

# Application of FO-MBR System to Utilize RO Concentrate as Draw Solution

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## Abstract

Reverse osmosis (RO) concentrate was utilized to produce high quality effluent from MBR system. High energy requirement and severe membrane fouling are the two main problems in the operation of MBR. This study investigated the feasibility of incorporating forward osmosis (FO) in wastewater treatment. The RO concentrate was used as draw solution in a closed-loop FO-MBR system. Permeate flux, draw solution (DS) conductivity, nutrient removal and membrane fouling were among the parameters checked for the evaluation of effluent quality. The flat sheet FO membrane with an effective membrane area of 0.03 m<sup>2</sup> was submerged in the oxic reactor of MBR. Different draw solution concentrations (35, 50, 100, 150 and 200 g/L) were used to investigate the suitable concentration for the operation of FO-MBR. Results showed that increasing the concentration of draw solution, increases permeate flux, which dilutes the RO concentrate that can be fed back to the RO system. Increase in the DS concentration increases the change in conductivity, and the same trend was observed with the increase in recirculation flowrate. The effluent quality of FO was better than the MBR when MF membrane was used. This suggests the consistency of water quality as compared to the conventional MBR effluent. The percent removal of TP, TN and COD reached 93.75%, 89.83% and 96.43%, respectively, which is more suitable as feed for RO process. In addition, membrane fouling was evident in MF membrane after the continuous operation. Further studies are needed, involving the integration of FO-MBR and RO systems, to evaluate the effectiveness of FO-MBR permeate to the recovery and performance of RO system.

## Introduction

Membrane bioreactor (MBR) is an attractive process for wastewater treatment due to its high effluent quality when operated on its optimized condition. The permeate from the MBR could be used as feed in RO system. The combination of membrane bioreactor coupled with reverse osmosis (MBR-RO) for water treatment has been successfully applied for the treatment of raw sewage, secondary effluent and municipal wastewater to produce reclaimed water [1-5]. However, some contaminants, such as organic matter and ions, which are not efficiently removed from the MBR system, could cause severe fouling on the RO membrane [6]. Several studies were conducted demonstrating the occurrence of organic and biological fouling, which remains a major challenge in reverse osmosis systems [7-10]. The deposited organic substances may further initiate biological fouling when microorganisms start to colonize the organic layer then multiply by feeding on biodegradable nutrients (C, P, N) from the feedwater and excreting more organic substances, leading to a significant build-up of biofilm. Moreover, the deposited sticky organic substances may enhance the deposition of more colloidal particles to the membrane and further aggravate the fouling [11]. In addition to membrane fouling, the concentrated brine produced from the RO system is a huge challenge for waste disposal [12]. Thus, utilization of RO concentrate should further be explored.

The objective of this study is to enhance the effluent quality of Forward Osmosis – Membrane Bioreactor (FO-MBR) system utilizing RO brine as draw solution. Batch tests were conducted to determine the relationship between the operating flux and the conductivity of the draw solution. The system was operated in a continuous mode, and nutrient removal as well as membrane fouling for both MF and FO membranes were analyzed and evaluated.

## Materials and Methods

### *Batch test*

The lab-scale FO-MBR system was operated in a closed loop mode. RO concentrate was used as draw solution. The RO concentrate was flowing on the permeate side while the active layer side was facing to the mixed liquor of the oxic reactor. Water flux and DS conductivity were measured at different draw solution concentrations to determine the most suitable DS concentration for the operation of FO-MBR. The change in salinity of the feed and the change in permeate flux are the two parameters that were monitored and evaluated. To investigate the effect of flowrate on the conductivity of draw solution, RO concentrate was recirculated with flowrates of 0.1, 0.2 and 0.3 L/min with constant temperature of 25 °C.

### *Continuous experiment*

The laboratory scale FO-MBR system is composed of two parts: (1) the membrane bioreactor with MF membrane treating wastewater, and (2) the closed-loop FO with RO concentrate as draw solution.

Concentration of 200 g/L NaCl was used as the initial DS concentration, and was replenished every 4 days. The flowrate of the closed-loop FO system was maintained to 0.2 L/min with constant temperature of 25 °C. AND GF-4000 analytical balance recorded the change in mass of the draw solution, which was used in the calculation of permeate flux.

Simultaneous filtration by MF and FO was performed, and samples were taken every other day, which was monitored for a month. Operating conditions for MF and RO were not similar due to their respective mechanisms in producing effluent. MF-MBR was operated through suction while FO-MBR generated effluent through osmotic gradient. The essential parameter similar for the two systems was the influent characteristic. Both MF and FO membranes were submerged in bioreactor having similar characteristics of sludge as well as nutrient content. Thus, comparison of the effluent qualities for both MF and FO were conducted, in terms of COD, TN and TP removals. The schematic diagram of the lab-scale FO-MBR system is illustrated in Figure 1. The MBR system used in the experiment was based on the optimization made by Sibag and the operating condition was summarized in Table 1 [13]. The synthetic wastewater used as feed in MBR was prepared by mixing the following chemicals: glucose (750 mg/L as COD),  $\text{NH}_4\text{HCO}_3$  (37.5 mg/L as TN),  $\text{KH}_2\text{PO}_4$  (5.5 mg/L as  $\text{TP-PO}_4^{3-}$ ), 300 mg/L  $\text{NaHCO}_3$ , 180 mg/L  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ , 28 mg/L  $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ , and 0.6 mL mineral salts solution. The mineral salts solution contained 1.5 g  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ , 0.15 g  $\text{H}_3\text{BO}_3$ , 0.03 g  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ , 0.18 g KI, 0.12 g  $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ , 0.06 g  $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$ , 0.12 g  $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ , 0.15 g  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ , and 10 g EDTA dissolved to 1 L deionized water.

### *Membranes*

PVDF flat sheet membrane from TORAY with pore size of 0.08  $\mu\text{m}$  and an effective filtration area of 0.03  $\text{m}^2$  was used in the MF-MBR experiment. The OsMem<sup>TM</sup> CTA-NW membrane from Hydration Technology Innovations (HTI) was used in the FO-MBR experiment. This FO membrane has a fouling resistant feature and casted on a weldable nonwoven support. The maximum operating temperature is 71 °C and the pH range is 3 to 8 as provided by HTI. The effective area of the FO membrane is 0.03  $\text{m}^2$ .

### Analytical methods

To compare the fouling propensity of both MF and FO membranes, fouled membranes were subjected to scanning electron microscopy (SEM) analysis. TN, TP and COD of the effluent were measured using HACH DR-2800. Thermo Scientific Orion Star A222 measured the conductivity of the draw solution.

Table 1. Operating condition of MBR system.

Parameter	Target Value
Total Volume (L)	10
Hydraulic Retention Time (hr)	10
Total Filtration Volume (L/day)	4.3
Returned Sludge (%Inf. Flowrate)	
reactor 1	100
reactor 2	100
Permeate Flux (LMH)	6

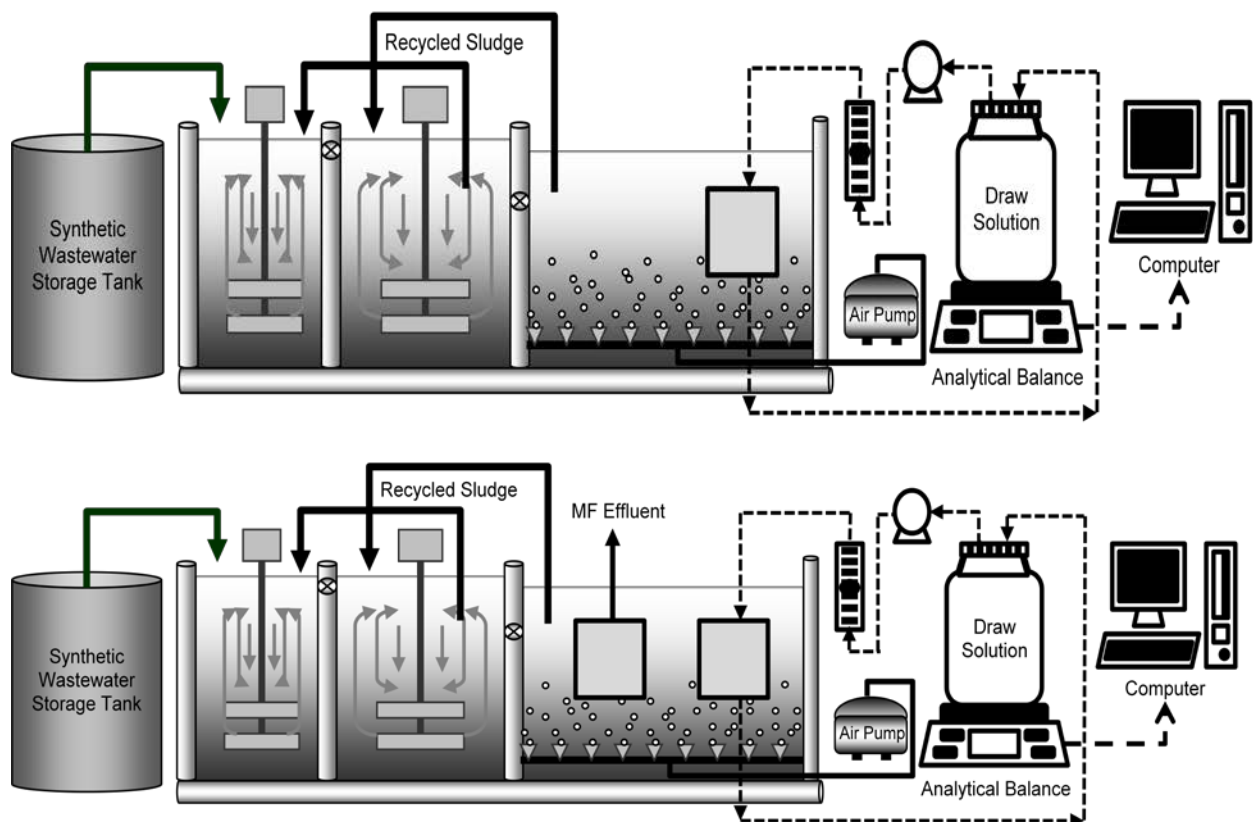


Fig. 1. Labscale FO-MBR systems used in batch mode (Top) and continuous mode (Bottom).

## Results and Discussion

### *Batch experiment at different DS concentrations*

The batch experiment was conducted by recirculating the draw solution in a flat sheet FO membrane submerged in MBR. Five concentrations of draw solution were subjected to batch test: 35, 50, 100, 150 and 200 g/L; and each test lasted for 60 minutes. The first parameter that was measured was the flux of each DS concentration. The generated scattered plot of each representative draw solution was presented in Figure 2. As the concentration of the draw solution increases, permeate flux also increases. The highest flux of 6.65 LMH was obtained by DS concentration of 200 g/L. It was also observed that the fluxes of 200 g/L and 150 g/L were nearly the same. To further visualize the differences in fluxes among the tested DS concentrations, average fluxes were generated. Figure 3 clearly illustrates the disparity among DS concentrations under study. This indicates that the flux difference between lower concentrations was higher than the difference between higher concentrations. It was proven that high DS concentration yielded the highest permeate flux, but the change in flux per DS concentration should not be neglected. It is also an important parameter in evaluating and selecting the suitable operating concentration for the draw solution. Extending the graph gives an unchanged or constant flux, even in increasing DS concentration.

The change in conductivity of the draw solution was also monitored. As shown in Figure 4, negative slope was observed after plotting the conductivity with respect to time, an indicator that filtration is occurring across the membrane. The water that permeated passed the membrane diluted the draw solution, which resulted to the decrease in conductivity. The change in conductivity with respect to time,  $\Delta C/\Delta T$ , was calculated and summarized in Table 2. Increasing the DS concentration increases the change in conductivity. However, it was observed that 200 g/L has lower  $\Delta C/\Delta T$  compared to 150 g/L. Highest rate of change in conductivity was attained by DS concentration of 150 g/L, which shows the highest amount of water that passed through the membrane. This observation was supported by the previous graph illustrated in Figure 3. Higher slope can be generated between DS concentrations of 150 g/L and 100 g/L as compared to the slope of the DS concentrations between 150 g/L and 200g/L. Also listed in Table 2 are the values of  $\Delta C/\Delta T$  at different operating flowrates. It was found that increasing the operating flowrate increases  $\Delta C/\Delta T$ , but then again, at higher flowrates, the value of  $\Delta C/\Delta T$  decreases. Thus 0.2 L/min was selected as the operating flowrate for the continuous experiment.

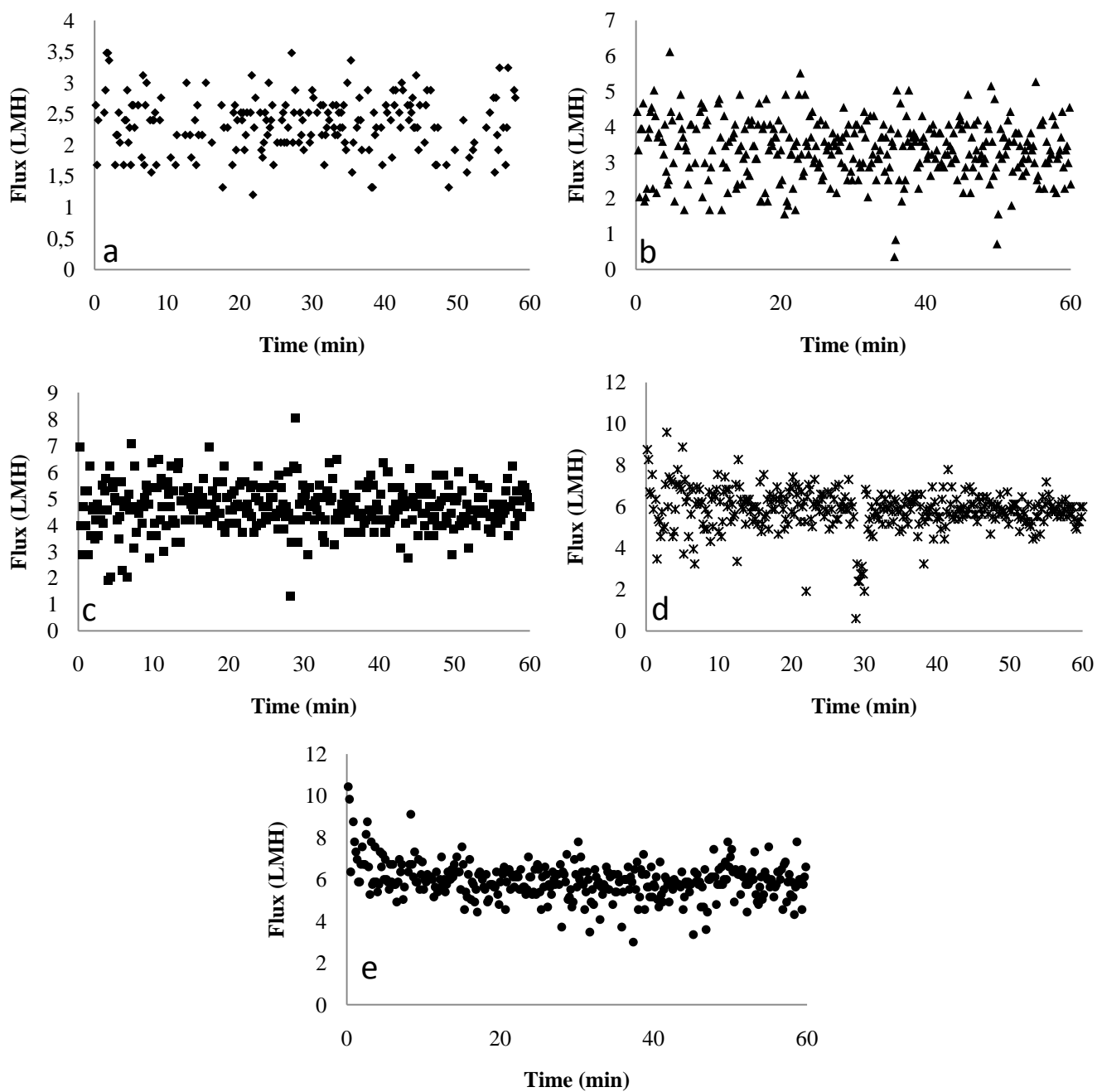


Fig. 2. Determination of flux at different DS concentrations: (a) 35 g/L; (b) 50 g/L; (c) 100 g/L; (d) 150 g/L; and (e) 200 g/L.

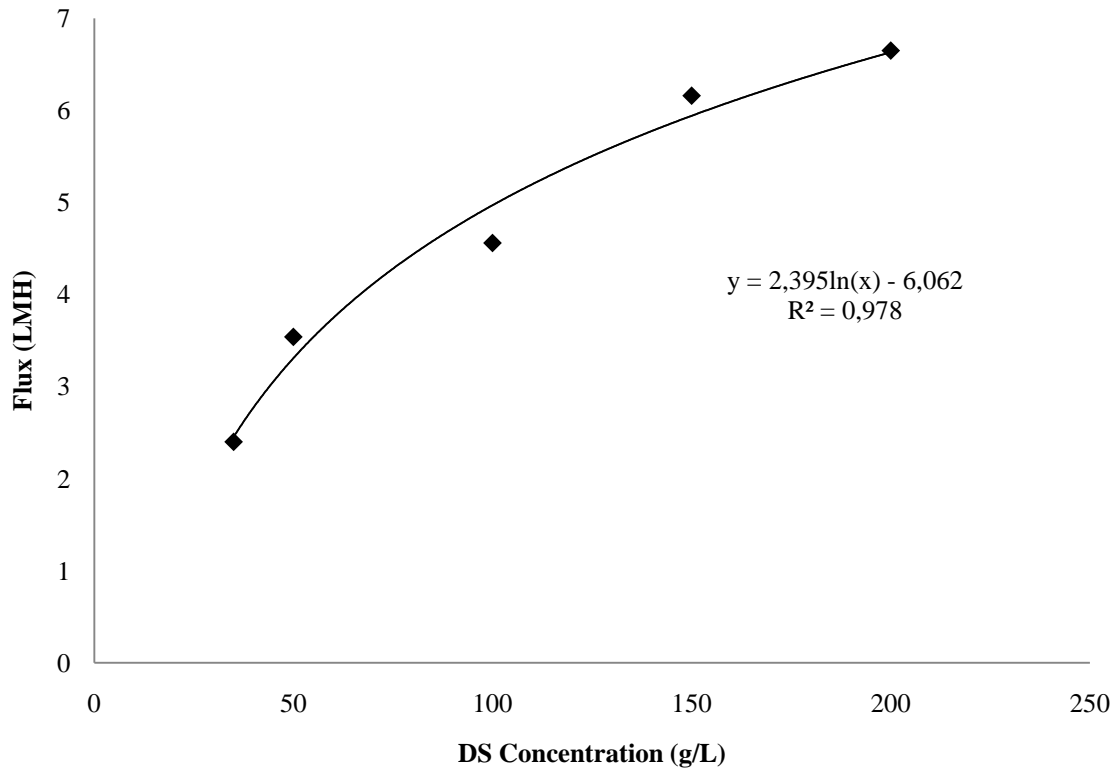


Fig. 3. Plot of average flux at different DS concentrations.

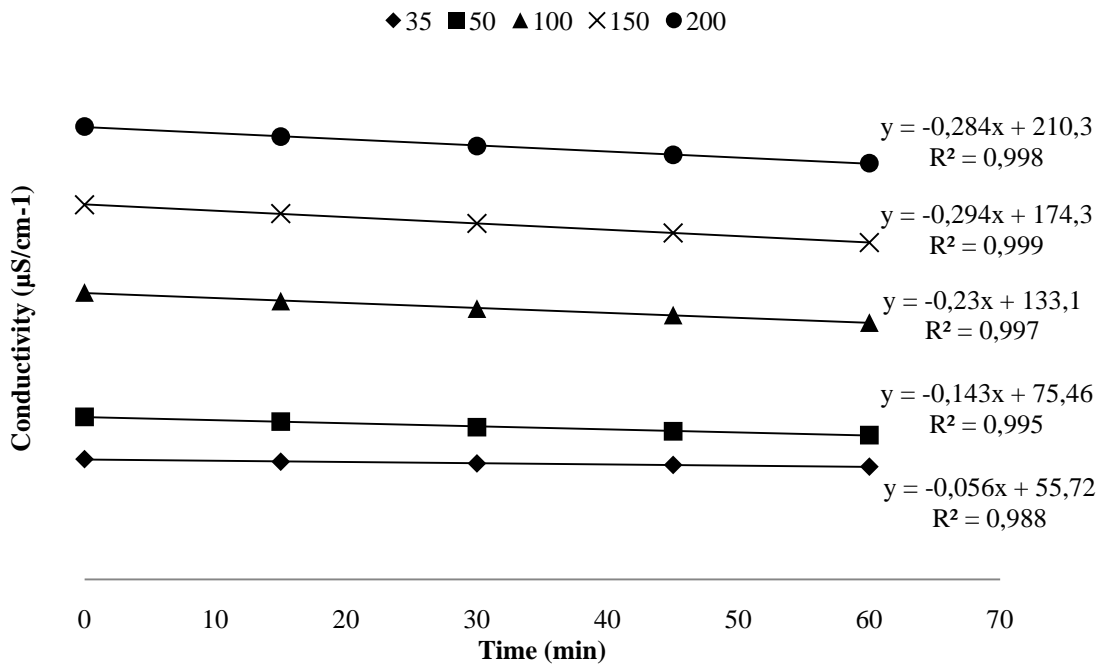


Fig. 4. Decrease in conductivity at different DS concentrations operated at 0.2 L/min.

Table 2. Change in conductivity of draw solution at various DS concentrations with different recirculation flowrates.

DS Concentration (g/L)	change in conductivity / change in time ( $\mu\text{S cm}^{-1} \text{ min}^{-1}$ )		
	0.1 L/min	0.2 L/min	0.3 L/min
35	0.04	0.06	0.06
50	0.10	0.14	0.17
100	0.17	0.23	0.28
150	0.23	0.29	0.32
200	0.20	0.28	0.30

#### *Operation of FO-MBR system in continuous mode*

Flat sheet MF and FO membranes were submerged in the oxic reactor of the MBR system and operated for a month. To evaluate the permeate quality, periodical sampling was conducted and subjected to TN, TP and COD analyses. The draw solution was changed every four days with freshly prepared DS concentration of 200 g/L. At the end of the operation, the fouled membranes were subjected to SEM analysis to evaluate the extent of fouling onto the membrane.

The permeate flux was monitored throughout the operation and separated into three parts as shown in Figure 5. The first part of the graph shows a very steep curved line, which illustrates the rapid decrease in flux. At this part, the rapid flux decline was attributed to the high initial DS concentration of 200 g/L. It can be observed, at the lower portion of the first part, that the slope of the graph slowly decreases. The dilution effect on the draw solution affected the water permeation across the membrane resulted to the decrease in flux. At the middle portion of the figure, the second part can be seen having a linear decrease in flux. The change in flux at this part was almost constant creating a linear relationship between flux and time. Lastly, the last part of the graph gave a much lower change in flux as compared to the middle part. This phenomenon occurred due to the dilution effect in the draw solution, and the low concentration difference between the draw solution and the mixed liquor inside the MBR. Decreased concentration of the draw solution affected the water filtration across the FO membrane. These results were supported by the decrease in conductivity throughout the operation as presented in Figure 6. Rapid decrease in conductivity was observed at the first part of the graph, then a constant decrease at the middle part, and a much lower difference of conductivity at the last part. Thus, a linear correlation between flux and conductivity was concluded.

Another essential aspect to consider is the quality of permeate. To make the permeate acceptable as feed for RO system, it should be of high quality, with less contaminant or low nutrient content. Thus, the concentration of TN, TP and COD were measured, and the effluent qualities of both MF and FO were compared. Results of TN, TP and COD concentrations in the effluent were presented in Figure 7. It was observed that the percent removal of FO is greater than MF. This can be attributed to the difference on the morphology of MF and FO membranes. Having a dense layer, the rejection of contaminants in FO membrane was much higher than FO membrane. The average percent TN, TP and COD removals of FO membrane reached 97.00 %, 97.05 % and 97.88 %, respectively. Even though the rejection of nutrients by FO membrane was high, it is important for the MBR system to be operated at optimized condition to prevent accumulation of nutrients inside the reactor, which could have an adverse effect on FO membrane. Membrane fouling is still a controversial issue up to date, thus, it is important to visualize the effect of foulants on membranes. SEM images were generated to show the extent of fouling on MF and FO membranes as shown in Figure 8. Severe fouling was observed in MF membrane while FO membrane, both active and support layers, was not significantly affected by fouling. This can be explained by the natural flow of water through the membrane. The only driving force used in FO process is the osmotic pressure across the membrane, unlike MF process, which uses suction to generate permeate. Therefore, high effluent quality and low membrane fouling tendency could be obtained by the application of FO, which is suitable as feed for RO system.

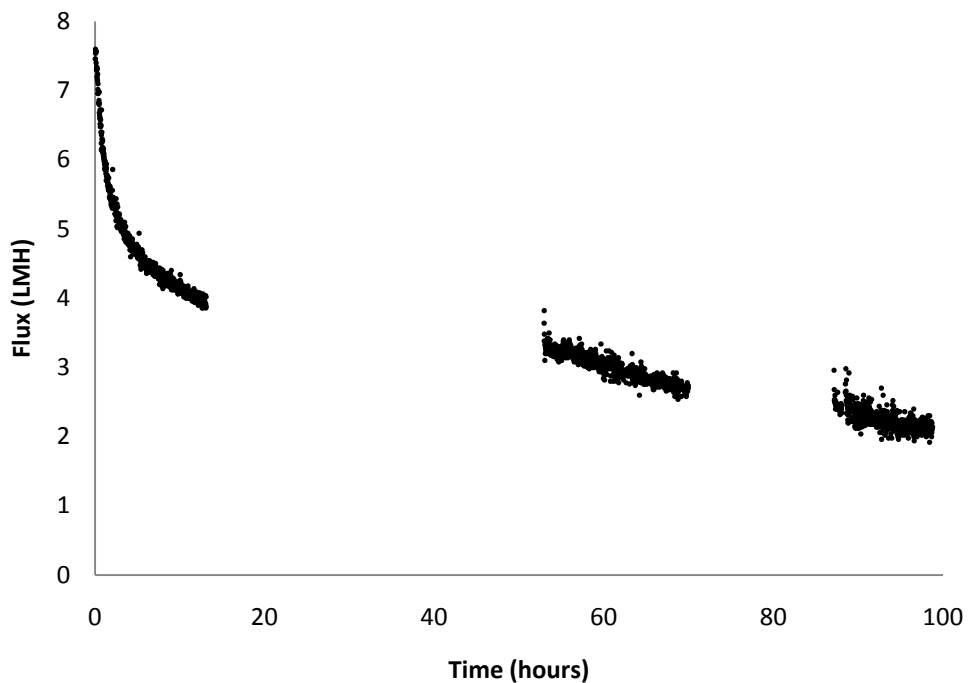


Fig. 5. Decline in water flux in an FO-MBR system during continuous operation of 60 hours.



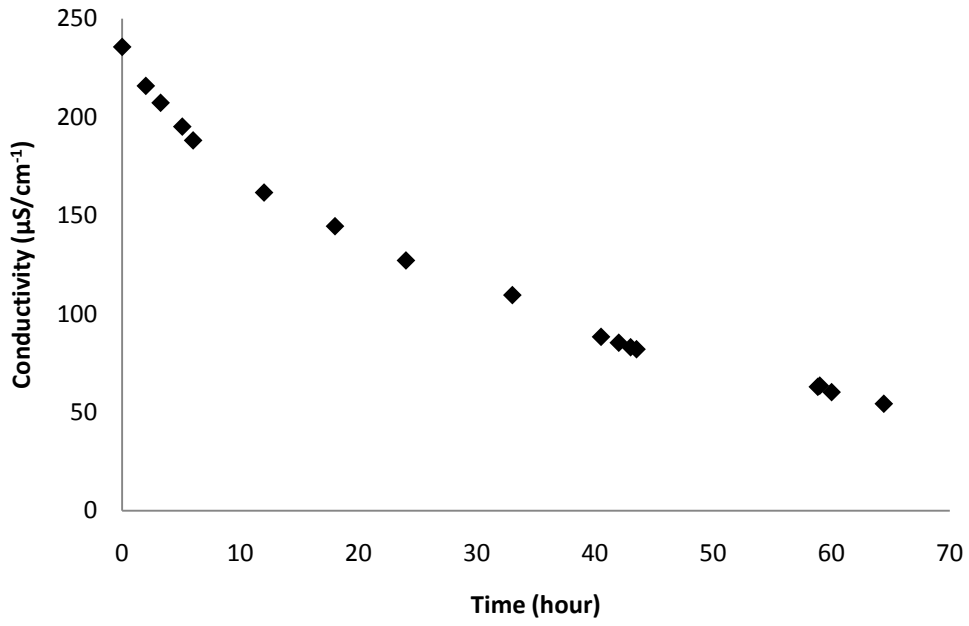


Fig. 6. Decrease in conductivity in a closed-loop FO-MBR system for 60-hour operation.

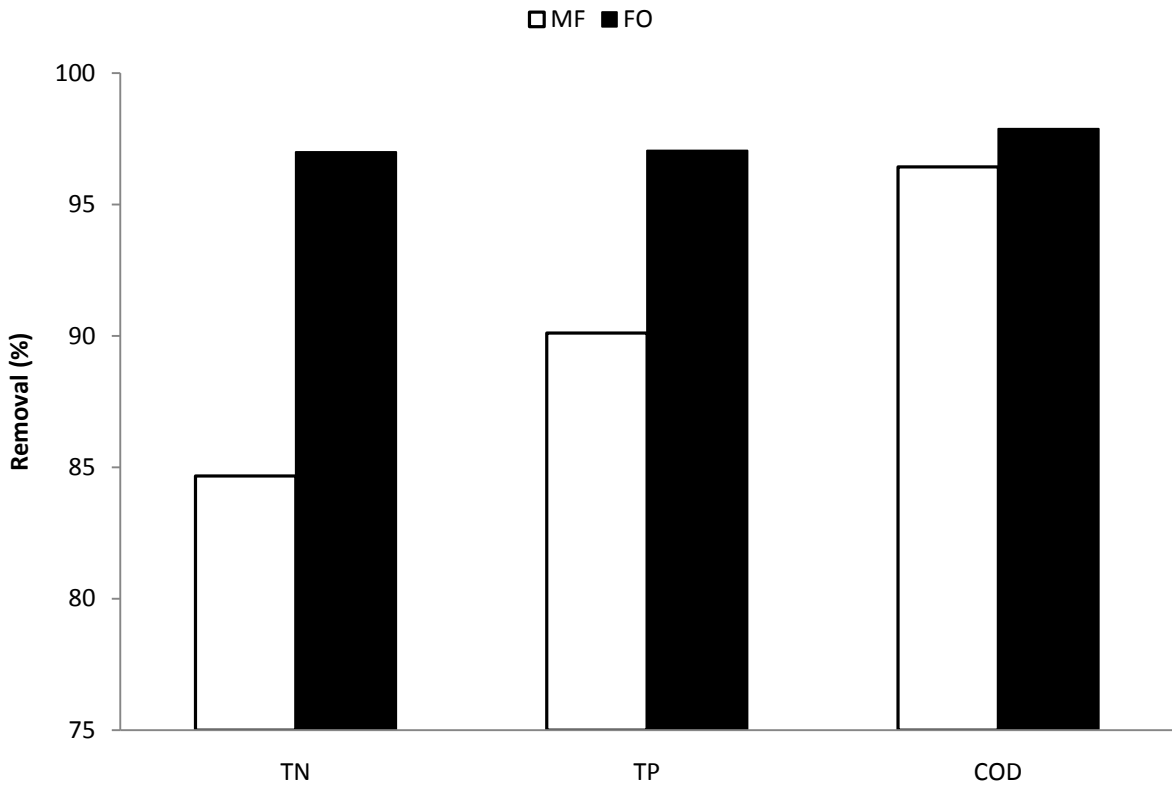


Figure 7. Average nutrient removal of MF-MBR and FO-MBR systems for continuous operation.

## Conclusion

Integrated FO-MBR system was successfully operated for both batch and continuous modes. For the batch test, different concentrations of draw solution was utilized, and found that the highest DS concentration of 200 g/L obtained the highest flux, but recorded the lowest change in conductivity with respect to time,  $\Delta C/\Delta T$ . It was also observed that increasing the operating flowrate increases  $\Delta C/\Delta T$ . For the continuous mode, a linear relationship between flux decline and decrease in conductivity was observed. Nutrient removal and membrane fouling was also investigated. FO membrane efficiently removed nutrients as compared to MF membrane. FO-MBR system obtained percent TN, TP and COD removals of 97.00 %, 97.05 % and 97.88 %, respectively. In addition, fouling on FO membrane, both active and support layers, was not evidently seen in SEM images. Thus, integration of FO to MBR system generated an improved effluent quality, which can be used as feed for RO system.

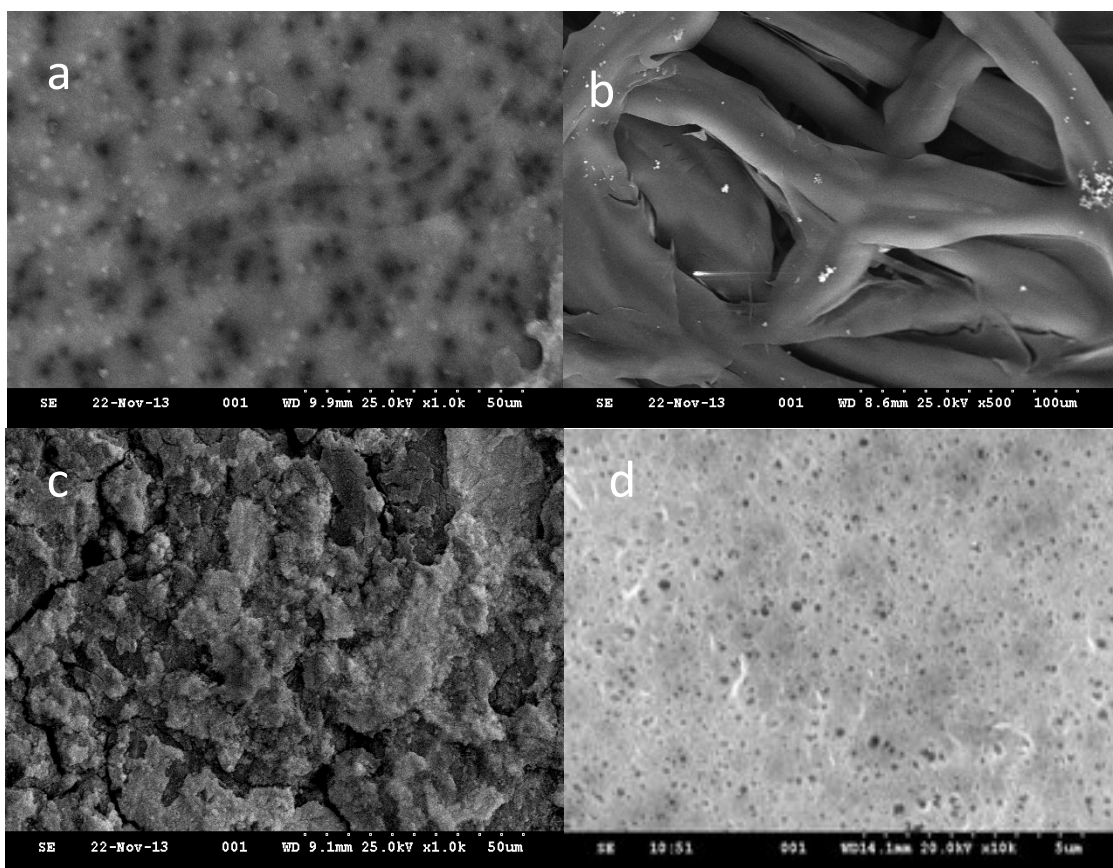


Fig. 8. SEM images of (a) fouled FO membrane – active layer; (b) fouled FO membrane – support layer; (c) fouled MF membrane; and (d) virgin MF membrane.

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