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Using tertiary-treated municipal wastewater as makeup water by reverse osmosis membrane

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ABSTRACT

The objective of this research was to study the possibility of using reclaimed municipal wastewater in cooling tower by treating the water using reverse osmosis (RO) membrane. In this study, RO membrane was operated at initial flow rate of 6.3 m^3/d, whereas the initial transmembrane pressure (TMP) was at 116 psi. During the operating period, permeate flux slightly decreased to 4.8 m^3/day with the increase of TMP up to 127 psi and membrane fouling resistance of $3.34E + 12\ m^{-1}$. Major membrane fouling mechanism was complete blocking followed by standard blocking and cake layer formation. Average permeate characteristics were 36.3 µS/cm of conductivity, 42.3 mg/L of TDS, 4.4 mg/L of hardness, 22.7 mg/L as $CaCO_3$ of alkalinity, 0.002 mg/L of iron, 0.05 mg/L as $CaCO_3$, 1.5 mg/L of chloride, 2.1 mg/L of silica, and there was no phosphate passing through RO membrane. Percent rejections compared to UF treated wastewater were in the range of 90.2–99.9% except for silica where rejection was only 85.1%. Scaling and corrosion index was studied in terms of Langelier saturation index (LSI) and Ryznar stability index (RSI). Without RO membrane, LSI value was −0.59 and RSI value was 8.5. With RO membrane, LSI value was −5.0 and RSI value was 17.0. From calculation of cycles of blowdown, it was found that when using the membrane, the cycles increased from 2.3 to 44 cycles. This, then, resulted in reduction of makeup water from 379.0 to 221.2 m^3/d, a decrease by 41.6%. Payback period of installation and operation of RO membrane system was 14 months.

Keywords: Makeup water; Reverse osmosis membrane; Tertiary-treated municipal wastewater

1. Introduction

Cooling systems utilizing water evaporation are usually the most cost-effective heat rejection systems as they provide the most cost-effective cooling technology for commercial air conditioning and industrial processes. However, makeup water quality is preferred with low conductivity, hardness, alkalinity, ion, colloidal, and dissolved solids. The additional concerns for operate the cooling systems are control of scale formation, corrosion condition, and biological fouling. As these increase the difficulty and costs associated with operating a cooling tower water system, chemical treatment of the cooling water is thus required to operate the cooling tower without scale formation and corrosion condition [1]. Wastewater reclamation is the treatment of wastewater up to reusable water quality standard [2,3]. The benefits of

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wastewater reclamation are, for example, (i) reduction of reliability on natural water resources, particularly important in the water sensitive areas, (ii) cost reduction due to less purchase of water supply, and (iii) environmental impacts reduction due to reduction of energy and chemical demands. Recently, wastewater reclamation was applied in various applications in various areas such as industrial, residential, irrigation, and agricultural areas [4–6]. Cooling tower is also one of the common options where reclaimed wastewater is used [7]. The principle of cooling tower involves providing cooled water by rejected heat through the natural process of evaporation. Loss of water from the evaporation results in concentrated dissolved solid which causes higher scale forming potential and leads to more corrosion problems [8]. To maintain the dissolved solids concentration within the standard used for cooling tower, bleed off (or blow down) of concentrated water with makeup water is required. Makeup water quality is the key factor that affects the maximum cycles of concentration; therefore, potable water is normally used as makeup water in a cooling tower. Previous study showed that the use of treated municipal wastewater resulted in higher scaling formation and corrosive potential [9,10]. Scaling could reduce heat transfer efficiency and overall performance of a cooling tower system [11]. The most common indicators used to predict the scaling and corrosive conditions are Langelier saturation index (LSI) and Ryznar stability index (RSI). However, these indices are designed to predict for calcium carbonate scaling only [12]. Tertiary treatment of treated municipal wastewater is necessary to reduce its scaling and corrosion potential for use in cooling tower systems [11]. Various types of tertiary treatment for reclaimed wastewater include activated carbon adsorption and ion-exchange process [13]. Reverse osmosis (RO) membrane is one of the suitable options for improving quality of reclaimed wastewater up to the standard used in a cooling tower. RO membrane is operated under high pressure and permeate pass through the semi-permeable membrane. It is an effective method for removing contaminants from feed water as large molecules and ions could not pass through a membrane. Disadvantage of RO membrane is membrane fouling. Membrane fouling during particle filtration occurs through a variety of mechanisms, including internal pore clogging by contaminants, coverage of pore entrances, and deposition on the membrane surface. Each of these fouling mechanisms results in a decline in the observed flow rate over time and the decrease in filtration efficiency [14]. Nevertheless, using a membrane is an advance on water or wastewater treatment due to its efficiency in solid removal, effluent disinfection, and keep permeate quality constant. The objectives of this research were to study (1) the possibility of using reclaimed municipal wastewater in cooling tower by RO membrane; (2) membrane fouling and membrane resistance; (3) corrosion index in terms of LSI and RSI; (4) cycles of concentration, blown down, and makeup water in cooling tower, and (5) cost analysis.

2. Materials and methods

2.1. Treated municipal wastewater characteristics

Wastewater was generated and collected from the various activities in a selected department store. The quantity of wastewater generated by the department store was in the range of 1,100–1,200 m^3/d. Activated sludge treatment process was used for the treatment of wastewater. The effluent wastewater characteristics are shown in Table 1. Some of the effluent wastewater (about 350 m^3/d) was treated further to be used in cooling tower by using sand filtration, carbon filtration followed by UF membrane process. Fig. 1 shows the schematic diagram for UF currently used and RO to treat the wastewater proposed by this study. The treated municipal wastewater after UF membrane was characterized according to the parameters listed in Table 1. This treated municipal wastewater was then used as influent for RO membrane experiment conducted in this study. The treated municipal wastewater will later be referred to as treated wastewater.

2.2. RO membrane experiment

Thin film RO membrane was used in this study. It was made from polyamide materials with an average pore size of 0.001 μm and effective membrane area of 7.9 m^2. The membrane was operated by 1 min running and 20 s pause at initial permeate flux of 6.3 m^3/d. Water temperature feed to the RO membrane was approximately 31.9°C. To observe the membrane fouling potential, TMP value was recorded every hour. The permeate was characterized by pH, conductivity, TDS, hardness, alkalinity as $CaCO_3$, iron (Fe^{3+}), chloride (Cl^-), silica, and phosphate (PO_4^{3-}).

2.3. Membrane fouling

Several models [16–20] have been developed to describe the membrane fouling during constant pressure filtration process. Among them, three kinds of blocking models (complete blocking, standard blocking, and intermediate blocking) and the cake filtration model are described by Eqs. (1)–(4), respectively.

Table 1
Average value of each treated wastewater characteristics

Parameter	Water quality standard for cooling tower [15]	Wastewater characteristics after each treatment process: Activated sludge + sand + carbon filtration	Wastewater characteristics after each treatment process: UF membrane
pH	7–9	7.3	7.3
Conductivity, µS/cm	<3,000	1,134.6	1,065.8
Total dissolved solids (TDS), mg/L	<2,000	745.7	687.1
Hardness, mg/L	<300	156.5	131.5
Alkalinity, mg/L as $CaCO_3$	<1,000	257.7	232.2
Iron, mg/L	<1.0	0.12	0.06
Calcium, mg/L as $CaCO_3$	–	73.53	56.63
Chloride, mg/L	<300	139.4	127.6
Silica, mg/L	<120	15.9	14.2
Phosphate, mg/L	<20	3.4	2.9

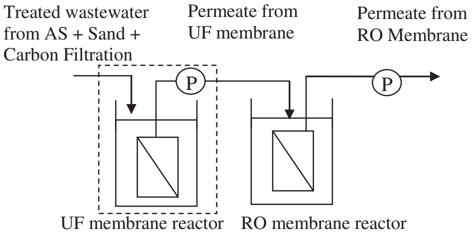


Fig. 1. Schematic diagram for the proposed wastewater treatment process: the unit in the box is currently used by the department store.

These equations have been commonly used to analyze the constant pressure dead-end membrane filtration process for MF [21], UF [22], and even NF/RO [23].

$$\frac{d(V/A)}{dt} = J_0 - k_b\,(V/A) \tag{1}$$

$$\frac{t}{V/A} = \frac{1}{J_0} + \frac{k_s \cdot t}{2} \tag{2}$$

$$\frac{dt}{d(V/A)} = \frac{1}{J_0} + k_i t \tag{3}$$

$$\frac{t}{V/A} = \frac{1}{J_0} + \frac{k_c}{2}(V/A) \tag{4}$$

where t is the filtration time, V is the cumulative permeate volume, A is the effective filtration area, J_0 is the initial flux, k_b, k_s, k_i, k_c (i.e., the well known modified fouling index) are the coefficients of complete blocking, standard blocking, intermediate blocking, and cake filtration models, respectively.

From these equations, four data plots have been proposed, where the linearity of the filtration data in the plot of $d(V/A)/dt$ and V/A, $t/(V/A)$ and t, $dt/d(V/A)$ and t, and $t/(V/A)$ and V/A offers proof of the complete blocking, standard blocking, intermediate blocking, and cake filtration model, respectively [22]. In these equations, V/A in place of V is used in order to compensate for the effects of the different membrane areas in this study.

2.4. Membrane fouling resistance

Membrane fouling resistance was analyzed using Darcy's law as shown in Eq. (5).

$$R_t = \frac{\text{TMP}}{\mu \cdot J} \tag{5}$$

where R_t = total membrane fouling resistance (m^{-1}); TMP = transmembrane pressure ($kg/m/s^2$); μ = permeate viscosity (kg/m/s); J = Permeate flux ($m^3/m^2/s$).

2.5. Scaling and corrosion index

In order to study the stability of water quality, LSI [24] and RSI [25] were applied to estimate the tendency toward $CaCO_3$ precipitation. LSI is a measure of a solution's ability to dissolve or deposit calcium carbonate, and is often used as an indicator of corrosive level of water. Calculation of LSI is shown by Eqs. (6) and (7).

$$\mathrm{LSI} = \mathrm{pH_a} - \mathrm{pH_s} \tag{6}$$

where $\mathrm{pH_a}$ is the actual pH of water and $\mathrm{pH_s}$ is the pH of water that has been saturated with calcium carbonate.

$$\mathrm{pH_s} = (9.3 + \mathrm{A} + \mathrm{B}) - (\mathrm{C} + \mathrm{D}) \tag{7}$$

where A, B, C, and D are water temperature (°C), TDS (mg/L), hardness (mg/L), and alkalinity (mg/L $CaCO_3$), respectively.

RSI has its basis in the concept of saturation level. RSI attempts to quantify a relationship between $CaCO_3$ saturation and scale formation as shown by Eq. (8).

$$\mathrm{RSI} = 2\mathrm{pH_s} - \mathrm{pH_a} \tag{8}$$

2.6. *Makeup water requirement for cooling tower*

Makeup water requirement for cooling tower was calculated based on windage rate, evaporation rate, cycle of concentration, and blow down. In this experiment, the cooling tower was composed of 14 cells and had approximately 14,000 m^3/d water recirculation in the system. The windage rate was set up at 0.05% while different exchange water temperature was set at 10°C. Windage, evaporation, cycles of concentration (or cycles), blow down, and makeup water were calculated using Eqs. (9)–(13), respectively.

$$\text{Windage } (\mathrm{m^3/d}) = \frac{\text{Water recirculation } (\mathrm{m^3/d}) \times \text{Windage rate } (\%)}{100} \tag{9}$$

$$\begin{aligned}\text{Evaporation } (\mathrm{m^3/d}) = {} & 0.00085 \times 1.8 \\ & \times \text{Water recirculation } (\mathrm{m^3/d}) \\ & \times \text{Diff. temp. } (^\circ\mathrm{C})\end{aligned} \tag{10}$$

$$\text{Cycles of concentration} = \frac{\text{Blow down concentration}}{\text{Makeup concentration}} \tag{11}$$

$$\text{Blow down } (\mathrm{m^3/d}) = \frac{[\text{Windage } (\mathrm{m^3/d}) \times (\text{Cylces} - 1)] - \text{Evaporation } (\mathrm{m^3/d})}{(1 - \text{Cylces})} \tag{12}$$

$$\begin{aligned}\text{Makeup } (\mathrm{m^3/d}) = {} & \text{Evaporation } (\mathrm{m^3/d}) \\ & + \text{Windage } (\mathrm{m^3/d}) \\ & + \text{Blow down } (\mathrm{m^3/d})\end{aligned} \tag{13}$$

3. Results and discussion

3.1. *Monitoring of TMP and membrane resistance*

The factor, which has the greatest influence on the RO membrane operation, is membrane fouling. Membrane fouling is caused by particles, colloidal materials, and mineral scaling results in permeate flux decline, and shorten membrane life due to possible membrane damage and harsh chemical cleanings employed for scale removal [26,27]. Fouling leads to increased TMP and decreased permeate flux with time. Fig. 2 shows the variation of TMP and permeate flux with time found in this study. It was observed that from 1 to 50 h of membrane operation time, permeate flux and TMP were almost constant. During 50–90 h, TMP slightly increased to 121.8 psi and after that it increased up to 127 psi. After 150 h, permeate flux decreased from 6.3 to 4.8 m^3/d and seemed to decrease continuously. High recovery RO is often limited due to biological fouling [28,29], organics [30–32], colloidal [33,34], and membrane fouling [35]. During the experiment of 150 h, permeate flux reduction rate was 0.009333 m^3/h.

The variation of permeate flux and membrane fouling resistance with time is as shown in Fig. 3. It was found that during 1–90 h, R_t continuously increased from 1.27E + 12 to 1.46E + 12 m^{-1}. During 100–150 h, the rapid increase of R_t was observed. It increased from 2.20E + 12 to 3.34E + 12 m^{-1}. The increase of R_t could indicate that the membrane was fouled. The fouling mechanism was quite complicated due to the various components of feed water.

In order to predict the RO membrane fouling, it was calculated based on complete blocking model,

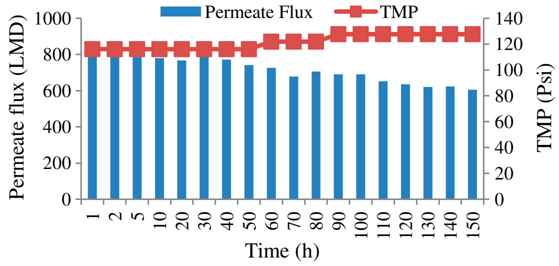


Fig. 2. Change of permeate flux and TMP with time.

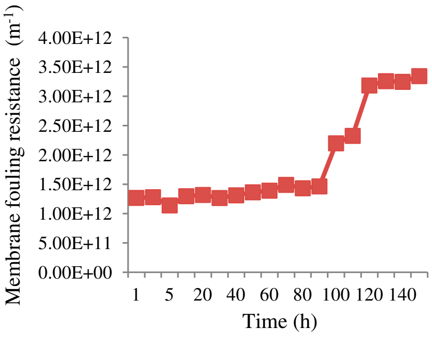


Fig. 3. Membrane fouling resistance.

standard blocking model, intermediate blocking model or cake layer formation. According to Eqs. (1)–(4), fouling mechanism on the RO filtration process is as shown in Fig. 4. The figure shows that complete blocking model gives a good linearity ($R^2 = 0.9990$) whereas standard blocking and cake layer models fit the data to a less extent ($R^2 = 0.9671$ and $R^2 = 0.9550$, respectively). Nevertheless, there was no relationship on intermediate blocking model ($R^2 = 0.2620$).

To observe complete blocking, standard blocking and cake layer formation models are consistent with the fact that the effective pore size of membrane was only 0.001 µm and the feed wastewater contained high concentrations of particulates and ions. Hence, the particulates and ions could accumulate on the membrane surface or block inside the pore of RO membrane. After the pore was completely blocked, the particles and ions could not pass through the membrane and could attach to the membrane surface. Then, cake layer formation occurred.

Membrane autopsy using scanning electron microscope (SEM: Hitachi version S-3400 N) could investigate the characteristic of membrane surface. The dried membranes (these were used for 150 h and underwent cleaning to recover the membrane flux) were cut at about 3 mm and pre-coated with a thin layer of gold prior to SEM analysis. The specimens were analyzed in a short period of time in order to protect the membrane destruction from a high-energy beam of electrons. The SEM results showed that carbon and oxygen were the main components on membrane as the membrane was made from synthetic materials. There were no particles adsorbed/attached on the membrane surface except particles of iron (0.83% wt) as shown in Fig. 5(a) and (b). This would be due to the high concentration in the feed flow and the cleaning process may not completely remove the adsorbed/attached iron on the membrane surface.

3.2. Permeate quality

The results of permeate quality with time is shown in Fig. 6. It could be seen that most particles and colloids were removed by RO membrane process. Compared to the feed wastewater characteristics as shown in Table 1, an average conductivity of RO permeate was

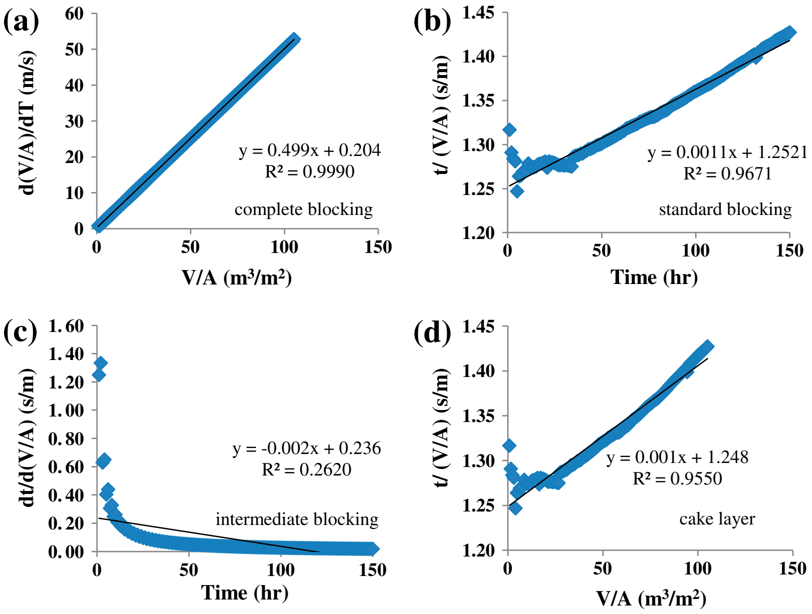
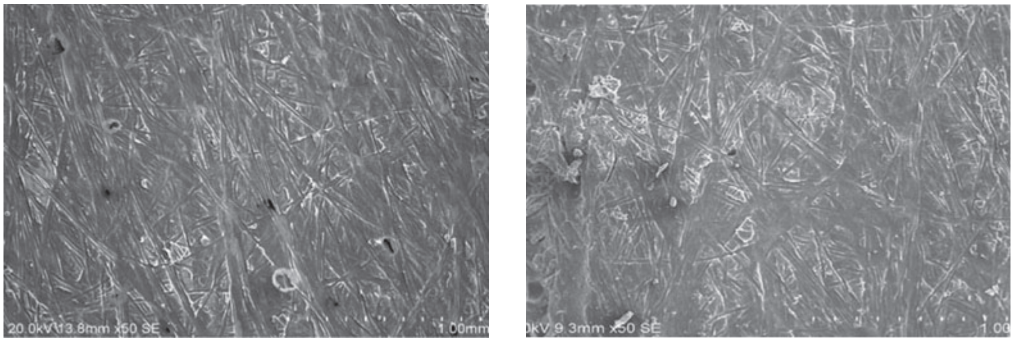


Fig. 4. Membrane fouling model. (a) complete blocking, (b) standard blocking, (c) intermediate blocking, and (d) cake layer.



(a) upper side of feed flow membrane

(b) lower side of feed flow membrane

Fig. 5. Surface of membrane after cleaning.
Note: (a) Main element consisted of carbon 80.75% wt, oxygen 18.42% wt, and iron 0.83% wt. (b) Main element consisted of carbon 80.75% wt, oxygen 18.42% wt, and iron 0.83% wt.

only 36.3 μS/cm. TDS, hardness, alkalinity, iron, calcium, chloride, and silica were also less in the permeate, with average concentrations of 42.3, 4.4, 22.7 mg/L as $CaCO_3$, 0.002, 0.05 mg/L as $CaCO_3$, 1.5, and 2.1 mg/L, respectively. There was no phosphate found in the permeate, and this may be due to the larger molecule of phosphate compared to other ions (molecular weights of phosphate, iron, calcium, chloride are 94.97, 55.85, 40.08, 35.43, and 28.09, respectively). Percent rejections of these particles and ions were in the range of 90.2–99.8% except for silica where the rejection was only 85.1%. The percent rejections were calculated using the UF treated wastewater as a basis. It could be concluded that permeate quality passed the standard criteria of makeup water quality in Thailand. However, pH of the permeate was lower than the pH of the feed water. From the experimental results, an average pH of the permeate was 6.7 while pH of the feed water, 7.3. It indicated the acidic nature of the permeate. This could be due to the fact that dissolved gases such as CO_2 could pass through the membrane and showed in the dissolved form in the permeate [36]. Therefore, additional treatment is required in order to produce non-corrosive and mildly alkaline water before being pumped as makeup water [37].

3.3. Corrosion index

LSI and RSI were calculated based on Eqs. (6) and (8). LSI values of the treated wastewater ($LSI_{\text{treated wastewater}}$) and the permeate ($LSI_{\text{permeate}}$) were −0.59 and −5.0 (Table 2). These indicate that no potential for scaling formation and $CaCO_3$ would be in the dissolved form based on the LSI values. Therefore, RO membrane could make treated water not corrosive and protect coatings of pipelines and equipment. However, considering RSI index, it was found that both the treated wastewater and the permeate showed the potential of corrosion. RSI of the permeate, RSI_{permeate} of 17.0, was larger than RSI value of the treated wastewater, $RSI_{\text{treated wastewater}}$ of 8.5 (Table 2). In fact, water with high corrosive property could not be transported in metallic pipes [38]. However, during the evaporation process of cooling tower, CO_2 could possibly be released from the water and the corrosion problem could be reduced [39].

3.4. Cycles of concentration

Loss of water in cooling tower in terms of windage and evaporation was calculated using Eqs. (9) and (10), respectively. The results are listed in Table 3. Loss of water recirculation from windage and evaporation resulted in higher dissolved solids concentration and the blowdown of water was required. In order to calculate the cycles of concentrations, hardness concentration was the key factor of treated wastewater while alkalinity concentration was the key factor of permeates to be substituted in Eqs. (14) and (15). Then, the cycles of concentration were 2.3 cycles for the treated wastewater while permeate were 44 cycles.

$$\text{Cycles}_{\text{treated wastewater}} = \frac{300\ \text{mg/L of Hardness}_{\text{standard makeup water quality}}}{131.5\ \text{mg/L of Hardness}_{\text{treated wastewater}}} = 2.3 \tag{14}$$

$$\text{Cycles}_{\text{permeate}} = \frac{1{,}000\ \text{mg/L of Alkalinity}_{\text{standard makeup water quality}}}{22.7\ \text{mg/L of Alkalinity}_{\text{permeate}}} = 44 \tag{15}$$

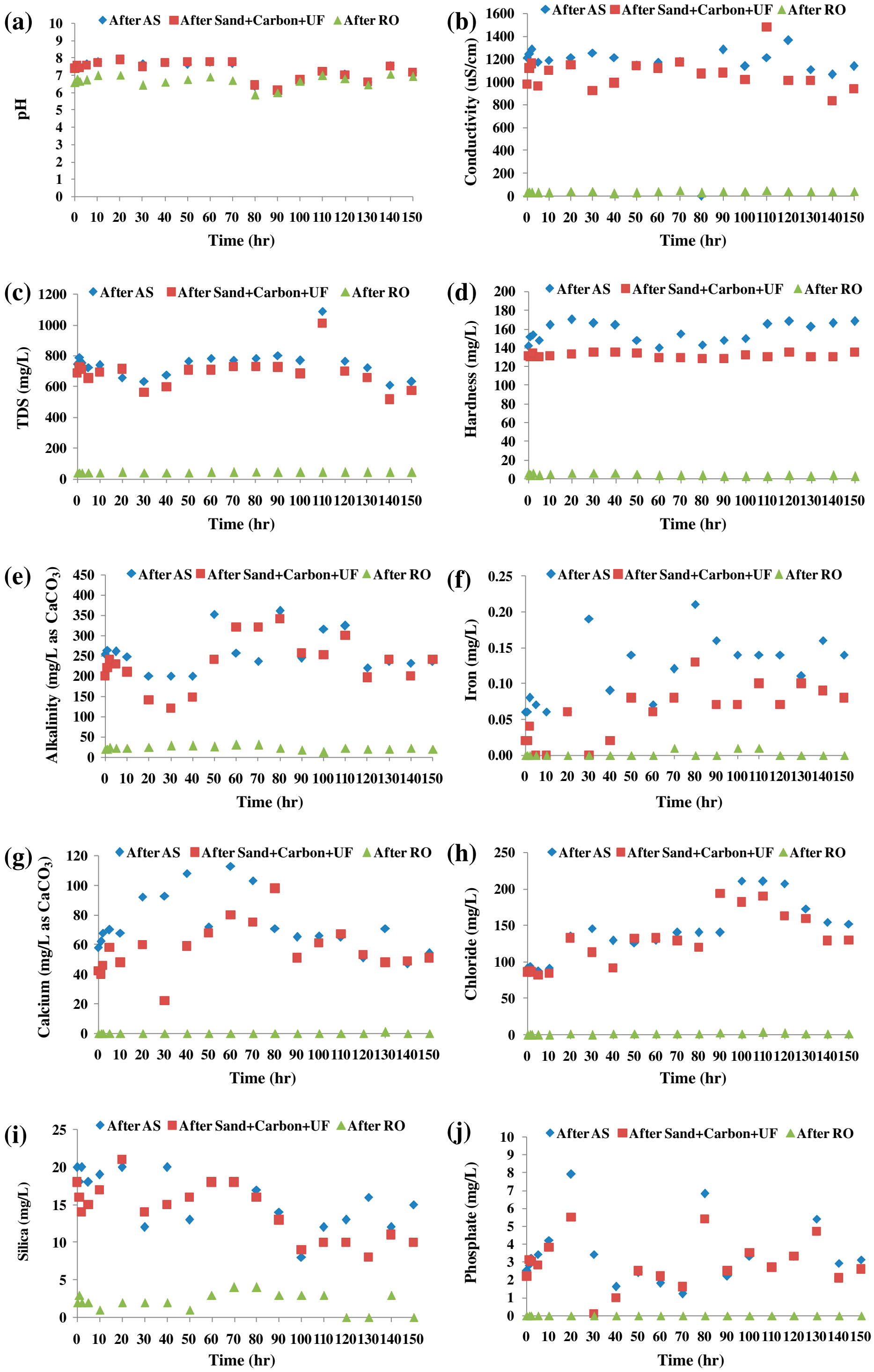


Fig. 6. Permeate quality during the operation period. (a) pH, (b) Conductivity, (c) TDS, (d) Hardness, (e) Alkalinity, (f) Iron, (g) Calcium, (h) Chloride, (i) Silica, and (j) Phosphate.

The blowdown required for the treated wastewater and the permeate were calculated using Eq. (12) and the results are shown in Table 3. Comparing to using the permeate, using treated wastewater (containing more impurities than the permeate) resulted in less cycles of concentration, and much more amount of blowdown was required. The blow down of 157.8 m^3/d was required when using the treated wastewater as makeup

Table 2
Comparison of LSI and RSI value

Index	Saturation pH	Treated wastewater	Permeate
LSI [40]	LSI (+); $CaCO_3$ precipitation likely, scale can form LSI (−); no potential for scaling, $CaCO_3$ will dissolve LSI (0); stable water. If the temperature of water change, scaling may occur	−0.59	−5.0
RSI [41]	RSI < 6; the scale tendency increase as the index decrease RSI > 7; the calcium carbonate formation probably does not lead to a protective corrosion inhibitor films RSI > 8; mild steel corrosion becomes an increasing problem	8.5	17.0

Table 3
Makeup water requirement when using the UF treated wastewater and RO permeate

Parameter	UF treated wastewater	RO permeate
Windage, m^3/d	7.0	7.0
Evaporation, m^3/d	214.2	214.2
Blowdown, m^3/d	157.8	0.0
Makeup water, m^3/d	379.0	221.2

water whereas the blow down was not necessary when the permeate was used. Clearly, this is because the water quality of the permeate was better. Finally, the quantity of makeup water was calculated using Eq. (13). It was found that using the treated wastewater required 379.0 m^3/d of makeup water whereas using the permeate required only 221.2 m^3/d equivalent to only 41.6% of the makeup water used when using the treated wastewater.

3.5. Cost analysis

To compare an advantage of treating water further with RO membrane using the conventional UF membrane system, cost is one key factor that influences on making the decision. The capital cost of RO membrane was approximately 2,000,000 THB (capacity of permeate production at 300 m^3/d) and its operating cost calculated based on electricity usage only was 774.2 THB/day or 23,226 THB/month. Since having the RO membrane could save makeup water of 157.8 m^3/d and the makeup water used in the conventional UF system cost 35 THB/m^3, the payback period of having the RO membrane was only 14 months as shown by Eq. (16).

$$\begin{aligned}\text{Payback period}\\ &= \frac{\text{Amount to be initially invested}}{\text{Estimated Annual net Cash Inflow}}\\ &= \frac{2,000,000\ \text{THB}}{(165,690\ \text{THB/month} - 23,226\ \text{THB/month})}\\ &= 14\ \text{month}\end{aligned} \tag{16}$$

Note: RO membrane system required the electricity of 1.25 kW/m^3 permeate; based on the electricity cost of 3.5 THB/kW.

4. Conclusions

In order to use reclaimed municipal wastewater in a cooling tower, RO membrane process could be used to treat the water. After the RO membrane process, permeate quality could pass the standard criteria of makeup water quality in Thailand. The permeate contained low concentrations of colloidal and ions which yielded in more cycles of concentration than the conventional UF-treated water. This also led to the lower makeup water demand. However, there is an issue of membrane fouling. From the results, it was found that the main membrane fouling mechanism for RO membrane was complete blocking, which then was followed by standard blocking and cake layer formation, respectively.

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