

Closed circuit desalination of O&G produced water: An evaluation of NF/RO performance and integrity

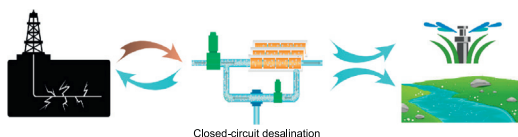
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GRAPHICAL ABSTRACT



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ABSTRACT

Reclamation and reuse of oil and gas (O&G) waste streams is being considered as an option to alleviate stress on freshwater supplies. While limited treatment is necessary to reuse O&G wastewater (e.g., produced water (PW)) for subsequent hydraulic fracturing, high removal of organic matter and total dissolved solids (TDS) is needed to enable broader reuse (e.g., irrigation, stream flow augmentation). This study focused on the use of closed circuit desalination with nanofiltration (NF) and reverse osmosis (RO) to remove TDS and organic matter from pre-treated PW. The performance and fouling propensity of three commercial NF and RO membranes was compared during the concentration of ~900 feed batches (440 h). Up to 99.6% TDS and 89% dissolved organic carbon (DOC) was removed, producing high quality permeate. Membrane characterization revealed minimal scaling and organic fouling of the three membranes, suggesting that closed circuit desalination (CCD) might be a promising method to facilitate reuse of PW.

1. Introduction

1.1. Water use in the O&G industry

Water scarcity and resource depletion are a growing concern as population grows and drought persists—magnifying stress on limited water supplies. Therefore, alternative water sources must be identified for sustainable development. One industry that has potential to substantially reduce the burden on freshwater supplies by implementing water treatment and reuse is the oil and gas (O&G) industry. Out of ~165 million m³ of water used each year in the U.S. for O&G well development, approximately 35% (60 million m³) returns to the surface as fracturing flowback and produced water (PW) and only 14% is reused [1]. Most water is disposed of via deep-well injection (i.e., Class II

disposal wells) for economic reasons, providing opportunities for PW reclamation and reuse if inexpensive treatment processes are developed [2].

Recently, more O&G operators are considering recycling of PW than in past years, mainly due to environmental concerns associated with deep-well injection (e.g., induced seismicity, groundwater contamination) and improved water transportation strategies that help reduce costs (e.g., temporary pipelines or planned infrastructure) [3]. Yet, depending on the target water quality, multiple treatment technologies may be required, making treatment of PW expensive and at this time economically unfavorable. Challenges associated with treatment of PW include high salinity (ranging from 1000 to 200,000 mg/L total dissolved solids (TDS)) and high concentrations of complex organic constituents (i.e., BTEX, oil and grease, hydrocarbons, and biopolymers)

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[4,5]. This water composition poses challenges to membrane filtration due to scaling potential (e.g., bromide, barium, calcium, iron, silica) and organic fouling, while salinity and organic matter pose challenges to biological treatment methods [6–8]. Therefore, flexible and robust treatment systems are necessary to withstand the harsh conditions and variable PW chemistry, and efficiently remove targeted constituents.

Various reuse applications for reclaimed PW require different water qualities. For example, reuse for hydraulic fracturing requires minimal treatment that focuses on the removal of hardness, scalants, and bacteria; while reuse for surface discharge or irrigation requires a higher level of treatment with removal of TDS and organic matter [9–11]. Reuse applications that return treated PW to the environment must take extra precaution to ensure consistent and predictable effluent/permeate quality, as the presence of some constituents (e.g., bromide, iodide, trace organics) might be harmful to humans or the environment [12]. While several technologies have demonstrated the ability to remove suspended solids (turbidity), organic matter, and TDS [5,13–15], the implementation of a short and simple treatment train tailored to the O&G basin and desired end use is likely the most cost effective treatment approach. Moreover, treatment trains including fewer technologies and requiring simpler operation with less maintenance will likely be favored by the O&G industry. Therefore, the current study focuses on use of a three-stage train to pre-treat and desalinate PW, consisting of biologically active filtration (BAF), followed by cartridge filtration and desalination with nanofiltration (NF) or reverse osmosis (RO).

1.2. Desalination of O&G wastewater

Efficient and sustainable desalination of PW requires thorough pretreatment to mitigate membrane fouling. BAF has demonstrated high removal of organic matter, exceeding 90% DOC removal from PW containing ~20,000 mg/L TDS [8], and has served as a pretreatment for NF desalination [16]. In a previous study by Riley et al., three O&G waste streams were treated by BAF for removal of organic matter, followed by ultrafiltration (UF) for removal of bacteria and particulates, and finally NF to remove TDS and organic matter [16]. NF90 membranes (DOW, Filmtec) were tested at varying pressures (150–300 psi) for each PW, producing final permeate of only 900 mg/L TDS (94% removal) and 1.3 mg/L dissolved organic carbon (DOC) (95% removal). However, the study was limited to only 60 h of operation at each pressure per water type and was conducted in recycling mode (not concentrating the feed); thus, limiting membrane fouling. While other studies have also evaluated the performance and fouling propensity of NF and RO membranes for desalination of PW, they were also limited in various aspects [17–21].

A study conducted by Xu and Drewes evaluated the efficacy of NF and RO membranes with varied antiscalants and cleaning methods during PW treatment [18]. However, the PW contained only 5300 mg/L TDS and < 2 mg/L DOC, and fouling experiments were conducted in recycling mode for only 75 h. While this study provides substantial insight to short-term fouling trends and discusses favorable membrane properties, the observed fouling might not be representative of fouling during long-term operations, particularly when concentrating the feed water with more challenging PW. Another study comparing the performance and fouling propensity of NF and RO membranes was very thorough in membrane characterization, examining the impacts of membrane properties on fouling, and analyzing changes to the membrane after PW treatment [19]. However, this study was also operated with low salinity PW (2500 mg/L TDS) in recycling configuration, and fouling tests were conducted for only 24 h. The studies by Kim et al. and Xu and Drewes are still probably more representative than another that evaluated NF and RO fouling in dead-end filtration, with only 90 min of exposure to PW [17].

The limited duration of the previous fouling studies, low salinity PW feeds, and operation in recycling or dead-end configuration warrants additional research, more representative of actual PW desalination

applications. Furthermore, there is lack of understanding of the long-term impacts of O&G related organic matter on polymeric membranes, particularly the potential for membrane degradation. Evaluation of such conditions is important to reveal limitations of commercial membranes and areas that need improvement to enable sustainable desalination and reuse of O&G wastewaters. Therefore, the present study uses the emerging, state-of-the-art closed circuit desalination (CCD) configuration to concentrate PW of ~20,000 mg/L TDS. CCD is an innovative mode that enables operation of RO or NF processes at high permeate flux, high water recovery, and lower pressures, thus reducing brine waste and operational/energy costs compared to traditional once-through RO configuration [22]. CCD is operated in a semi-batch configuration—quickly concentrating feed batches and replacing the brine with new feed while maintaining a constant permeate flux (contrary to traditional desalination systems that are operated as a batch with reduced permeate flux and cross-flow in subsequent stages). This reduces the membrane exposure time to damaging supersaturation concentrations, improving overall membrane sustainability and water recovery.

To date, no studies have investigated the application of CCD for PW treatment and in conjunction with NF desalination. Therefore, the main objective of this study was to assess the long-term sustainability and performance of NF and RO membranes during operation in CCD configuration at the bench-scale. This was accomplished through (a) evaluation of membrane performance (i.e., ion rejection, organic matter rejection, transmembrane pressure increase) (b) investigation of fouling propensity and membrane integrity (e.g., chemical cleaning and membrane characterization), and (c) comparison of NF and RO membranes. Furthermore, two promising membranes with potential for high rejection and low fouling propensity were evaluated in this study that has not been applied in PW desalination in the past (i.e., DOW Filmtec BW30-XFR and ECO-PRO).

2. Materials and methods

2.1. Membranes tested

Three commercial polyamide NF and RO membranes were evaluated in this study, including DOW Filmtec's (Midland, MI) NF90, BW30-XFR (fouling resistant; referred to as BW30 in this study), and ECO-PRO (referred to as ECO in this study) membranes. While NF90 is commonly used in various water treatment applications and is well documented in the literature [20,23], few studies with BW30-XFR [24], and only one paper (albeit very limited results) with ECO-PRO could be found [25]. ECO-PRO is reportedly a looser membrane like NF, but with rejection similar to RO, offering potential for reduced energy consumption (lower operating pressures than RO), and superior performance like RO. Membranes were tested in cross-flow membrane cells with a membrane active area of 139 cm² each. A 34 mil (0.86 mm) spacer was installed on the feed side to ensure adequate mixing, and a tricot spacer was installed on the permeate side. Prior to experiments, membranes were compacted for 20 h at 600 psi and tested with a 2 g/L NaCl solution to ensure membrane integrity and ion rejection.

2.2. PW feed stream and pretreatment

The PW tested in this study was acquired from a well-pad in the Denver-Julesburg (DJ) basin (Colorado, USA) after processing through oil/water separators. It was collected in 1 m³ (~250 gal) plastic totes and was stored at room temperature (~21 °C) until pretreatment by BAF. BAF pretreatment was applied to remove the majority of organic matter (70–90% DOC) from the PW and is described in detail elsewhere [16,26]. After BAF, the effluent was collected and filtered using a 0.5 µm cartridge filter to remove small particulates and some bacteria, and was stored at 5 °C until subsequent testing by NF or RO. The average feed water quality used throughout the duration of the study is

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