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Pilot study for sewage wastewater reclamation and reuse using RO membrane: comparison of different pre-treatment systems

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ABSTRACT

A pilot plant study was designed to monitor the performance of two parallel lines with a capacity of 50 m^3/day using different pre-treatment technologies prior to reverse osmosis (RO) units for water reclamation from a local-based effluent treatment plant in Malaysia. Line 1 consisted of coagulation-pore controllable fiber filter (PCF) and was denoted as PCF-RO, while line 2 was sand filter-ultrafiltration (UF) and was denoted as UF-RO. The pilot plant was operated continuously everyday for three months throughout the study. In the PCF-RO line, ferric chloride was chosen as a coagulant for the system. The performance and efficiency of PCF-RO compared to UF-RO in terms of system operability, percentage reduction of parameter tested, system deterioration, and the effectiveness of ferric chloride as coagulant were investigated. The results showed that permeate quality for both systems met the WHO drinking water standard for drinking water. However, membrane performance for PCF-RO which was deteriorated over the operation period, had led to lower rejection of BOD, COD, and other parameters. The long-term performance and efficiency of UF-RO in terms of percentage reduction were better than PCF-RO line indicating that membrane performance in the PCF-RO line was affected by coagulant. Interactions of membrane with coagulant seemed to affect RO membrane stability. SEM images were obtained to compare the membranes after long-term operations.

Keywords: Ultrafiltration; Pore controllable fiber (PCF) filter; Sewage treatment; Water reclamation

1. Introduction

Membrane filtration process is gaining acceptance rapidly as a promising technology for wastewater reclamation due to the improvement of production process in terms of cost effectiveness and consistent high-quality permeate. Filtration processes, such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) have been widely used

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in different applications, such as desalination and demineralization of saline water, drinking water production, wastewater treatment, chemical, food, and pharmaceutical industries [1–4]. However, the major drawback of this technology is membrane fouling which can affect membrane performance in the terms of stability of product quality and operation cost over the operation period [5]. In order to expand the membrane life time and performance, pre-treatment units are used prior to RO to remove organic matter and biological species present in the sewage wastewater. Coagulation, sand filtration (SF), and UF are some examples of pre-treatment processes used prior to RO for the purpose of improving the quality of feed water to the level which would result in optimal performance of the RO membranes. Coagulation in wastewater treatment plant can be accomplished with coagulants, such as synthetic polymer, ferric salts, and alum by neutralizing the forces that make colloidal particles repel each other. As results, the colloidal particles collide to form larger flocs, whereas SF is excellent to remove the suspended solids present in water. In the previous studies [6], MF/RO system was used for a pilot study in wastewater reclamation and reuse. The results showed that rejections of RO membrane in terms of *E. coli*, alkalinity as $CaCO_3$, and NO_2-N were 100, >73, and >90%, respectively. Another study [7] demonstrated that MF/RO pilot plant can reject the parameters, namely conductivity and nitrate higher than 96 and 85%, respectively. Pre-treatment was conducted for both of these studies to reduce fouling occurrence on RO membrane. Therefore, the purpose of this study was to study the performance of two parallel RO membrane systems which were pre-treated by a pore-controllable filter (PCF) and a UF membrane, respectively. The local sewage treatment plant basically treats the sewage water using physical and chemical processes. For primary treatment, screening and sedimentation were used to remove some of the suspended solids and organic matter. The effluent from the primary treatment will still contain high amounts of organic matter. In this case, the primary effluent will be channeled to the secondary treatment units. Secondary treatment used biological unit processes, such as aeration, activated sludge systems, and sequencing batch reactors for the removal of biodegradable organic and suspended solids. The secondary effluent from this sewage treatment plant was then used in this study.

2. Material and method

2.1. Design of pilot plant

The pilot plant used in this study consisted of two parallel units of 50 m^3/day PCF technology and 50 m^3/day UF membrane technology as pre-treatment prior to the RO membrane unit. The performance of the pilot plant, with the two units running in parallel, for water reclamation from a sewage effluent treatment plant based in Malaysia was monitored. The pilot plant was operated continuously for 24 h for over three months. The treated sewage effluent was used as raw feed water in this study. The schematic diagram of the pilot plant is presented in Fig. 1. The PCF-RO line consisted of coagulation tank, PCF unit 1, PCF unit 2, and RO, whereas UF-RO line consisted of sand filter (dual-media filter), UF unit, and RO. In PCF-RO line, ferric chloride with 40% concentration was chosen as coagulant agent for the system to enhance the removal efficiency of colloidal elements in the feed water. PCF filter consisted of flexible fiber bundles surrounding the strainer which can be pulled-up to the longitudinal direction to reduce its pore size up to a thickness of 0.5–0.6 cm to form the filter media. The pore size and the operating flux of the PCF filter are 5–10 μm and 6.25 m^3/h, respectively. The filtration pressure of water from outside to inside of the strainer was up to 0.03–0.7 bar. This PCF filter also has a backwash function to clean the fiber bundles with backwash water by compressed air after releasing them. The PCF filter was chosen as one of the pre-treatment units in this study due to its benefits, such as the following:

(1) High efficiency and excellent water quality.
(2) Compact size and low cost.
(3) Very effective backwashing.
(4) Long life-time of filter media and simple to exchange.
(5) Simple design.
(6) Easy to coat for anticorrosion.

The UF model, SFP2860 was chosen as a pre-treatment unit in this study. This UF unit has 0.03 μm pore of PVDF membrane. It is excellent in removal of bacteria, viruses, and colloids prior to RO system. Moreover, this PVDF membrane also has features, such as high strength and chemical resistance which allow long membrane lifespan. The operating flux of the UF unit is within the range of 40–120 m^3/m^2 h. For PCF units 1 and 2, the system was backwashed for 120 and 60 s, respectively, after 20 min of operation. In comparison with PCF unit, UF unit was backwashed for 60 s after 20 min of operation. The details of raw water are shown in Table 1. The RO membrane that was used for both systems was polyamide thin-film composite (DOW FILMTEC BW30-4040). Table 2 shows the detail specification of the RO unit.

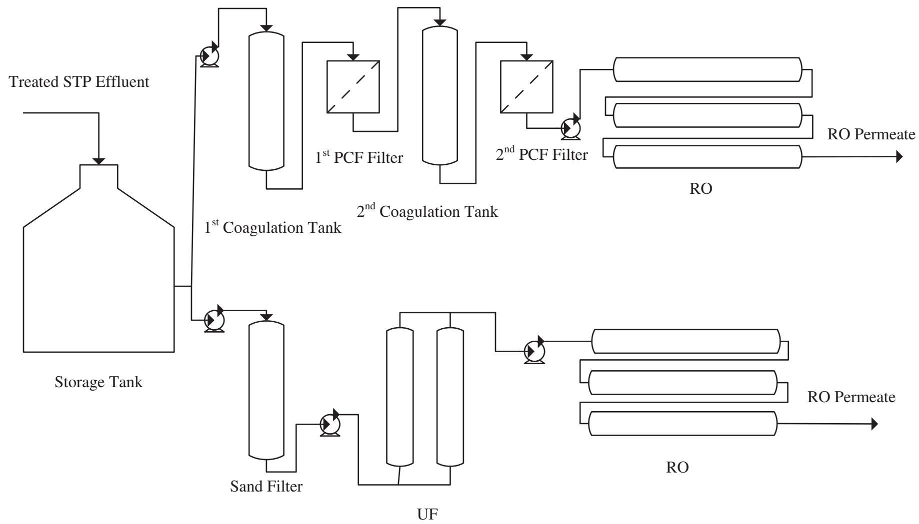

Fig. 1. Schematic diagram of pilot plant.

Table 1
Feed water quality characteristics

Parameter	Units	Average	Max	Min
pH	–	7.18	7.40	6.99
BOD_5	mg/L	19.30	48.0	1.15
COD	mg/L	29.3	39.0	24
TSS	mg/L	15.8	24.0	8.1
TDS	µS/cm	225.51	255.08	197.30
Conductivity	µS/cm	445.75	507.00	363.00
Ammonia	mg/L	15.2	22.3	11.8
Nitrate, as N	mg/L	2.10	6.75	0.16
Nitrite, as N	mg/L	2.27	12.12	0.12
Silica	mg/L	11.59	13.31	7.25
Color	Pt–Co	127.13	211.00	94.00
TKN	mg/L	22	24	20
E. coli	Cfu/100 mL	6,500	9,600	3,400
Alkalinity as $CaCO_3$	mg/L	46	53	39

Table 2
RO unit specification

Item	Specification
Model	DOW FILMTEC BW30-4040
Membrane type	Polyamide thin-film composite
Dimensions	3.9″ W × 40″ L
Feed flow rate	1.5–3.6 m^3/h
operating pressure	11–15 bar
Recovery ratio	93–95%

2.2. Analytical methods

Sampling was carried out weekly on site. Water samples were analyzed for parameters, such as pH, BOD_5, COD, total suspended solid (TSS), total dissolved solids (TDS), conductivity, ammonia, nitrate, nitrite, silica, color, total kjeldahl nitrogen (TKN), *E. coli*, and alkalinity as $CaCO_3$ as per the standard methods. Some of the parameters, such as silica, TKN, and *E. coli* were analyzed periodically. At the completion of the pilot study, RO membranes for both lines were sent to scanning electron microscope (SEM) unit for analysis. Membrane surfaces and cross-sections were analyzed using SEM (Gemini model SUPRA 55VP-ZEISS) coupling with X-ray detector (Oxford instrument model INCA PentaFET-x3). For cross-sectional analysis, the membranes were fractured in liquid nitrogen and then coated with gold prior to analysis. Samples were further analyzed with Fourier transform infrared spectroscopy (ATR-FTIR) using FTIR Nicolet 6700.

3. Results and discussion

Table 3 shows the result for UF effluent and RO permeate quality for the UF-RO line, while Table 4 shows the result for PCF effluent and RO permeate quality for the PCF-RO line.

The performance of both the PCF-RO and UF-RO were monitored in terms of their permeate quality. The performances of both PCF-RO and UF-RO were excellent and produced permeate that met the WHO standard for drinking water. The rejection percentage of RO units is based on the calculation below in terms of concentration (mg/L):

$$\text{Rejection}\,\% = \left(\frac{\text{Permeate}_{\text{UF or PCF}} - \text{Permeate}_{\text{RO}}}{\text{Permeate}_{\text{UF or PCF}}}\right) \times 100\% \quad (1)$$

As listed in Table 3, rejection of nitrite and nitrate in RO permeate for UF-RO line was lower than PCF-RO line in the first two months. However, this situation became contradictory starting from July until completion of the study. The concentration of nitrite and nitrate are illustrated in Figs. 2 and 3. All nitrite and nitrate concentrations for UF-RO line were below 0.15 and 1 mg/L, respectively.

It can be seen that the UF and PCF were not efficient in ammonia removal for the feed water. However, the obtained results showed that RO for both lines was sufficient to reject ammonia where the rejection percentages of UF-RO and PCF-RO are 79.2–88.1 and 33.6–71.5%, respectively. The concentration of ammonia for both lines is illustrated in Fig. 4.

The relationship between ammonia, nitrate, and nitrite over operating time are shown in Figs. 2–4. It can be seen that ammonia, nitrate, and nitrite concentrate in UF-RO permeate showed negative relationship over the operating time. These trends showed that the rejection of ammonia, nitrate, and nitrite for UF-RO increased gradually with time. For PCF-RO line, all the three measured parameters for PCF-RO permeate increased with time. These indicated that RO permeate in terms of ammonia, nitrate, and nitrite became lower and performance of PCF-RO system was deteriorated.

When dosing ferric chloride as coagulant to raw water, it can be seen that overall performance of PCF-RO was excellent at the initial stage of the study. This was strongly supported by the results where the parameters tested were within target. However, the performance of PCF-RO started to decline and showed lower performance than UF-RO after the first two months. This resulted in some parameters exceeding the drinking water standard. These parameters were BOD_5, COD, ammonia, color, and *E. coli*. This could be caused by the damage to the membrane due to the use of coagulants. The

Table 3
Permeate quality UF-RO

Parameter	Units	UF effluent			RO permeate			Percentage rejection %
		Average	Max	Min	Average	Max	Min	
pH	–	7.02	7.28	6.20	6.83	7.47	6.07	–
BOD_5	mg/L	13.72	45.00	0.54	3.33	14.00	0.13	20–92.3
COD	mg/L	12.3	22.0	1.0	2.7	8	0	50–100
TSS	mg/L	5.8	14.0	1.0	2.3	6.1	0.4	30–86.7
TDS	μS/cm	216.60	246.14	184.19	13.53	18.85	10.03	92.3–95.3
Conductivity	μS/cm	433.25	492.00	358.00	28.15	37.5	24.96	92.2–94.3
Ammonia	mg/L	16.32	28.10	10.60	2.35	4.50	1.32	79.2–88.1
Nitrate, as N	mg/L	1.83	4.00	0.33	0.28	0.93	0.02	28.9–98.7
Nitrite, as N	mg/L	0.78	2.05	0.29	0.05	0.13	0.01	80.1–98.5
Silica	mg/L	9.10	10.98	6.66	1.89	6.97	0.19	36.5–98
Color	Pt–Co	21.13	31.00	12.00	1	3	0	90.3–100
TKN	mg/L	22	25	19	5	6	4	76–79
E. coli	Cfu/100 mL	96	192	0	0	0	0	100
Alkalinity as $CaCO_3$	mg/L	27.5	48	7	13.5	18	9	0–81.3

Table 4
Permeate quality PCF-RO

Parameter	Units	PCF effluent			RO permeate			Percentage rejection %
		Average	Max	Min	Average	Max	Min	
pH	–	6.87	7.13	6.30	7.05	7.84	6.49	–
BOD_5	mg/L	16.63	42.00	0.85	9.68	29.00	0.01	0–99.1
COD	mg/L	15.4	27	4	5.5	13	0	27.3–100
TSS	mg/L	7.1	10	1.5	2.6	5.1	0.1	49–93.3
TDS	μS/cm	225.65	256.66	200.44	71.27	112.00	16.12	45.6–92
Conductivity	μS/cm	450.63	515.00	389.00	144.41	238.90	32.10	45.5–91.8
Ammonia	mg/L	14.68	21.40	10.70	7.49	9.22	5.70	33.6–71.5
Nitrate, as N	mg/L	1.72	7.75	0.14	0.90	3.50	0.08	12.5–54.8
Nitrite, as N	mg/L	1.12	4.15	0.04	0.12	0.32	0.02	2.3–98.3
Silica	mg/L	9.71	12.18	5.94	4.61	8.62	1.39	28–88.5
Color	Pt–Co	38.75	74	16	12.5	22	1	47.1–93.8
TKN	mg/L	20	25	15	6	7	5	66.7–72
E. coli	Cfu/100 mL	7,675	11,700	3,650	950	1,900	0	48–100
Alkalinity as $CaCO_3$	mg/L	44	52	36	10	14	6	61.1–81.3

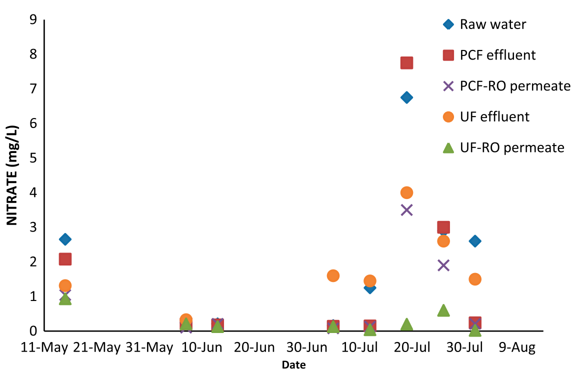

Fig. 2. Concentration of nitrate in pilot unit.

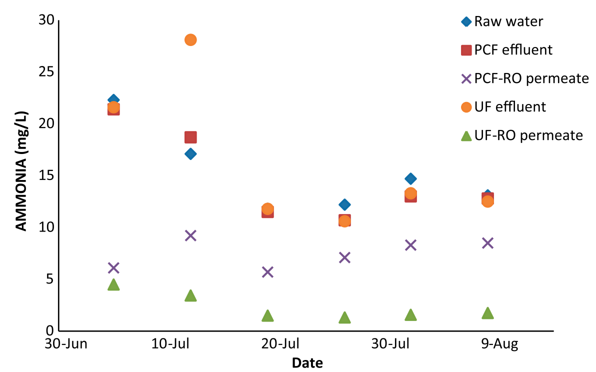

Fig. 4. Concentration of ammonia in pilot unit.

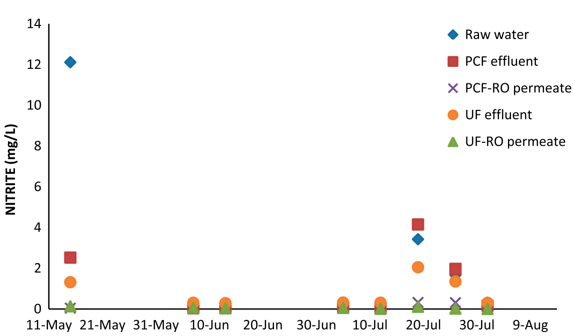

Fig. 3. Concentration of nitrite in pilot unit.

effect of ferric chloride coagulant residuals on polyamide membrane performance has been investigated previously [8]. Their studies revealed that ferric chloride residuals caused the chlorination reaction and led to polyamide membrane degradation. BOD_5 test is a measurement of the quantity of oxygen required for biochemical oxidation of organic matters in wastewater. BOD_5/COD ratio has been widely used as an indicator of biodegradability of the wastewater. Results showed that BOD_5 values of some samples were higher than COD values. This could be due to the concentration of ammonia which for certain sampling weeks was high compared with others and thus led to high-oxygen demand for nitrification process. The linear regression result obtained between ammonia concentration and BOD_5 values is depicted in Fig. 5. The trend showed the high positive correlation between the two variables with correlation coefficient being 0.85.

Comparative performance in terms of rejection percentage between PCF-RO and UF-RO were shown in Fig. 6. Overall, all the RO rejection percentages of tested parameters for UF-RO were higher than PCF-RO except for BOD_5, TSS, and alkalinity as $CaCO_3$ when the pilot plant was operated at optimal condition and setting. This was due to that the UF unit produced better quality of UF permeate compared to PCF unit especially in terms of BOD_5, TSS, and alkalinity as $CaCO_3$, and thus, less quantity of contaminants to be fed into RO unit which resulted in low RO rejections. The final RO permeate quality for UF-RO line in terms of BOD_5, TSS, and alkalinity as $CaCO_3$ were better than PCF-RO line. This indicates that the UF is superior as pre-treatment method for RO system to produce high-quality permeates and cost-effective in operation.

3.1. SEM-EDS analysis of membrane

Two samples of the used RO membranes from each line were chosen for SEM analysis. SEM images of fouled RO membrane for PCF line and UF line are presented in Fig. 7(a)–(b). The membrane surfaces were imaged at relatively high-magnification level which was 3.00 kx. It was found that membrane surface for Fig. 7(b) is clearer than (a).

The micrograph in Fig. 8(a)–(b) shows the cross-section of RO membrane for both operation lines. The top layer of the membrane was deposited with foulants and it can be observed that the thickness of foulants deposited on PCF-RO membrane was about nine times as that of UF-RO. The thickness of foulants was in the range of 4.29–6.43 μm and 0.13–0.75 μm for both systems, respectively.

Application of $FeCl_3$ lowered down the permeate flux decline for the system. Fig. 9 shows the flux decline for both lines. However, severe membrane fouling (i.e. pore blocking of foulants and formation of cake layer on the membrane surface) [9] could have occurred due to the presence of hydrophilic compounds in the feed water which were poorly removed by coagulants [10]. Another study [11] also reported

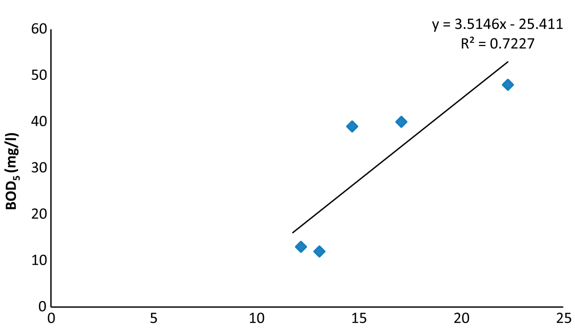

Fig. 5. Linear regression of BOD and ammonia.

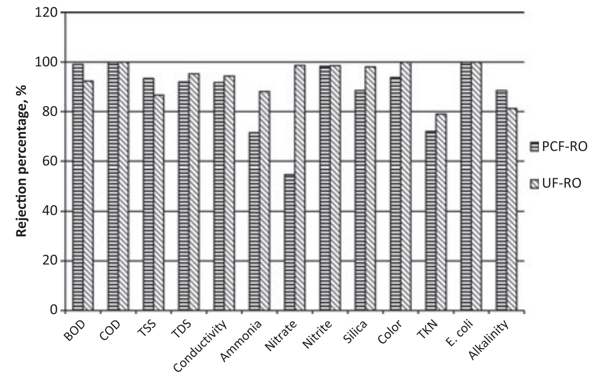

Fig. 6. Comparison of rejection percentage between PCF-RO and UF-RO.

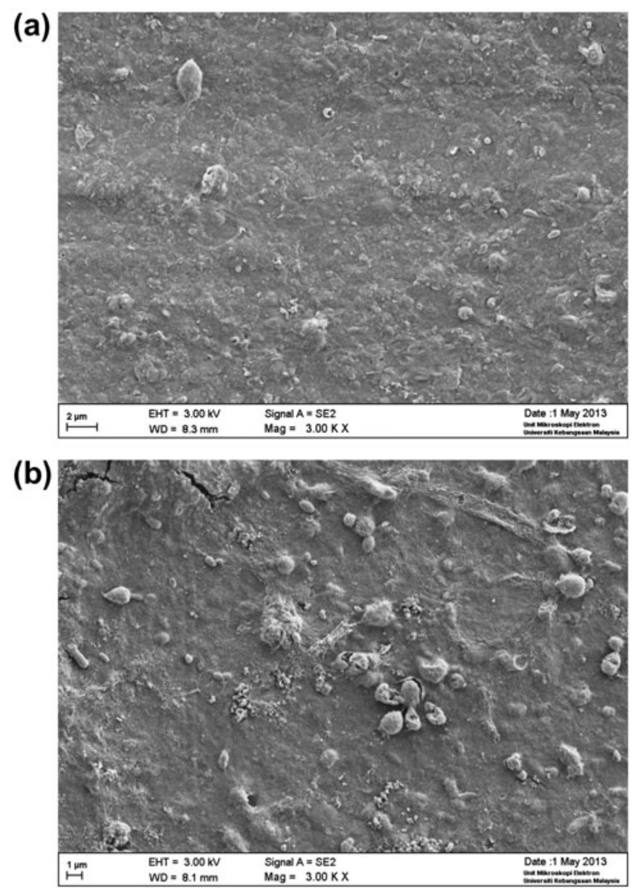

Fig. 7. SEM images of fouled RO membranes: (a) PCF-RO and (b) UF-RO.

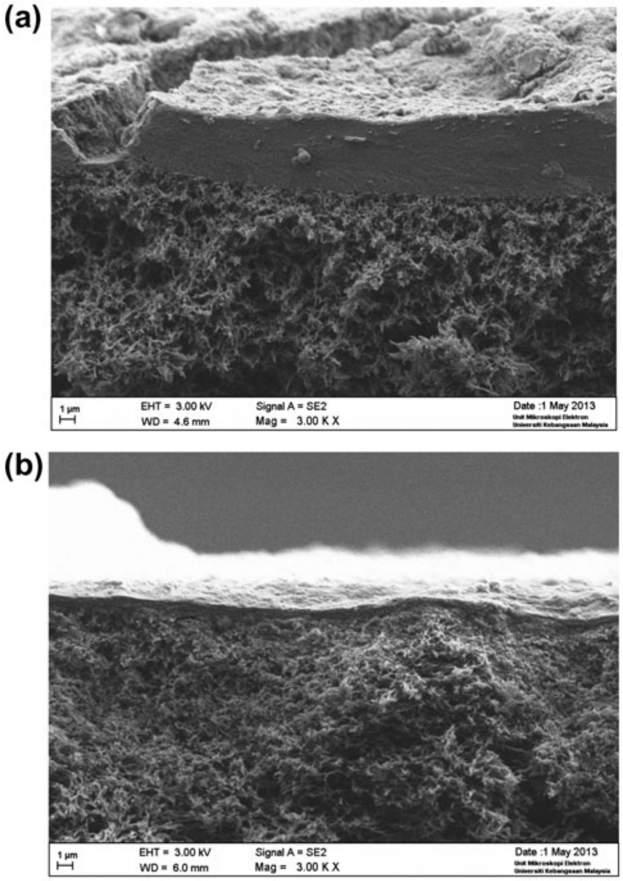

Fig. 8. Cross-section of fouled RO membranes: (a) PCF-RO and (b) UF-RO.

that the SEM analysis of fouled membrane with ferric chloride-treated water sample showed denser and gel-like appearance cake layer.

The two RO membranes were then sent for scanning electron microscopy-energy dispersive X-ray spectrometry (SEM-EDS) analysis. The EDS result showed that the foulants for PCF-RO membrane was mainly comprised of C, O, and Fe elements at 34.71, 37.56, and 14.81% by means of weight, respectively, while UF-RO comprised of C, O, and S elements at 63.98, 26.17, and 7.03%. Moreover, the Fe element in UF-RO membrane is just 1.86% in weight percentage and it was relatively smaller in comparison to the PCF-RO membrane. The elements S and Fe were found on both the RO membranes. They entered sewage treatment plant in the form of urine [12], food residuals [13], detergent [14], and pharmaceutical drugs [15]. Degradation of proteins containing ferrous and sulfur compounds from urine in sewage treatment plant contributed to detection of elements S and Fe in RO membrane. However, these results are inadequate to identify the possible root cause of membrane fouling. Therefore, ATR-FTIR was carried out.

For this section, FTIR analysis was conducted to investigate the functional groups that were present on the fouled RO membrane. Two samples were subjected for analysis. The IR spectra are depicted in

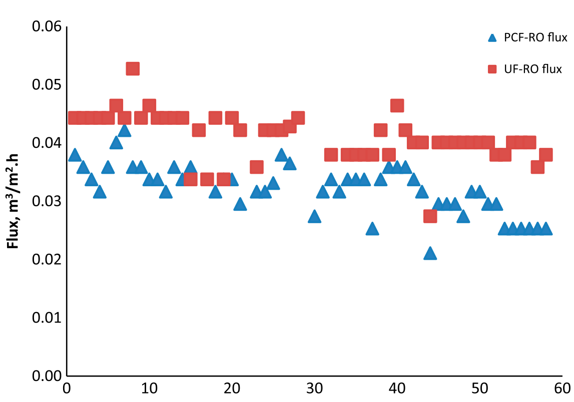

Fig. 9. RO membrane flux decline for both lines.

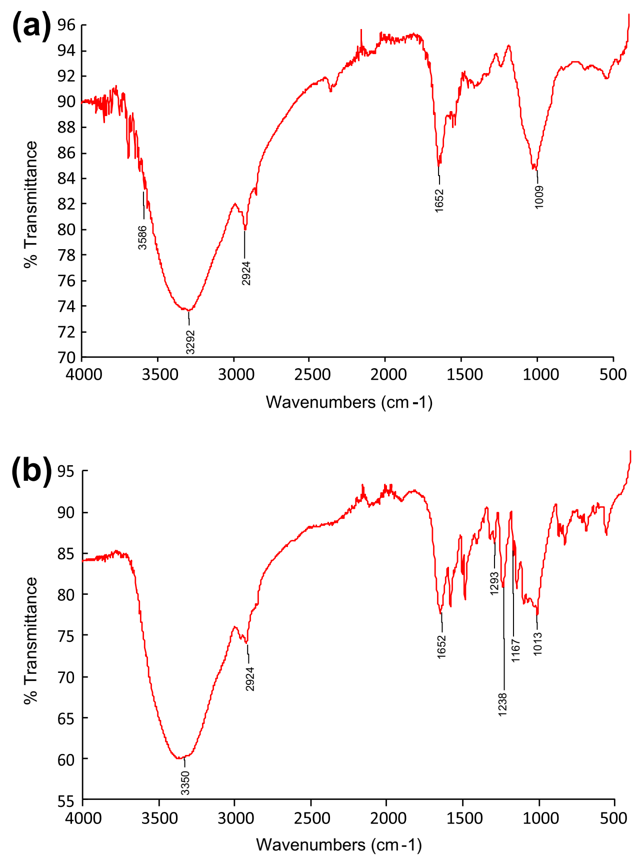

Fig. 10. FTIR results for RO membranes: (a) PCF-RO and (b) UF-RO.

Fig. 10. Two IR peaks at 2924 and 1652 cm^{-1} appeared for both Figs. 10(a) and (b) which are most likely referred to as alkane functional group (C–H stretch) and amide group, respectively. A strong peak at region 3350 cm^{-1} with a broad wavelength in Fig. 10(b) is due to the stretching vibration of –OH functional group. There are two sharp IR peaks depicted in Fig. 10(a) compared to (b) at 3,292 and 3,586 cm^{-1}. This proved that the presence of free O-H functional group in PCF-RO membrane was higher than UF-RO. These results were in tally with the SEM-EDS results where the weight percentage of Fe element present in PCF-RO was twice than that present in UF-RO. The patterns are similar to the study of pre-treatment of petrochemical wastewater by coagulation previously. In the study [16], sludge after treatment by $FeCl_3$ depicted the broad wavelength in regions between 3,150 and 3,650 cm^{-1}. Other strong IR peaks could be seen in 1,000–1,300 cm^{-1} regions for both spectra, which were at 1,009 cm^{-1} and 1,238 cm^{-1} for Fig. 10(a) while at 1,013, 1,167, 1,238, and 1,293 cm^{-1} for Fig. 10(b), showing that C–O group was present. This significantly proved that carboxylic acid content existed in the foulants.

4. Conclusions

Both UF and PCF are good as pre-treatment units prior to RO membrane formation in the reclamation of treated sewage system. The results showed that permeate quality for both systems met the WHO drinking water standard for drinking water. However, the performance and efficiency of PCF-RO deteriorated over operation period. It was found that the percentage reduction of parameters tested such as BOD_5, COD, etc. decreased with the function of time. In contrary, the long-term performance and efficiency of UF-RO in terms of percentage reduction were better than PCF-RO line. This indicated that membrane performance in the system was affected by coagulant residuals presence in water. In addition, the thickness of foulants deposited on PCF-RO membrane was thicker than UF-RO. FTIR analysis showed the similar patterns of wavelength in regions between 3,150 and 3,650 cm^{-1} for PCF-RO system with previous study which was also used $FeCl_3$ as coagulant.

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