Desalination 394 (2016) 91–100

Contents lists available at ScienceDirect

Desalination

journal homepage: www.elsevier.com/locate/desal

Fouling characteristics and their implications on cleaning of a FO-RO pilot process for treating brackish surface water

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HIGHLIGHTS

- Fouling characteristics of a pilot FO-RO plant using natural surface water was studied.
- Severe flux decline was caused by high scaling/fouling potential feed solutions.
- Fouled membrane autopsy found inorganic scaling was the main cause of fouling.
- Physical and chemical cleaning was ineffective to restore declined flux.
- A sufficient cleaning strategy is important to remove inorganic scalants.

ARTICLE INFO

Article history:
Received 27 February 2016
Received in revised form 25 April 2016
Accepted 28 April 2016
Available online 14 May 2016

Keywords:
Forward osmosis
Reverse osmosis
FO-RO hybrid system
Autopsy
Membrane fouling characterisation

ABSTRACT

In this study, a laboratory pilot scale forward osmosis-reverse osmosis (FO-RO) hybrid system was used to desalinate both actual and spiked brackish surface water (BSW). An overall performance evaluation was conducted and the membrane foulings were characterised by comprehensive techniques. Severe flux decline was observed during the treatment of high scaling/fouling potential feed solutions. It was found that when the raw BSW was used as feed, the flux can be completely recovered by hydraulic cleaning. When the raw BSW was spiked with nutrients and/or scaling ions for accelerated scaling and biofouling, more severe flux decline was observed. Inorganic scaling caused by calcium and phosphate, and their interactions with organic constituents in the feed solutions were the dominant cause of the declined system performance. For the spiked feed water, the combined physical and chemical cleaning using two chemical agents was not able to restore reduced flux to its initial value. This study identified the need for implementing a sufficient cleaning strategy targeting different membrane foulants, particularly for inorganic scalants; as well as confirms the need of fouling-resistant membrane.

1. Introduction

Forward osmosis (FO) is an emerging membrane technology, and has drawn growing attention from researchers and industries over the last decades [1,2]. FO membrane processes have high rejection of contaminants and low irreversible fouling tendency [3]. As a result, FO processes require less pre-treatment when working with impaired water and less periodic cleaning than pressure-driven membrane processes [4]. With these benefits, earlier studies have investigated the potentials for use of FO processes in wastewater treatment, brackish/seawater desalination, food processing, and power generation [1]. Despite recent advances in FO, there are several challenges remaining for successful application of this technology including economic sustainability, selection of optimal draw solution (DS), and identification of fouling mechanism [5].

Many researchers have attempted to combine osmotically-driven membrane systems with nanofiltration (NF), reverse osmosis (RO) or membrane distillation (MD) to investigate the viability of the process as an alternative to conventional seawater reverse osmosis (SWRO) systems [6]. FO hybrid systems provide several operational advantages including reduction of fouling and scaling compared to pressure-driven membrane process, recovery of osmotic energy of RO brine, minimisation of the use of chemicals required for conventional pre-treatments, provision of multiple barriers for water purification, and dilution of seawater for lowering energy use and operating costs when it is used as a pre-treatment for the pressure-driven membrane

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http://dx.doi.org/10.1016/j.desal.2016.04.026

processes in water purification systems [7–9]. Yangali-Quintanilla et al. [10] used FO coupled with low pressure RO (LPRO) to treat wastewater effluent as feed, and seawater from Red Sea as the DS. The results were promising. The system consumes 50% of the energy used for high pressure seawater RO desalination, and diluted DS can be used as an alternative water source for aquaculture. Shaffer et al. [11] investigated the feasibility of seawater desalination using a FO-RO process for agricultural irrigation. Boron and chloride concentrations were met for agricultural irrigation. A modelling study revealed that less energy was consumed than for a conventional two-phase RO process. A pilot scale FO-RO hybrid system was used for treating wastewater effluent [12]. Although both reversible and irreversible membrane fouling was observed, the water flux remained stable and achieved long-term operation (~1300 h). Robust rejection by the dual barrier was also achieved for inorganic and organic foulants.

A potential drawback of pressure-driven membrane processes is fouling, resulting in increased normalised pressure and deceased normalised flux [13]. Eventually, such technical issues increase operating costs. FO processes are known to be less susceptible to fouling compared to conventional membrane processes such as NF and RO due to less cake layer formation and lower compaction of foulants on the membrane surface [1,14]. However, fouling in FO is still considered one of the major limiting issues for successful application [15]. Fouling studies on FO processes have been carried out extensively in recent years, ranging from organic [16,17] and biofouling [18–21], and colloidal fouling [22, 23] to inorganic scaling [24–28] under varying feed quality, membrane materials and orientations, hydrodynamic conditions, and draw solutions. With these studies, the understanding on fouling in FO process has been extensively improved. Despite the significant attentions given to this fouling research, the knowledge about fouling characteristics of pilot- and full-scale FO processes is still limited. Very few studies have investigated FO processes in more 'real world' membrane applications. As a result, there are still knowledge gaps in fouling in large- or full-scale FO applications.

This study investigated spiral-wound FO membrane performance under a series of accelerated extreme fouling conditions in the FO-RO pilot process for treating brackish surface water (BSW). FO performance including water flux and fouling behaviours, and flux recovery were evaluated with BSW as a feed with different compositions. Autopsy of the fouled membrane and comprehensive analytical techniques were performed to elucidate heterogeneous membrane fouling and provide insights into the fouling in the large-scale FO system. By obtaining these fouling characteristics, targeted pre-treatment and necessary cleaning protocols could be developed which can ensure the long-term sustainable operation of the FO-RO hybrid system.

2. Materials and methods

2.1. Feed and draw solution

BSW was collected from Mawson Lakes, South Australia, Australia in March 2015. Four different feed solutions were prepared in order to examine the full effect of different foulants on the FO process flux behaviours. In Stage I, raw BSW without pre-treatment was used directly. In Stage II, the feed was prepared by adjusting the C:N:P mass ratio to 100:20:10 (400 mg/L C, 80 mg/L N, and 40 mg/L P) via spiking with 0.9216 g sodium acetate (Merck, Australia), 0.1036 g sodium dihydrogen phosphate (Chem Supply, Australia), and 0.3246 g sodium nitrate (Chem Supply, Australia) per litre of BSW. In Stage III, the feed was prepared by dissolving 35 mM of $CaCl_2$ (Merck, Australia) 20 mM of Na_2SO_4 (Merck, Australia) and 19 mM of NaCl (Sigma-Aldrich, Australia) corresponding to a gypsum saturation index (SI) of 1.3 [29]. In Stage IV, the feed was prepared by adjusting the C:N:P mass ratio to 100:20:10 and adding gypsum to achieve a SI of 1.3. The DS was 0.675 M sodium chloride (NaCl) solution.

2.2. FO-RO hybrid system set up

A hybrid FO-RO pilot system was used in laboratory for this study. The dual barrier pilot system was designed and constructed in our previous study [24] and is shown in Fig. 1. The FO system comprises two spiral-wound thin-film composite (TFC) FO membrane elements (2521FO-TFC-MS-P-3H, Hydration Technology Innovations, USA) which are contained in a separate vessel, a high pressure pump (Calpeda, Italy) for the feed, a gear pump (Cole-Parmer, Australia) for the DS, flow-meters (Cole-Parmer, Australia), pressure gauges (Swagelok, Australia) and valves. The two FO elements were installed horizontally and connected in series for both feed and DS. The two digital flow-meters (Cole-Parmer, Australia) were installed on the feed circuit before and after the membranes and connected to a computer in order to record/monitor the water flux of the FO process. The concentration of the DS was monitored using a conductivity meter (HQ 40d, Hach, USA). The RO system comprised a spiral-wound TFC membrane (SW30–2521, Dow Filmtec, USA), a tank for the diluted DS (feed for RO system), a high pressure pump (Hydra-Cell Pump, Wanner Engineering, Inc., USA), flow-meters (Cole-Parmer, Australia), pressure gauges (Swagelok, Australia) and valves. The water flux and the pressure of the RO process were monitored using a digital flow-meter (Cole-Parmer, Australia) and pressure sensors, both connected to a computer.

2.3. FO-RO hybrid system operation

The FO experiment was first conducted in a start-up batch mode until reaching the desired outlet DS concentration, then the system was switched to continuous mode. A water production rate of 32% was selected. The BSW feed was pumped into the feed side of the first FO element at a rate of 4 L/min and 15 psi (1.03 bar). The DS was pumped into the draw side of the first FO element at 10 psi (0.69 bar). The water production rate of the FO process is determined by the ratio of the fresh water flux extracted from the feed to the initial feed flow rate. The diluted DS was collected separately and reconcentrated to its initial concentration (0.675 M) by the hybridised RO process (Fig. 1). The DS was pumped into the inlet of the RO element at a rate of 2 L/min and the operating pressure was kept at 67 bars. The reconcentrated DS was pumped back into the DS tank and reused for the next stage of the FO process. Fresh DS was prepared and added to fill the DS tank to its initial volume which is 50 L when desired. Duplicate experiments were carried out for each feed solution. Physical cleaning using MilliQ (MilliQ™, Australia) water was performed between each stage for 30 min. At the end of the operation of the FO process, the system was stopped and the cleaning experiment was conducted. The chemical cleaning solutions for FO were prepared at a high pH using 2.0% (w) of sodium tripolyphosphate ($Na_5P_3O_{10}$; Ajax, Australia) and 0.8% (w) of Na-EDTA ($C_{10}H_{14}N_2Na_2O_8\cdot 2H_2O$; sodium salt of ethylenediaminetetraacetic acid; Merck, Australia) and 0.025% (w) Na-DDBS ($C_6H_5(CH_2)_{12}\cdot SO_3Na$, sodium salt of dodecylbenzene sulfonate; Tokyo Chemical Industry, Japan), and a low pH using 2.0% (w) of citric acid ($C_6H_8O_7$), respectively. The high pH cleaning solution is specifically recommended for removing calcium sulphate and light to moderate levels of organic foulants of natural origin, heavier levels of organic foulants of natural origin and the low pH cleaning solution is useful in removing inorganic scale (e.g. calcium carbonate, calcium sulphate, barium sulphate, strontium sulphate) [29].

2.4. Membrane autopsy

In order to characterise the fouling layers formed on the FO membrane surface, the FO membrane modules were autopsied after the Stage IV filtration experiment. Each membrane module was cut into three parts; inlet, central, and outlet section, in order to examine the relationship between the flow direction of the membrane element