Performance evaluation of reverse osmosis (RO) pre-treatment technologies for in-land brackish water treatment

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HIGHLIGHTS

• PV-RO as an emerging desalination technology treating in-land brackish water
• Filmtech membrane filtration performance was better than Hydranautics membrane.
• FO is an effective pretreatment minimizing RO membrane cleaning frequencies.
• MgCl₂ as draw solution performed better than NaCl due to its divalent structure.

ABSTRACT

Integration of renewable energy with desalination technologies has emerged as an attractive solution to augment fresh water supply sustainably. Fouling and scaling are still considered as limiting factors in membrane desalination processes. For brackish water treatment, pre-treatment of reverse osmosis (RO) feed water is a key step in designing RO plants avoiding membrane fouling. This study aims to compare at pilot scale the rejection efficiency of RO membranes with multiple pre-treatment options at different water recoveries (30, 35, 40, 45 and 50%) and TDS concentrations (3500, 4000, and 4500 mg/L). Synthetic brackish water was prepared and performance evaluation were carried out using brackish water reverse osmosis (BWRO) membranes (Filmtec LC-LE-4040 and Hydranautics CPA5-LD-4040) preceded by 5 and 1 μm cartridge filters, 0.02 μm ultra-filtration (UF) membrane, and forward osmosis (FO) membrane using 0.25 M NaCl and MgCl₂ as draw solutions (DS). It was revealed that FO membrane with 0.25 M MgCl₂ used as a draw solution (DS) and Ultra-filtration (UF) membrane followed by Filmtec membrane gave overall 98% rejection but UF facing high fouling potential due to high applied pressure. Use of 5 and 1 μm cartridge filter prior to Filmtec membrane also showed effective results with 95% salt rejection.

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1. Introduction

Energy and drinking water supply remain unsolved issues for many countries around the world [1]. Therefore, integration of desalination technologies with renewable energy has become an attractive solution to overcome water scarcity problem [2]. Major part of the world’s water is seawater, brackish water and groundwater. Approximately, 97.4% of the entire water available on earth is salty and 1.984% is located in the ice caps and glaciers, while 0.592% is located as groundwater and only 0.014% of the earth’s water is available as fresh water [3]. Many water-stressed countries are supplementing their fresh water supply with desalinated water to meet their increased water demand caused by population growth, rapid urban sprawl, agriculture development, industrialization, and tourism [4].

Inland salinity of ground water having total dissolved solids (TDS) of varying concentration, usually below 10,000 ppm, has been found in four provinces of Pakistan. For example, in the Punjab most ground water has TDS < 1500 while in Balochistan it typically exceeds 3000 mg/L (see Fig. 1) [5].

However, these areas also receive 5.1–6.2 kWh/m²/day of annual average mean daily solar radiation, making photovoltaic electricity an attractive solution to conventional energy to fulfill their water requirements via desalination [6]. Treatment of brackish water using desalination technologies is an effective option to overcome fresh water scarcity problem [7]. Brackish water desalination represents over 21% of the total worldwide desalination capacity due to its low operating cost and energy requirement [8]. Reverse osmosis (RO) is a water treatment technology that has gained world-wide acceptance. Over the years, remarkable advancement has been made in RO technology [9–11]. Recent study revealed that reverse osmosis (RO) is the most optimized technology for water desalination related activities [12].

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Membrane scaling and fouling are among the most serious concerns in membrane-based treatment processes. In brackish/sea water desalination process, pre-treatment of the saline feed is a crucial step in designing of the process to avoid membrane fouling and scaling and to reduce its cleaning frequency [13]. Proper pre-treatment is an essential aspect in design process via reverse osmosis technology for successful plant operation to ensure treatment performance [14–16].

Compared to conventional pre-treatment technologies, membrane technologies were found to be more cost effective and give better results by removing the particles having size greater than the pore size of the membrane. This results in low silt density index (SDI) value, which make them more attractive pre-treatment technologies for the water having high total dissolved solids (TDS) [17–20]. Among membrane processes, micro-filtration (MF) and ultra-filtration (UF) are the technologies that have gained global acceptance as suitable pre-treatment technologies for saline water [21].

Ultra-filtration (UF) was found to be cost effective and efficient technique for the removal of suspended solids and bacteria [13]. The selection of the pre-treatment option is site specific and is mainly based on feed water quality, but in some cases the feed water quality is influenced by seasonal variation (i.e. flood, drought, and climatic impact) which make pre-treatment design more complicated. Forward osmosis (FO) was found to be a feasible pre-treatment option for variable quality feed water and for the feed having high fouling potential. It is capable of providing uniform treated water quality with less fouling potential instead of variable feed quality.

Forward osmosis is an emerging technology used in water reuse and desalination [22–25]. It is regarded as a natural process that utilizes osmotic pressure gradient to draw the water from the dissolved solutes in feed solution across a semi-permeable membrane [26–28]. Among the other RO pre-treatments, FO has much lower fouling tendency and can operate over a longer period of time without cleaning [29]. Increasing interest in FO is fueled by the global demand for less fouling, high recovery and low energy consuming process increasing lifespan of the membrane compared to the pressure-driven membrane process [30, 31].

Though UF and MF has been widely used in RO pre-treatment, the use and availability of FO is relatively recent. Side-by-side comparison of pre-treatment options is lacking. This study sets out to provide pre-treatment comparison for a stand-alone photovoltaic (PV) powered RO plant designed to meet the growing water needs of inland areas of Pakistan.

2. Materials and methods

2.1. System configuration

In this study, pilot-scale reverse osmosis plant was designed to investigate rejection of RO membranes at different water recoveries and TDS concentrations. To make process performance more effective and sustainable, multiple pre-treatment options were coupled prior to both (RO) membranes operated in parallel. The general layout of the pilot scale plant showing all components is shown in Fig. 2, while the actual picture of the system is depicted in Fig. 3.

A 2 kWh photovoltaic (PV) system consisting of eight monocrystaline silicon solar panels (model: CS6P-265M, Canadian Solar) connected with a grid inverter was installed to provide solar energy input. The specification of the photovoltaic (PV) system and modules is illustrated in Table 1.

The pilot-scale reverse osmosis (RO) unit consists of a feed tank, high pressure submersible feed pump (model: SQF 0.6-3, Grundfos, UK), clean-in-place (CIP) tank, and clean-in-place pump (model: MSP 230, Marchmay, UK) along with membrane modules comprising of two spiral wound RO membrane (Filmtec LC-LE-4040 and Hydranautics CPA5-LD-4040) in combination with different pre-treatment technologies comprising of 5 and 1 μm cartridge filter (CF), 0.02 μm pore size ultra-filtration (UF) membrane and a cellulose tri-acetate (CTA) flat sheet forward osmosis (FO) membrane. Permeate and feed flow rate were measured by rotameters and recycled to the feed tank to make operation continuous. Membrane inlet and outlet pressure was measured using bourdon gauge (model: 233.55 LBM, WIKA Instrument Corporation, USA) which was under the permissible limit recommended by...
the membrane manufacturers (Table 2). Feed and permeate TDS and pH were also measured with in-line meters.

2.2. Synthetic brackish water feed conditions

Filtration tests were performed using synthetic brackish water [2]. Three brackish water feed conditions of 3500, 4000, and 4500 mg/L TDS concentration were prepared in accordance to target feed water quality found in the substantial areas of Pakistan (Table 3) [5]. Sodium metabisulphite (Na2S2O5) at a concentration of 2 mg/L was also added to neutralize residual chlorine in the tap water and inhibit bacterial growth [32].

2.3. Operational conditions

The synthetic water was fed to RO membranes preceded by different pre-treatment technologies including 5 and 1 μm melt blown cartridge filter, 0.02 μm ultrafiltration (UF) membrane and forward osmosis (FO) membrane. For FO system, 0.25 M NaCl and MgCl2 were used as draw solutions (DS). Experiments were performed in batch recirculation mode and the flux across the FO membrane, active layer facing feed side was measured by digital data logging weight balance (UX 6200H, Shimadzu, Japan). The diluted DS was then fed to the RO membrane for the separation of clean water and regeneration of DS for reuse. System was operated for 2 h for each set of recoveries 30, 35, 40, 45, and 50% along with each pre-treatment option. For each TDS condition, the duration was sufficient enough to achieve steady-state condition [33].

\[
\% \text{ Rejection} = \left(1 - \frac{C_p}{C_f}\right) \times 100\% \tag{1}
\]

where \(C_p\) and \(C_f\) are permeate and feed concentration respectively (mg/L). Rejection value was calculated under each condition (after 2 h operation)

Water recovery from each membrane arrangement were measured using Eq. (2)

\[
\text{Water recovery} = \frac{Q_p}{Q_f} \times 100\% \tag{2}
\]

where \(Q_p\) and \(Q_f\) are the permeate and feed water flow rates respectively (L/h).

Flux across the RO membranes were calculated using Eq. (3)

\[
J_v = \frac{L_p}{\Delta P - \sigma \Delta \Pi} \tag{3}
\]

where \(J_v\) is the hydraulic permeate flux (L·h\(^{-1}\)·m\(^{-2}\)), \(L_p\) is the membrane permeability (L·h\(^{-1}\)·m\(^{-2}\)·bar\(^{-1}\)), \(\Delta P\) and \(\Delta \Pi\) are the transmembrane pressure and osmotic pressure (bar) respectively and \(\sigma\) is the local reflection coefficient.

The solute flux across the RO membrane is the sum of diffusive and convective flux. Therefore, mass transfer across the RO membrane can be expressed as Eq. (4) [33,34].

\[
J_s = J_v \cdot C_p = J_{\text{diff}} + J_v \cdot C_{\text{conv}} \tag{4}
\]

where \(J_s\) is the solute flux (mg·m\(^{-2}\)·s\(^{-1}\)), \(C_p\) is the solute permeate concentration (mg·L\(^{-1}\)), \(J_{\text{diff}}\) is the diffusive flux (mg·m\(^{-2}\)·s\(^{-1}\)) and \(C_{\text{conv}}\) is the solute permeate concentration due to convective transport. The above equation can also be re-written as.

\[
C_p = \frac{J_{\text{diff}}}{J_v} + C_{\text{conv}} = P_s \Delta C_s / J_v + C_{\text{conv}} \tag{5}
\]
where $P_s$ is the membrane solute permeability (m·h$^{-1}$), and $\Delta C_s$ is the concentration difference ($C_b - C_p$) across the membrane (mg·L$^{-1}$), and $C_b$ is the solute brine concentration.

After each run, forward washing of RO membrane was performed with treated water using clean in place (CIP) pump to avoid membrane surface deposition. Each pre-treatment option was evaluated using parameters including membrane inlet pressure, feed TDS, feed pH, membrane outlet pressure, permeate pH, and permeate TDS.

### Table 3
Concentrations used for synthetic feed preparation [2].

<table>
<thead>
<tr>
<th>Compounds</th>
<th>Amount (TDS-mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Feed I</td>
</tr>
<tr>
<td>NaCl</td>
<td>889</td>
</tr>
<tr>
<td>CaCl$_2$</td>
<td>941</td>
</tr>
<tr>
<td>MgCl$_2$·6H$_2$O</td>
<td>983</td>
</tr>
<tr>
<td>NaNO$_3$</td>
<td>45</td>
</tr>
<tr>
<td>Na$_2$SO$_4$</td>
<td>617</td>
</tr>
<tr>
<td>NaHCO$_3$</td>
<td>18</td>
</tr>
</tbody>
</table>

### 3. Results and discussion

Figs. 4 and 5 show the percent rejection performance of both RO membranes coupled with multiple pre-treatment options at different recoveries obtained against different trans-membrane pressure for the feed TDS condition i.e. 3500 mg/L. From the filtration tests with Filmtec membrane, minimum 95% salt rejection with cartridge filter as a pre-treatment (CF-RO) and 98% salt rejection with ultrafiltration as a pre-treatment (UF-RO) was observed with a maximum permeate TDS of 175 mg/L and 68 mg/L, respectively. While for the similar set of arrangement with forward osmosis as a pre-treatment (FO-RO), 93 and 96% salt rejection with NaCl and MgCl$_2$, respectively as DS was observed with a maximum permeate TDS of 234 and 120 mg/L, respectively (Fig. 4). In parallel, from the Hydranautics membrane, minimum 85% salt rejection with cartridge filter (CF-RO) and 97% salt rejection with ultrafiltration (UF-RO) as a pre-treatment was observed with maximum permeate TDS 500 and 98 mg/L, respectively. While with forward osmosis as a pre-treatment, minimum 77% rejection with NaCl and 96% salt rejection with MgCl$_2$ as DS was observed with a maximum permeate TDS of 775 and 130 mg/L, respectively (Fig. 5).

Ultrafiltration (UF) membrane followed by RO membranes showed effective salt rejection at low operating pressure as compared with CF-
RO and FO-RO. Both membranes showed consistent operational behavior with all the pre-treatment arrangements over a wide range of pressures applied in order to achieve high recovery except for FO-RO arrangement with NaCl as DS using Hydranautics membrane resulting in decline in rejection performance at higher pressure condition accompanied with significant increase in permeate TDS (Fig. 5). Moreover, the performance of CF-RO in terms of permeate TDS was also poor among the pre-treatment options tested.

For FO as a pre-treatment with NaCl as DS using Hydranautics membrane, a 4% decline in rejection at per bar increase in pressure was observed. MgCl₂ as a draw solution was more effective as compared to NaCl because of its high osmotic potential and divalent ionic structure. It was observed that the flux across the FO membrane has a direct relation with the molar concentration of draw solution (DS) and inverse with the feed solution (FS) concentration [35]. Higher molar concentration of DS resulted in higher flux across the FO membrane and ultimately concentrated FS and diluted DS which required high operating pressure for the regeneration of DS and separation of pure water.

Figs. 6 and 7 show the percent rejection performance of RO membranes for the feed condition II i.e. 4000 mg/L. Under feed condition II, the filtration test using Filmtec membrane showed minimum 95% salt rejection with the cartridge filter (CF-RO) and 97% salt rejection with ultrafiltration (UF-RO) as a pre-treatment was observed with a maximum permeate TDS of 186 and 110 mg/L, respectively (Fig. 6).

Increased feed water TDS inversely effects rejection performance of RO membranes with all the pre-treatment options. Major decline in rejections was observed in FO-RO with NaCl as DS followed by CF-RO combinations using Hydranautics membrane. The significant increase in permeate TDS in FO-RO with NaCl as DS was due to presence of mono-valent ionic structure of NaCl and consequently its poor rejection (Fig. 7).

Despite high rejection efficiency of UF-RO as compared with CF-RO and FO-RO (MgCl₂ as DS), UF being high pressure pre-treatment option may experience high fouling tendency over long term operation as compared to FO operated under natural osmotic concentration gradient. Zaviska et al. [29] reported that the fouling potential of UF membrane was higher as compared with FO membrane due to its high pressure application and less removal of scaling agents (i.e. sulfate and carbonate).

Figs. 8 and 9 show the percent rejection performance of RO membranes for the feed condition III i.e. 4500 mg/L. Under feed condition III, pressure drop and decline in flux across the UF membrane was observed over the passage of time for high TDS feed condition indicating its fouling characteristics. On the contrary, FO-RO arrangement with MgCl₂ as DS offered relatively lower pressure drop and sustained flux.
FO membrane operation offers limited deposition of scaling and fouling agents and extracts water under natural gradient which results in reversible, uncompact fouling on membrane surface due to concentration polarization only [29]. Furthermore, fouling on FO membrane surface does not significantly affect membrane flux due to its uncompact structure and can be easily removed by rinsing with di-ionized (DI) water [29].

Considering all three feed TDS conditions, the optimum water recovery was found to be 40% for Hydranautics membrane and 45% for Filmtec membrane. It was also observed that the initial permeability i.e., permeability for pure water (DI water) of both membranes decreased during the filtration test with brackish water, although no irreversible fouling was observed after each test with forward membrane flushing. Teychene et al. [33] reported that 30% decrease in permeability for Energy-Saving Polyamide-Boron (ESPAB) membrane while an average 10% decrease for other sea and brackish water membrane was observed. On average, our study revealed 13% decrease in permeability for Filmtec membrane whereas 10% decrease in permeability for Hydranautics membrane over two-hour operation due to concentration polarization on the membrane surface.

4. Conclusions

Ultrafiltration (UF) and forward osmosis (FO) were found as an effective pre-treatments with less fouling characteristic to avoid membrane cleaning frequencies but at high operating cost in term of high operating pressure for UF and MgCl₂ as DS for FO process. Operating cost of FO can be justified for the brackish water having complex constituents and varying concentrations posing high fouling tendencies. MgCl₂ as draw solution presented better results as compared to NaCl due to its divalent structure and osmotic potential. Filmtec membrane LC-LE-4040 provided better performance than Hydranautics membrane CPA5-LD-4040 over a wide range of pressure and TDS conditions. 40 and 45% recoveries for Hydranautics and Filmtec membranes, respectively were found as optimum values for all the feed conditions.

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