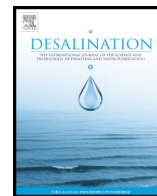




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Forward osmosis as a pre-treatment for treating coal seam gas associated water: Flux and fouling behaviours

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HIGHLIGHTS

- Different membranes were employed to treat real coal seam gas associated water.
- Flux decline was caused by the compounding effects of ICP and membrane fouling.
- Hydraulic cleaning restored declined flux.
- High rejection of foulants in coal seam gas feed water was achieved.

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ABSTRACT

In this study, a bench scale forward osmosis (FO) process was operated using two commonly available FO membranes in different orientations in order to examine the removal of foulants in the coal seam gas (CSG) associated water, the water flux and fouling behaviours of the process were also investigated. After 48 h of fouling simulation experiment, the water flux declined by approximately 55 and 35% of its initial level in the TFC-PRO and CTA-PRO modes (support layer facing the feed), respectively, while the flux decline in the TFC-FO and CTA-FO modes (active layer facing the feed) was insignificant. The flux decline in PRO modes was caused by the compounding effects of internal concentration polarisation and membrane fouling. However, the declined flux was completely recovered to its initial level following the hydraulic cleaning using deionised water. Dissolved organic carbon (DOC), adenosine tri-phosphate (ATP) and major inorganic scalants (Ca, Mg and silica) in the CSG feed were effectively removed by using the FO process. The results of this study suggest that the FO process shows promising potential to be employed as an effective pre-treatment for membrane purification of CSG associated water.

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1. Introduction

Coal methane or coal seam gas (CSG) is an unconventional natural gas resource. It is essentially methane and can be used in the same way as conventional natural gas [1]. Since its first commercial production commenced in Australia in the mid-1990s, this industry has developed very fast, with 90% of the CSG production based in Queensland. CSG has become an important energy source and will gradually gain importance in many countries as conventional gas productions decline [2,3]. The rapid development of the CSG industry has resulted in environmental issues in water resources. Large volumes of water are produced and released during the CSG production. This water may contaminate and affect the quality of the surface or underground water if it is discharged without proper treatment [4,5]. CSG associated water exhibits lower

total dissolved solid (TDS) concentrations than conventional produced water such as oil and gas production process water. For example, the TDS concentrations in Rocky Mountain basins in the US, ranging from 370 to 43,000 mg/L compared to 1000 to 400,000 mg/L for conventional oil and gas resources [6]. The lower TDS concentrations of CSG water suggest that these types of water can be utilised as an alternative water resources for irrigation and can be recycled in the production process. However, there are still knowledge gaps of the suitability of CSG water composition and ranges of constituent concentrations for water treatments [7].

Some technologies used for CSG water treatment include coagulation, microfiltration (MF) or ultrafiltration (UF) and reverse osmosis (RO) [1]. For treatment of this produced water, forward osmosis (FO) can be hybridised with processes such as RO or distillation to continuously reconcentrate the draw solution and produce high quality product water [5]. FO is an osmotically driven membrane process that takes advantage of the osmotic gradient to draw water across the semipermeable

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membrane from the feed side to the draw side. Due to the absence of hydraulic pressure during the process, FO provides many advantages such as lower fouling tendency, high water recovery, and high rejection of wide range of contaminants [8,9]. There have been a number of studies for the treatment of produced water using FO processes. Hickenbottom et al. [5] investigated the FO process performance for treating drilling wastewater from a shale gas field. In this study, the FO process was able to recover (>80%) water from the wastewater with minimal irreversible membrane fouling and reject more than 99% of dissolved organic carbon (DOC). Hydration Technology Innovations (HTI) developed a portable oilfield wastewater reclamation system using the FO process [10]. Using this system on each well, over 80% of the drilling waste can be recycled to provide approximately 20% of the water required for hydraulic fracturing. Furthermore, this system showed high pollutant rejection. It can reject >90% of undesired solutes in the impaired process water. Another pilot scale study by Oasys Water, Inc. using a FO membrane brine concentrator (MBC) has been conducted in shale gas exploration [11]. It was found that feed water recoveries were similar to those of evaporative brine concentration technologies (>60%). The product water generated in the pilot system was found to meet surface water discharge quality criteria of the local standard. This study investigated the impacts of membrane selection on the performance of FO membranes for treating CSG process water. The actual process water obtained directly from a CSG field in Queensland Australia was used as feed water. Two commercially available FO membranes were used in different orientations to evaluate the water flux, membrane fouling, and foulant rejection. The feasibility of using the FO process as a pre-treatment of treating CSG process water before the pressure membrane process is investigated.

2. Materials and methods

2.1. FO membranes

Two most commonly used FO membranes were used in this study that is polyamide (PA) thin-film composite (TFC) and cellulose tri-acetate (CTA) FO membranes (HTI, OR, USA). These membranes were prepared with an effective membrane surface area of 48 cm² (80 mm long and 60 mm wide) to fit the custom-made cross-flow membrane cell with dimensions of 80 mm long, 60 mm wide, and 2 mm deep. Membrane samples were soaked in MilliQ water (MilliQ™, Australia) for 24 h prior to the fouling simulation experiments. FO membranes were installed in both FO (active layer facing the feed channel) and pressure-retarded osmosis (PRO; supporting layer facing the feed channel) orientations. Mesh spacers (Low foulant spacer, Sterlitech, WA, USA) were cut into 80 mm long and 60 mm wide strips and placed in the membrane holders to support the FO membrane and to reduce external concentration polarisation (ECP) on both sides of the membrane.

2.2. Feed and draw solutions

High purity NaCl (purity ≥ 99.5%, Sigma-Aldrich, Australia) was used as draw solute in all FO experiments. CSG co-produced water was collected from a site in Queensland, Australia and was used as the feed solution. The main composition of the feed solution is summarised in Table 1.

2.3. Bench-scale forward osmosis system

A FO membrane system was used to perform FO fouling and cleaning experiments as described in our previous study with a minor modification [12]. The draw and feed solutions flowed co-currently at flow rates of 500 ml/min in each channel on both sides of the membrane, using peristaltic pumps (Cole-Parmer, Australia) and flow meters (Cole-Parmer, Australia). The permeate flux was monitored using an automated balance (MS 1600I, Mettler-Toledo, Switzerland) connected to a computer data logging system. During the experiment, the cross-

Table 1

The main composition of coal seam gas feed water.

Coal seam gas water composition	Concentrations, mg/L
pH	[8.8]
Conductivity (μS/cm)	1855
Total dissolved solids (TDS)	941
Dissolved organic carbon (DOC)	1.8
Aluminium, Al	0.006
Bicarbonate, HCO ₃	1220.543
Boron, B	0.306
Calcium, Ca	1.600
Chloride, Cl	367.770
Copper, Cu	0.004
Iron, Fe	0.003
Lead, Pb	0.073
Magnesium, Mg	0.560
Manganese, Mn	0.002
Nickel, Ni	0.003
Nitrate, NO ₃	14.947
Nitrite, NO ₂	68.042
Potassium, K	1.710
Silicon, Si	1.655
Sodium, Na	747.455
Total silica, SiO ₂	11.000
Sulphate, SO ₄	10.647

flow rate of the draw and feed solutions was maintained at 9.9 cm/s. The concentration of draw solution was maintained constant at 1 M to maintain the constant osmotic pressure of the draw solution using a conductivity sensor and dosing pump (8850-3 Conductivity/Resistivity Transmitter, GF Signet, VT, USA). The NaCl rejection was determined using a conductivity meter (HQ40d, Hach, CO, USA). The temperature of both feed and draw solutions was maintained at 25 °C. In each batch of the experiments, MilliQ water was used as the feed and 1 M NaCl as the draw solution. After a stable water flux was achieved in one hour, the feed water was changed to CSG production water and the FO system was operated continuously for 48 h. Membrane hydraulic cleaning was performed immediately after another 48 h fouling experiment. The membrane was rinsed with MilliQ water at a cross-flow rate of 9.9 cm/s for 30 min. After the cleaning, the membrane water flux was measured again for one hour, finally, 1 M NaCl was used as the draw solution to assess flux recovery with CSG production water as the feed.

2.4. Analytical methods

2.4.1. Feed and draw solution compositions

CSG feed water, and draw solutions after each filtration simulation, were analysed in order to investigate the composition of feed water and rejection of major ionic species, and ion chromatography (IC, Dionex ICS-3000, CA, USA) and inductively coupled plasma optical emission spectrometry (ICP-OES, Perkin-Elmer Optima 5300 DV, MA, USA) were used for determination of anions and cations, respectively.

2.4.2. Membrane surface characterisations

The hydrophobicity of the membranes was measured using a contact angle goniometer (Model 200, Rame-Hart Inc., NJ, USA) by the sessile drop method. Ten measurements were taken for each sample and the results were averaged.

Atomic force microscopy (AFM) was used to determine topographic image and height profile of the membranes (PSIA XE-100, Park Systems, Korea). The samples were observed in 5 × 5 μm area using a non-contact cantilever (NCHR 10 M, Park Systems, Korea). The arithmetic average roughness (R_a) of the fouled membranes was measured.

Functional groups of the membrane surfaces were identified with Fourier-transform infrared (FTIR) spectroscopy (Varian 600-IR, CA, USA). The spectrum of each membrane was collected between 4000 and 650 cm⁻¹ wavelength ranges.

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