux measurement. The cleaning efficiency is estimated by flux recovery identification, followed by SEM and EDX analysis to observe the membrane surface.

2.0 METHODOLOGY

2.1 Membrane Material

The initiating material used in the membrane fabrication is Sayong ball clay from Perak, Malaysia. The clay was crushed into powder form and sieved through a 75 µm mesh standard screen. Sayong ball clay consists mainly of SiO_2 (74.3%) and Al_2O_3 (19.6%), followed by Na_2O (2.4%), K_2O (1.22%), MgO (1.09%) and CaO (1.08%). Corn starch was milled to clay powder with ratio of 30:70 weight percentage and acted as a pore former. Flat disc membranes with dimension of 2.5 mm thickness and 3 mm diameter were fabricated through a compaction method by using universal testing machine (Instron 5982) at a pressure of 200 MPa. The green body was then sintered at temperatures from 900°C to 1100°C with holding time of 2 hours, by using a laboratory high temperature furnace (Nabertherm). The performance of the fabricated membranes before and after cleaning was compared with commercial microfiltration ceramic membrane available in industry.

2.2 Characterisation Techniques

The porosity of the membrane is measured according to Archimedes principle with an immersion medium of distilled water, following the ASTM C373-88 standard. The 3-point bending test to determine the mechanical strength of membrane was done by using a Universal testing machine (Instron 5982). The testing was done by following the ASTM C-1161-02c standard with a span of 40 mm and crosshead speed of 0.5 mm/min.

The effectiveness of chemical cleaning was accessed using a table top microscope (TM3000, Hitachi) with the energy dispersive x-ray spectroscopy (EDS) unit (Quantax70, Hitachi). The membranes cleaned with chemical were analysed with this equipment together with a fouled membrane for comparison.

2.3 Filtration Test and Chemical Cleaning

The filtration tests were performed by using a laboratory size filtration system, as shown in Figure 1. Different pressures were applied during the filtration process, which were from 0 bars to 3 bars. The filtration process was performed at 25°C temperature. The water source was taken from UTM lake water in Johor, Malaysia. Each fouling and cleaning process was done through five steps: initial pure water flux measurement, fouling, rinsing, NaOH cleaning and final pure water flux measurement.

The initial and final pure water flux was measured by using distilled water. The membrane was then fouled with lake water at a pressure of 1 bar for 2 hours. The flux reduction within these 2 hours was observed. Then the NOM-fouled membranes were removed from the filtration system and rinsed with distilled water to remove loose bound particles on the membrane surface.

Chemical cleaning was carried out by soaking the fouled membrane in 0.1 M NaOH solution for 24 hours to desorb the NOM foulants [7]. Lastly, the final pure water flux of the cleaned membrane was measured to calculate flux recovery to estimate the cleaning efficiency. Water flux recovery was assessed by comparing the flux of virgin membrane (Jo) with the flux after cleaning (Jc); measured under the same condition. The percentage of water flux recovery (FR) can be expressed as: FR = (Jc/Jo) x 100% [15].

2.5 Water Analysis

Quality of the filtered water, which includes pH, chemical oxygen demand (COD), biochemical oxygen demand (BOD_5), total suspended solid (TSS), turbidity, hardness and salinity, were determined. COD analysis was conducted by using DR4000 spectrophotometer machine through a digestion reactor method to measure the chemical content of the waste water. On the other hand, BOD_5 analysis was done to determine the changes in the dissolved oxygen in waste water after filtration. The total suspended solid analysis will detect the amount of the remaining small solid particles in the water after filtration process. The degree of clarity of the filtered water was measured by turbidity in Nephelometric Turbidity Units (NTU).

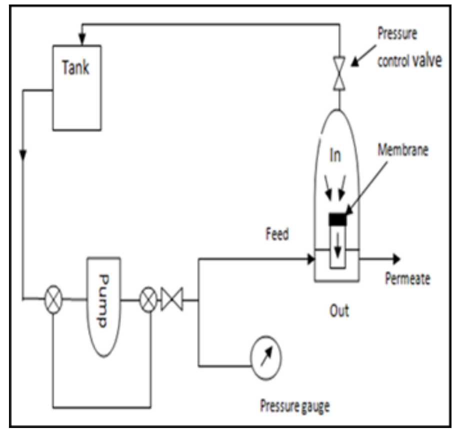


Figure 1 Schematic diagram of water filtration system

3.0 RESULT AND DISCUSSION

3.1 Properties of Membrane

The properties and performance of the ball clay membranes were compared with commercial membranes available in the market. The properties of both types of membranes are shown in Table 1. The apparent porosity of ball clay membranes was reduced from 40% to 0.07% at temperatures of 900°C to 1100°C. The decrease in porosity was due to the densification of the porous structure during sintering. More consolidated structures were formed at a higher sintering temperature that reduced the number of open pores. The strength of the ball clay membrane was inversely proportional to the porosity value. However, the strength value increased with increasing temperature, owing to densification and growth of the grain boundaries [22-23]. The pure water flux value increased as the sintering temperature was increased, but inversely proportional to porosity. This may be due to the increased pore size at a higher sintering temperature. Large pores facilitate water filtration process, but do not contribute to the porosity percentage since it may be close pores rather than open pores.

3.2 Effect of Transmembrane Pressure (TMP) on Flux

The variation of fluxes with TMP between 0 bars to 3 bars is shown in Figure 2. Overall, the initial pure water flux was higher than the flux of lake water. However, both showed an increase in flux value with increasing TMP. The initial pure water flux showed a large increase at higher TMP while the lake water fluxes increased slowly. Similar result was also found by others [23-28]. Bhave [29] noticed that the flux at different pressures is influenced by the presence of particulates which favour the formation of concentration polarisation layer. This layer will cause an increase in resistance to solvent transport across the membrane, and hence limiting the flux.

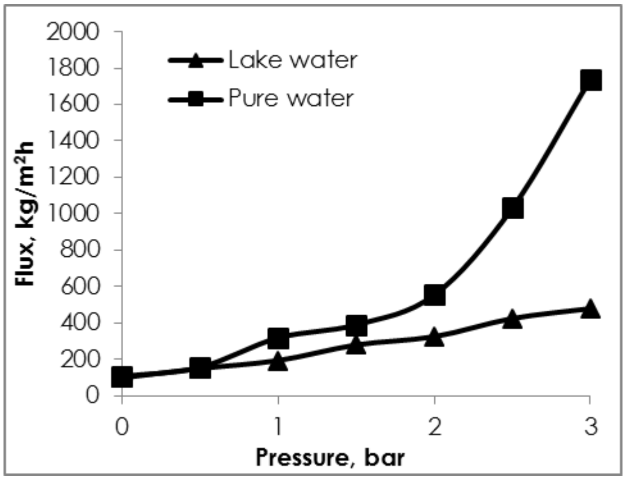


Figure 2 Variation of initial pure and lake water flux at different transmembrane pressure during filtration process

3.2 Effect of Sintering Temperature on Flux

Sintering temperature during membrane preparation also affected the water flux value. As can be seen in Figure 3, the initial pure and water flux both increased with increasing sintering temperature of up to 1050 °C due to the increase in membrane pore diameter. The flux was then reduced at 1100°C. This may be related to the reducing porosity as well as the presence of closed pores at 1100 °C, which was not favourable in the filtration process. This result is in good agreement with other researchers [23, 24 and 30]. According to Sarkar *et al*. [24], the reduction of flux at higher sintering temperature is caused by poorer pore connectivity which results from smaller pore size. Wang *et al*. [30] concluded that the flux increased with increasing temperature was caused by the increment of pore size while the decrease of pore length resulted in a sharp reduction in the flow resistance. On the other hand, membranes with larger pores are easier to be blocked, causing a rapid reduction of flux and higher fouling extent [31]. Overall, the value of pure water flux is always higher than lake water flux.

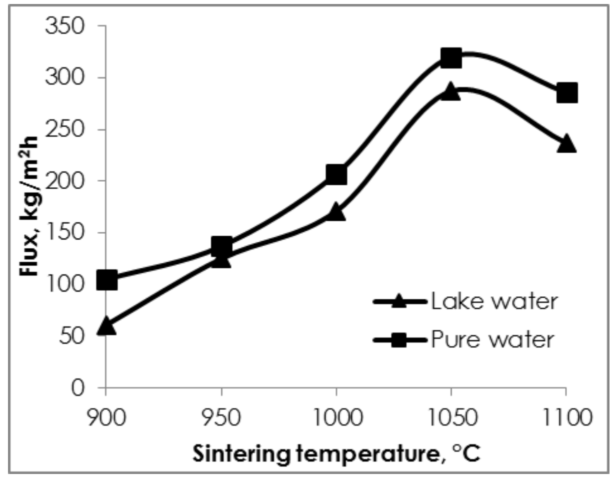


Figure 3 Effect of sintering temperature on the fluxes

3.4 Membrane Performance after NaOH Cleaning

Typical flux declined within 2 hours during the microfiltration of lake water, such as shown in Figure 4 for the ball clay membrane, before and after cleaning and as compared to the flux of lake water filter with commercial membrane. The experiment was done with ball clay membrane sintered at 1050°C and at temperature of 25°C and TMP of 1 bar throughout the filtration process. As expected, the flux rate improved 1.8 times after chemical cleaning with 0.1M NaOH. However, the membrane before and after cleaning showed a rapid flux decline over time at the first 15 min filtration. After 1hour of filtration time, the flux value was nearly constant. The rapid flux decline occurred when the membrane pores were clogged and fouled by particulates in lake water [19].

According to Jana *et al*. [32], the flux decrease during the first phase was due to the membrane fouling while the second phase was due to concentration polarisation. The commercial membranes still showed higher flux than ball clay membrane since their porosity are higher (62%). Nevertheless, the flux continues to decrease over time. Several authors also reported similar results for flux over time [27, 32- 33]. The maximum flux recovery achieved after cleaning with NaOH was 50%.

Table 1 Properties of ball clay and commercial ceramic membrane

Properties	Porosity (%)	Strength (MPa)	Cross-sectional area (m²)	Pure water flux (kg/m²h)
Clay membrane				
900 °C	40	1.05	0.7069x10-3	105
950 °C	37	1.37	0.7069x10-3	137
1000 °C	31	2.29	0.7069x10-3	207
1050 °C	17	5.48	0.7069x10-3	320
1100 °C	5	17.15	0.7069x10-3	286
MF Commercial membrane	62	5.48	0.03	833

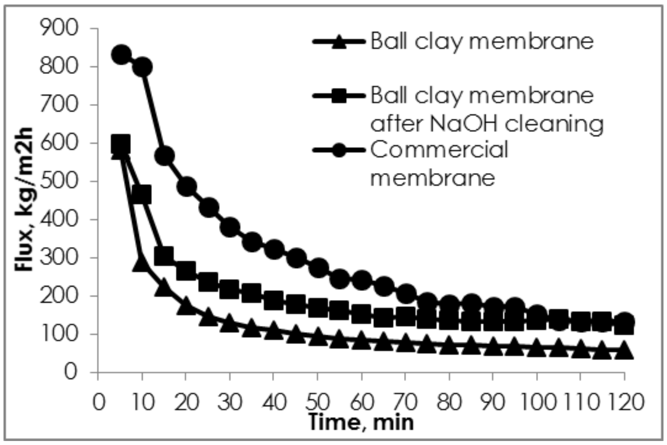


Figure 4 Flux evaluation in function of time for ball clay membrane before and after NaOH cleaning compared with commercial membrane

3.5 SEM/EDX Analyses on the Membrane Surface

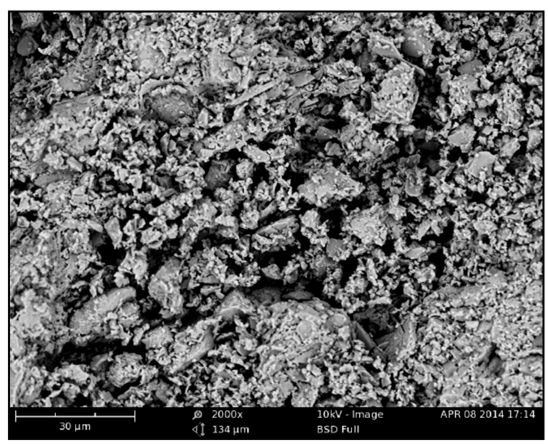
Figure 5 shows the SEM of virgin, fouled and cleaned ball clay membrane sintered at 1050°C. As compared to the virgin membrane (Figure 5(a)), some impurities were observed on the fracture surface of the fouled membranes, as shown in Figure 5(b). After the cleaning process, the impurities were completely removed (Figure 5(c)). These results were further confirmed with EDX analysis on the virgin, fouled and cleaned ball clay membrane (Figure 6). In the fouled membrane, iron was present (Figure 6(b)). Originally, the iron peak were absent in the virgin ball clay membrane (Figure 6(a)). According to Wallberg *et al*. [18], iron is one of the substances that are difficult to remove by cleaning. However, after cleaning with NaOH, the iron peak was completely removed, as shown in Figure 6(c).

3.6 Water Quality Analysis

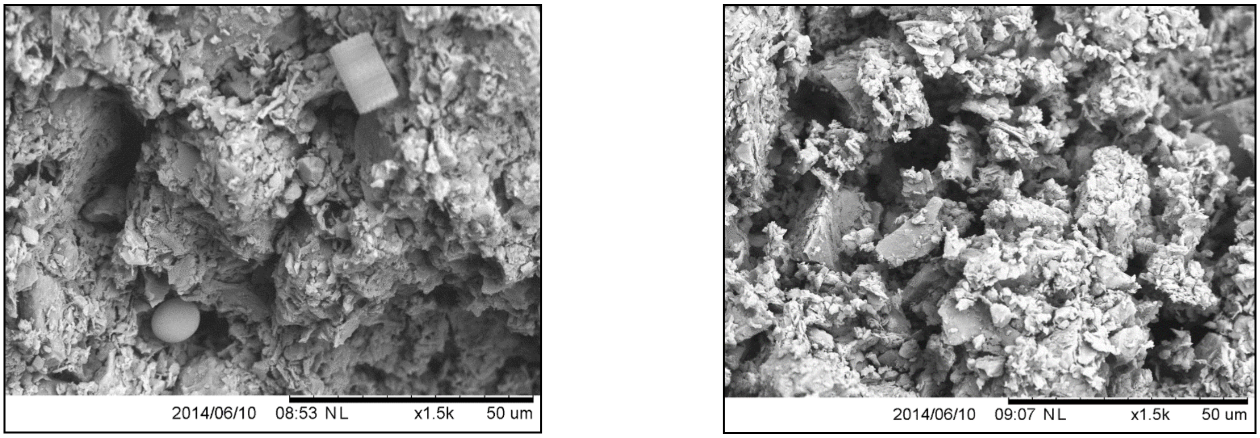
Table 2 shows the result for water quality analysis of lake water, before and after filtration with ball clay membrane. The results obtained were compared with the quality of lake water filtered with a commercial membrane from the industry. As can be seen in the results, the overall quality of the water was improved after being filtered with ball clay membranes and gave better results than the water filtered with a commercial membrane.

As shown in Figure 7, the COD value maximally reduced up to 38% by ball clay membrane, slightly higher than commercial membrane. This result indicated that the use of microfiltration membrane was sufficient in removing pollution from waste water [28]. Almost 99.9 % of suspended solid from lake water was removed by ball clay membranes, which showed that the elimination of colloidal particles or suspended solid was successful.

The BOD_5 of the lake water was reduced up to 50% with ball clay membranes. This showed that these ball clay membranes were able to remove some organic matter dissolved in the lake water. This result was much better than with commercial membrane, where it can only reduce the BOD_5 value to about 11%.

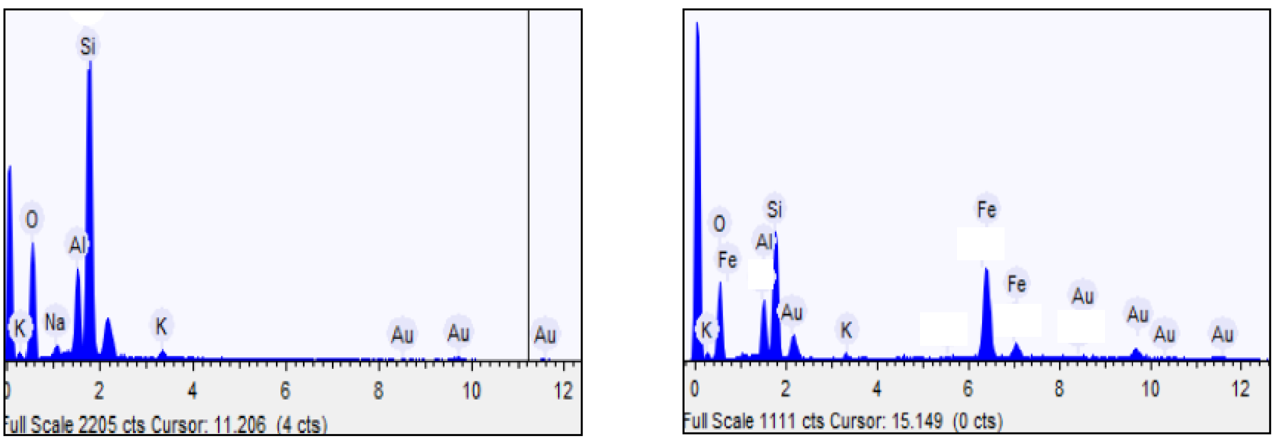


(a)

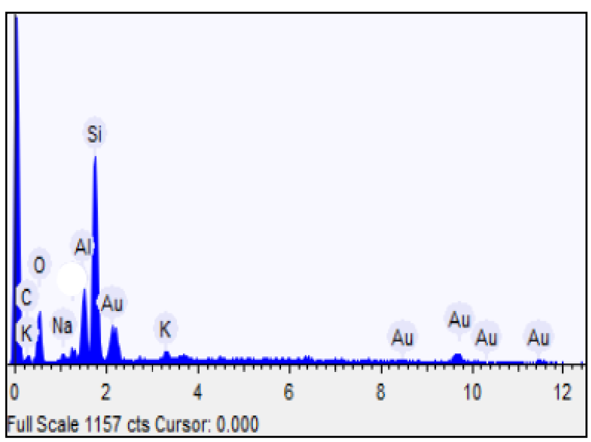


(b) (c)

Figure 5 SEM of the fracture surface of a (a) virgin ball clay membrane, (b) ball clay membrane after fouling, and (c) ball clay membrane after cleaning



(a) (b)



(c)

Figure 6 EDX analysis of the fracture surface of a (a) virgin ball clay membrane, (b) fouled ball clay membrane, and (c) cleaned ball clay membrane

Table 2 Water quality data for lake water before and after filtration with ball clay and commercial membrane

Membrane Type	COD (mg/l)	TSS (mg/l)	BOD_5 (mg/l)	pH	Turbidity (NTU)	Hardness (mg/l)	Salinity (mg/l)
Ball Clay Membrane							
900 °C	5	0.0096	3	7.15	1	22	1.75
950 °C	5	0.012	3	6.94	2	24	1.98
1000 °C	5	0.0156	3	7.04	2	25	2.09
1050 °C	5	0.038	2.7	6.37	2	27	2.79
1100 °C	6	0.0427	2.5	6.43	2	27	3.17
MF Commercial Membrane	**6**	**0.29**	**5**	**6.27**	**4**	**24**	**3.49**
Raw lake water	**8**	**26**	**5.6**	**5.92**	**6**	**34**	**3.99**

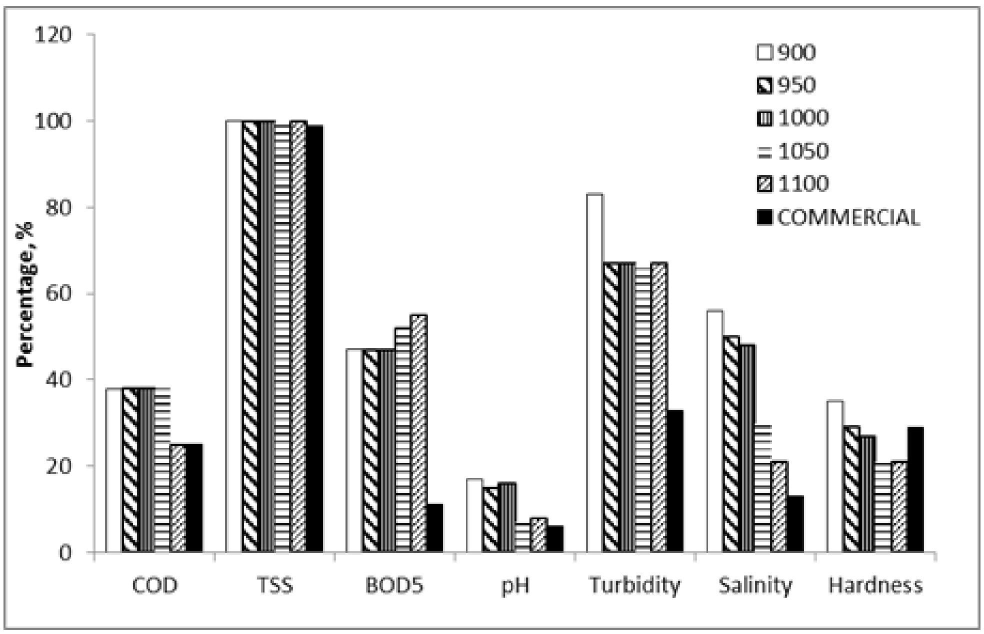


Figure 7 Percentage of reduction of water quality of lake water filter with ball clay and commercial membranes

4.0 CONCLUSION

A ball clay membrane was fabricated by using compaction method and sintered at different sintering temperatures. The porosity was reduced while the strength increased with increasing sintering temperatures. Both fluxes of pure water and lake water increased when the transmembrane pressure was larger. However, the presence of suspended particles in the lake water caused the flux to be lower than pure water flux. When comparing the fluxes of membranes sintered at different sintering temperatures, the fluxes were higher for the membrane of higher sintering temperature. At 1100°C membrane, the flux was then reduced slightly since more closed pore were present than opened pores. The flux was also observed as a function of time for 2 hours. As expected, the flux was reduced as the time increased to an almost constant state at the end. After chemical cleaning, the flux improved and the achieved flux recovery was up to 50%. This showed that NaOH was able to remove the foulant from the membrane surface. This result was further confirmed with SEM and EDX analyses. Iron peak was detected on the foulant membrane surface. After the cleaning process, the iron peak was completely removed. The quality of the lake water was also improved after the chemical cleaning process. Overall, the lake water, filtered with ball clay membrane and sintered at 900°C, produced the most clean water with 5 mg/l of COD, 0.0096 mg/l of TSS, 3 mg/l of BOD_5, pH of 7.15, 1 NTU of turbidity, 22 mg/l of hardness and 1.75 mg/l salinity.

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