



Calcium carbonate scaling by reverse draw solute diffusion in a forward osmosis membrane for shale gas wastewater treatment



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ABSTRACT

In this study, we investigated the potential of membrane scaling and its reversibility in the FO process for produced water treatment. Sodium chloride (NaCl) and ammonia-carbon dioxide (NH₃-CO₂) were used as draw solutions to examine the effect of draw solute on membrane scaling behavior when treating model produced water, with a high concentration of calcium ions. While a negligible water flux decline was observed for NaCl draw solution, a severe water flux decline occurred for NH₃-CO₂ draw solution due to calcium carbonate (CaCO₃) scaling. Bidirectional solute diffusion analysis and scanning electron microscopy results (SEM) revealed that the CaCO₃ scaling formed only on the active layer of the membrane at the feed side due to the reverse diffusion of carbonate species from the draw solution and the high level of removal of Ca²⁺ by the FO membrane. Scaling formation enhanced the reverse diffusion of draw solutes implying a significant loss of draw solutes in the FO process. Osmotic backwashing did not recover the initial water flux by CaCO₃ scaling. Although ethylenediaminetetraacetic acid (EDTA) cleaning is effective for scaling, the rapid flux decline indicates that a more fundamental strategy is required for long-term FO operation when NH₃-CO₂ draw solution is used for water treatment. A two-step FO process (first FO using a NaCl draw solution and second FO using an NH₃-CO₂ draw solution) was proposed for the pretreatment of feed solution and stable FO operation for shale gas wastewater treatment.

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1. Introduction

Natural gas production in unconventional resources, such as shale gas formation and tight sands, has grown dramatically in recent years around the world as advances in horizontal drilling and hydraulic fracturing technology have made it economically viable [1–3]. The hydraulic fracturing technology uses water to fracture the shales and create pathways for natural gas flow [4]. The water quantity in hydraulic fracturing wells varies with the shale-gas play, well depth, the length of laterals, and other factors [5]. In the case of the Marcellus shale, 20,000 m³ of freshwater per well is consumed over its life cycle, with 65% being direct water consumption at the well site and 35% being indirect water consumption [6].

Hydraulic fracturing injects water containing chemical additives and proppant at high pressure to fracture the shales. For 1–3 weeks, approximately 15–25% of the injected water returns from the well as “flowback” at a high flow rate, but with relatively low levels of salinity, heavy metals, and naturally occurring radioactive materials (NORM) [6,7]. Produced water, accounting

for 70–90% of the total wastewater, is generated during gas production at lower flow rates, but with significantly higher levels of salinity, heavy metals, and NORM throughout the life of the well [8]. The flowback water can often be reused in the fracturing process; however, the produced water must be treated prior to reuse due to its high level of total dissolved solids (TDS) ranging from 1–400 g/L [7,9].

Currently, management options for the produced water include disposal via deep-well injection and desalination for reuse or discharge to the environment. Deep-well injection costs 0.59–13 dollars/m³ of produced water depending on the trucking distance from the well to the disposal site [6,10]. In addition to the high cost of trucking, environmental concerns and water shortages in shale gas regions drive the need for produced water treatment. Treatment of produced water is challenging due to its complex physicochemical composition, including organics, suspended solids, chemicals from the hydraulic fracturing fluid, and TDS. The desalination of the high-salinity produced water is the most challenging issue for external reuse.

Among the emerging water treatment technologies, forward osmosis (FO) has received increased attention in the last decade. The crucial advantage of the FO process is that it can be operated at low hydraulic pressure utilizing the osmotic pressure difference as a driving force. Recently, two companies providing FO

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technology have conducted a pilot FO membrane process for oil and gas wastewater [8,11,12]. One FO pilot (Green Machine), which operates without the pretreatment of drilling wastewater (2.5% w/w TDS), uses 26% w/w NaCl draw solution, while another FO pilot (membrane brine concentrator (MBC)) uses $\text{NH}_3\text{-CO}_2$ draw solution to treat 73 g/L TDS produced water with pretreatments such as oxidation and soda ash softening. Although these pilot studies have shown the feasibility of the FO process for oil and gas wastewater treatment, few studies have been conducted regarding the effect of feed and draw solution on FO performance and FO fouling mechanisms.

Based on the FO process studies using NaCl draw solution to treat drilling waste water [13] and coal seam associated water [14], FO membrane showed high rejection of organic and inorganic contaminants and the declined flux was effectively recovered by osmotic backwashing or hydraulic cleaning. When NaCl was used as draw solution, the inorganic fouling mechanism was similar to RO process i.e., supersaturation of scale precursor near membrane surface by concentration polarization depending on the feed water quality [15,16]. However, when other types of draw solution is used such as ammonium bicarbonate [17] and diammonium phosphate [18], scaling can be also formed by reverse draw solute flux from draw solution to the feed solution through reaction with the ions in the feed solution. Although this reverse solute flux led to calcium carbonate [17] and magnesium phosphate scaling [18] causing a severe water flux decline, the fouling layer was reversible using hydraulic cleaning.

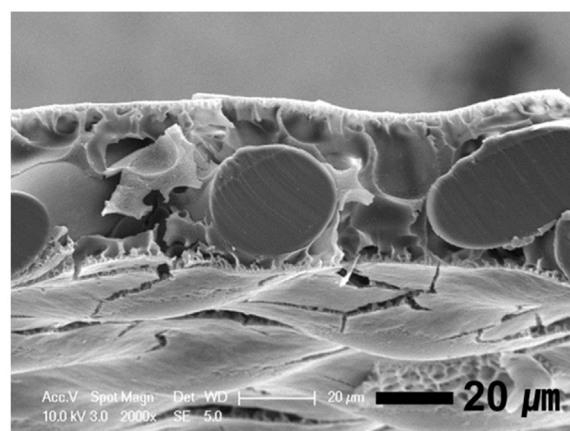
However, the reversibility of the fouling layer of FO could be different in produced water applications because produced water contains a high concentration of scale-forming compounds, such as Ca^{2+} , Mg^{2+} , Ba^{2+} , Sr^{2+} , and SO_4^{2-} [5,7]. The concentrations of the scale precursors in produced water vary widely by well, but generally Ca^{2+} is the ion present at the highest concentration. In the Marcellus shale, the Ca^{2+} concentration ranges from 2.6–43 g/L in produced water, which is 6- to 100-fold higher than in seawater [5]. The high concentrations of scale-forming compounds would necessitate chemical cleaning, in addition to hydraulic washing or osmotic backwashing, for FO membrane operation.

In this study, we investigated the potential of membrane scaling and its reversibility in the FO process to treat shale gas produced water containing high concentrations of scale forming ions. Calcium ion was used as a representative scale forming ion to simulate produced water, and two widely investigated draw solutions, NaCl and $\text{NH}_3\text{-CO}_2$, were used to investigate inorganic scaling by bidirectional diffusion. In addition, the efficiencies of osmotic backwashing and chemical cleaning were compared for FO membrane operation. The bidirectional diffusion results of this study show the importance of chemical reactions between the feed and draw solutes and extend our understanding of the FO membrane fouling mechanism to cope with inorganic fouling in the FO process.

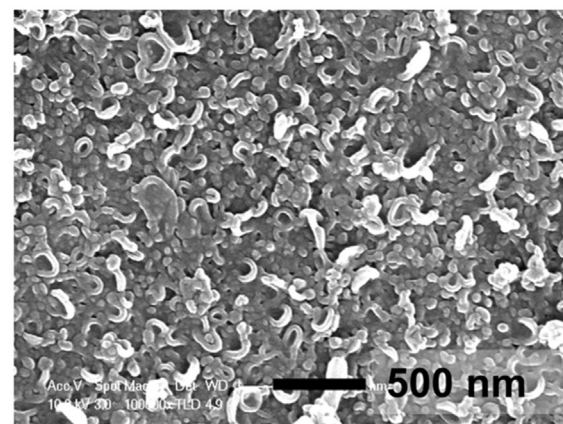
2. Materials and methods

2.1. FO membrane and systems

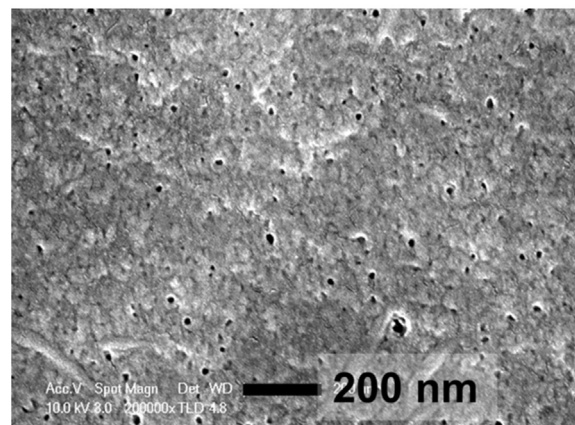
A flat-sheet, thin-film composite (TFC) FO membrane was obtained from Porifera (Hayward, CA, USA). The membrane consisted of a selective polyamide active layer on top of a polysulfone support layer, with an embedded woven mesh (Fig. 1). The polyamide active layer is known to have an abundance of carboxyl groups, which produce a negative charge on the surface [19]. All experiments were conducted in FO mode, with the active layer of the membrane facing the feed solution. A custom-built cross flow membrane test cell, with symmetrical flow channels



(a)



(b)



(c)

Fig. 1. SEM images of the TFC-FO membrane. (a) cross-section, (b) active layer, and (c) support layer.

(50 × 100 × 2 mm) was used for the FO experiment. A commercial tricot and mesh-type spacer were used on both the draw and feed channels to simulate the hydrodynamic conditions inside a spiral wound membrane element. Two gear pumps (Cole-Parmer, Vernon Hills, IL, USA) were used to circulate the feed and draw solutions in separate closed loops at a flow rate of 0.5 L/min, corresponding to a cross-flow velocity of 13.6 cm/s, considering the void volume of spacers. Water baths (Lab Companion, Jeio Tech, Seoul, Korea) were used to maintain the temperature of both the

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