The Effect of Concentration Staging on Performance of Produced Water treatment using Forward Osmosis

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Abstract. During oil and gas production, a large amount of produced water (PW) is generated, which would be harmful to the environment if not treated properly. Treatment of such wastewater requires multi-processing units and typically includes fouling prone pressure driven membranes. Forward osmosis (FO) seems promising due to its high salt rejection, high total dissolved solid removal, more reversible fouling type, does not require external pressure to operate and potentially can be used as a standalone unit with minimum pretreatment. However, FO still suffers from low permeate flux, high reverse solute flux (RSF) and membrane fouling, although not as severe as in pressure driven membrane. Since FO depends largely on osmotic pressure difference, the effect of concentration staging was investigated. It was found that multi-stage concentration was able to achieve up to 80% water recovery due to sufficient osmotic pressure.

1. Introduction
Produced Water (PW) is wastewater from oil and gas production partly from water flooding process in during enhance oil recovery (EOR) [1]–[3]. PW is the major waste stream produced from oil and gas industry [4]. PW generally consists of various hydrocarbon, organic and inorganic matters, dispersed oil and various solutes [4]. It must be treated properly due to the high level of contaminants present in it.

Conventional treatments offer its own advantages and disadvantages. For example, while adsorption is able to remove heavy metals, benzene, toluene, ethylene and xylene and dispersed oil, suspended particles may plug the adsorbent media hence reducing its efficiency overtime [5]–[7]. Regeneration of absorbent media requires usage of chemicals in which requires an extensive treatment for the chemicals used. Another conventional treatment includes evaporation pond which has been used for both onshore and offshore treatment. However, it requires a relatively large space to function efficiently [8]. In short, conventional PW treatment faces a few challenges such as high treatment cost, usage of toxic chemicals, requires large installation space and generation of secondary waste [7]. To solve these problems, membrane separation technologies have been continuously researched.

Common membrane based technology includes the usage of pressure driven membrane such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). These membranes use differential pressure as driving force to facilitate mass transport. The degree of particle separation depends on pore size of the membrane. Example, MF has the largest pore (0.1-3μm) and is used to remove suspended solids while RO has the smallest pore (about 0.1nm) and is used to remove
salts [7], [8]. However, pressure driven membrane are prone to membrane fouling [9], [10]. The usage of high pressure tends to have an irreversible fouling effect on the membrane [11].

Another alternative for membrane separation technology is forward osmosis (FO). FO is an emerging technology and have been investigated in PW treatment [12]–[14]. In contrary to pressure driven membrane, FO relies on osmotic pressure difference between two solutions; the feed solution (FS) and the draw solution (DS) where the DS has the highest osmotic pressure to facilitate mass transport. FO results in dewatering and concentrating the feed while diluting the DS.

In the treatment of PW, FO offers a variety of advantages. Firstly, FO does not require any external pressure to operate unlike in RO since it utilizes osmotic pressure difference between two solutions. The absence of high external pressure promotes lower energy consumption and reduce fouling tendency [15]. It also renders the fouling type to be more reversible [11] and therefore a simpler cleaning method such as osmotic backwashing can offer complete flux recovery [16]. As such, FO can potentially operate as a stand-alone unit without the use of pretreatment, unlike NF and RO [15]. Moreover, FO is reported to have high salt rejection up to 96% while operating at ambient condition [15]. FO can also treat hyper saline wastewater (>65 bars) in which RO is unable. However, FO also have a number of setback such as lower water flux (depending on osmotic pressure difference) [17], reverse salt flux due to concentration polarization [18] and membrane fouling (despite of its reversibility), although it is not as severe as in NF or RO [10 - 11].

Since FO relies much on osmotic pressure difference, this study aims to investigate the effect of concentration staging performance on the treatment of PW via FO. Concentration staging will be compared based on single-stage and multi-stage concentration. Rejection analysis was also done for both single-stage and multi-stage concentration.

2. Materials and Method

2.1. List of Chemicals and Materials
PW feed solution (FS) was obtained from a petroleum production well in peninsular Malaysia. The characteristic of the PW sample is summarized in Table 1. Note that the conductivity of the feed sample is 2 mS/cm. Control feed was also used as to mimic the PW in terms of osmotic pressure. To produce such solution, sodium chloride (NaCl) was added to deionized (DI) water to obtain around 2000 µS/cm. The control feed was used to measure the performance of FO without any fouling agent while still having similar osmotic pressure of the feed sample. The control feed was used mainly in the study on effect of staging. Seawater was used as the DS and was obtained locally from Teluk Batik, Perak, Malaysia. It has a conductivity of 58 mS/cm and a pH of 6.5. The large difference in conductivity between the feed and the DS ensures sufficient driving force for FO.

2.2. Experimental Setup
Figure 1 shows the schematics for the experimental setup. The feed is in direct contact with the DS through the FO membrane where both are kept under ambient condition. The membrane was kept on AL-
FS (active layer facing feed solution) mode to reduce the effect of fouling [19]. The membrane area was $3.4 \times 10^{-3}$ cm$^2$. Two Seko PR7 Peristaltic Dosing Pumps were used to recirculate both FS and DS to the membrane module at 150 ml/min. The FS was kept on a Mettler Toledo MS 3002S/01 weighing balance to record the changes of the weight which was later used to determine the flux according to equation (1). The weigh balance was connected with a computer for data logging through Mettler Toledo Serial-Port-to-Keyboard software. All of the data were logged and computed in Microsoft Excel.

$$J = \frac{\Delta V}{A \Delta t}$$  \hspace{1cm} (1)

where $J$ is permeate flux in L/m$^2$h, $V$ the volume of the feed (L), $A$ the area of the membrane (m$^2$) and $t$ the time (h).

2.3. Effect of Staging: Single-stage concentration vs Multi-stage Concentration

For both experiments, a TFC-PA membrane from Toray was used. The objective was to concentrate PW from 300 ml to 60 ml. The initial ratio of feed to DS was kept at 1:1. Particularly, for single-stage concentration, the same DS (taken at once) was used throughout the experiment. However, for the multi-stage concentration, the replenishment of DS was introduced. This was done stage by stage i.e.; for each 60 ml decrement of PW FS, the DS is replenished with fresh 300 ml DS. The replenishment of DS was necessary to provide enough driving force for FO process. The process continues until PW reached 60 ml, leading to 4 stages of DS replenishment.

2.4. Rejection Analysis

The rejection analysis was conducted for both experiment. Several parameters were gauged which were conductivity, pH value, total oil and grease (TOG), turbidity, chemical oxygen demand (COD), total nitrogen (TN) content, total phosphorus (TP) content. These parameters were measured by using conductivity meter (Eutech Instrument CON 700), pH meter (Eutech Instrument pH 700), total oil in water analyzer (Turner Design TD-500D), turbidity meter (Hach 2100Q Turbidity Meter), and Hach-Lange kits for COD, TN and TP respectively.

3. Results and Discussion

3.1. Single-stage concentration vs Multi-stage Concentration

Based on Figure 2, for the single-stage concentration, the flux for control feed was in the range of started from 16.27 L/m$^2$h and declined to 6.07 L/m$^2$h. The final conductivity value for both control feed and DS were 8.14 mS/cm and 48.7 mS/cm respectively. However, when using real PW as the feed, the flux was declining much further from 9.49 L/m$^2$h to 0.39 L/m$^2$h. Furthermore, the amount of water that was able to recover was only up to 56.67%, instead of 80% when using control feed. At the end of the experiment, it was found that the conductivity for PW and draw solution were 31.4 mS/cm and 31.8 mS/cm. The finding suggested that the draw solution was not able to draw water from the feed. Prolonged operation resulted in balanced water transport. This effect can also be explained by the effect...
fouling, more specifically cake enhanced concentration polarization (CECP) [20]. The gap of flux between the control feed and PW suggested that fouling occurred on the membrane, thus reducing the effective area of the membrane. Moreover, RSF occurred due to concentration polarization. Therefore, the osmotic pressure shifted towards the membrane side that was facing the feed. Consequently, the osmotic pressure differences reduced and permeate flux decreased.

For multi-stage concentration, replenishment of draw solution was done for every 60 ml reduction of the feed. Based on Figure 3, the flux for control feed started from 20.15 L/m²h and declined to 8.01 L/m²h, which was more than that of single-stage concentration. The final conductivity for control feed and draw solution were 8.46 mS/cm and 48.4 mS/cm respectively. When real PW was used as the feed solution, the permeate flux obtained was decreased from 15.48 L/m²h to 1.72 L/m²h. Indeed, fouling also occurred in multi-stage concentration, which explained the flux gap between control feed and PW besides osmotic dilution of the draw solution. However, multi-stage concentration was able to achieve 80% water recovery due to sufficient osmotic pressure difference provided by replenished draw solution.

3.2. Summary of Rejection Analysis

Table 2 shows the summary of the rejection analysis done for both single-stage concentration and multi-stage concentration. Note that for multi-stage concentration, the rejection analysis was done at the
final stage \((n=4)\). The parameters of the DS for both single-stage concentration and multi-stage concentration were almost similar and both have meet the demands of standard B discharge effluent.

4. Conclusion

This study investigates the effect of concentration staging where multi-stage concentration outperformed single-stage concentration. However, fouling still persist despite the claims of FO membrane having lower fouling tendency. Further study on membrane fouling control strategy is thus required.

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Table 2. Summary of Rejection Analysis on Draw Solution

| Parameter                  | Single-stage | Multi-stage \((n=4)\) |
|----------------------------|--------------|-----------------------|
| Conductivity (µS/cm)       | 31876        | 47121                 |
| pH                        | 6.5          | 6.5                   |
| COD (mg/L)                | 230          | 220                   |
| TOG (mg/L)                | 7            | 7                     |
| Total Nitrogen (mg/L)     | 0            | 0                     |
| Total Phosphorus (mg/L)   | 1.3          | 1.2                   |
| Turbidity (NTU)           | 1.47         | 1.13                  |
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