Contents lists available at ScienceDirect

Desalination

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

Combined silica and sodium alginate fouling of spiral-wound reverse osmosis membranes for seawater desalination

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ARTICLE INFO

Keywords: Combined fouling Membrane fouling Sodium alginate Colloidal silica Reverse osmosis Spiral wound

ABSTRACT

Reverse osmosis efficiency depends on the precise prediction, among other factors, of membrane fouling. Although extensive research has been devoted to single foulants, rather less attention has been paid to simultaneous foulants which are more representative of real feed solutions. The main purpose of this research was therefore to characterize and quantify the fouling potential that results from the coexistence of simultaneous foulants. Silica and sodium alginate were used as respective models of inorganic and organic colloidal foulants. In order to understand the possible interactions, membrane fouling was first forced with a single foulant and the results compared to combined fouling. The experiments were carried out under constant pressure and employed synthetic seawater with 32 g/L of NaCl in a pilot-scale reverse osmosis unit. A standard commercial spiral-wound thin-film composite membrane was used. The results showed a more aggravated permeate flux decline in the concentration of sodium alginate was equal to or higher than the concentration of silica. In these cases, the permeate flux decline of combined fouling was more severe than the sum of flux decline of individual foulants.

1. Introduction

There is considerable concern in the scientific community as a whole that membrane fouling in membrane-based desalination technologies, and especially in reverse osmosis (RO) membranes, remains a major drawback for the implementation of such technologies [1–3].

Membrane fouling is an unavoidable consequence of the accumulation of undesirable foulants on, in or near the membrane. Depending on the nature of the foulant, fouling is classified into four main categories [4,5]: (1) particulate and colloidal matter deposition on the membrane surface; (2) organic fouling; (3) scaling and inorganic fouling; and (4) biofouling. The consequences of this phenomenon include a negative effect on efficient membrane performance and reductions in system efficiency and membrane lifetime [6].

Considerable research efforts have been made by the scientific community in this field during the last 30 years, reflected in the number of published papers. Furthermore, the number of publications in 2016 in this field was around 20 times higher than in 1992 and twice as high if compared to 2011 [2]. Numerous research studies have attempted to address directly how membrane fouling occurs and how to predict fouling [7], while others have focused on the mitigation and prevention of fouling using, for example, nanocomposite membranes [8–10].

In relation to attempts to directly address how membrane fouling occurs, particular and complex studies on organic and inorganic fouling of RO membranes are of great relevance [11-16]. To date, most studies on RO fouling have focused on fouling caused by either one single foulant or interaction with specific ions. The findings show that fouling behavior is strongly affected by feedwater environment (foulant type, concentration, pH, specific ions such as Ca⁺², ionic strength), membrane properties and hydrodynamic conditions.

Nevertheless, while membrane fouling by individual foulants is an extremely complex process, it is clear that the combined impact of the coexistence of multiple foulants may differ from the impact of individual foulants. Therefore, one of the most important challenges in industrial applications is to comprehend the fouling mechanisms that mark the synergistic effects of the coexistence of multiple foulants. Such an understanding can lead to the development of more effective approaches for control of membrane fouling.

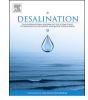
Despite its importance, few studies have been published dealing with RO fouling mechanisms as a result of multiple foulants. Combined organic-organic [13,17–21] and inorganic-organic [22,23] foulant studies are the most common in RO systems. Previous research highlighted a synergistic behavior between foulants where simultaneous foulants caused a more severe membrane fouling compared to fouling

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https://doi.org/10.1016/j.desal.2018.03.032

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Received 28 June 2017; Received in revised form 15 March 2018; Accepted 26 March 2018 0011-9164/@ 2018 Elsevier B.V. All rights reserved.

caused by single foulants. Conversely, R. Higgin et al. [22] reported a synergistic behavior between silica and alginic acid. In this case, a lower permeate flux decline was observed in the combined fouling test than the flux decline produced by individual silica.

In spite of these advances, most research papers focus on a single or combined foulant type with a feedwater solution characterized by low salinity, with the experiments performed in a laboratory unit which employs flat sheet membrane pieces - which is not representative of real scale seawater industrial applications.

In order to broaden our knowledge on this subject, the aim of this experimental study is to provide a greater understanding of fouling behavior by combined organic and inorganic fouling under more representative feed solutions and operating conditions. In this study, fouling is characterized and the fouling potential quantified for a commercially spiral-wound thin-film composite (TFC) RO membrane element.

2. Experimental methods and materials

2.1. Model foulants and chemical reagents

Commercial sodium alginate from brown algae (Sigma-Aldrich, Madrid, Spain) and colloidal silica Ludox[®] TM-40 (Sigma Aldrich Madrid, Spain) were selected as the organic and inorganic colloidal foulant models, respectively.

Sodium alginate was selected in order to model polysaccharides which are quite abundant in natural waters. Its behavior is similar to that of extracellular polymeric substances (EPS). These EPS are produced by microorganisms and play an important role in biofouling. Also, colloidal silica is one of the main foulant present in natural waters.

Sodium alginate was supplied in powder form (12–80 kDa). The colloidal silica was supplied as a 34 wt% suspension in deionized water, with a particle size of 20 nm as reported by the manufacturer. Sodium alginate and silica colloid solutions were prepared by dissolving each foulant for each experiment in deionized water with vigorous stirring. The organic and inorganic foulant concentrations used in the experiments were 100, 200 and 300 mg/L for silica and 50, 100 and 300 mg/L for sodium alginate.

The experiments were carried out using synthetic feedwater. Table 1 shows a detailed composition of the feed solution which included tap water and commercial sea salt (99.4% Regenia, Union Salinera S.A., Spain) at a fixed concentration of approximately 32 g/L. A feed water pH of 8 was maintained in all tests by adding small amounts of NaOH. Feed solutions were stirred for 3 h at room temperature and then passed through a $0.45 \,\mu\text{m}$ filter.

The chemical agents used for chemical cleanings were 0.1 wt%

Table 1

Physicochemical	characteristics	of	the	synthetic	seawater
used in the tests.					

Parameters	Value (mg/L)		
Aluminum	< 0.005		
Bicarbonate	69.2		
Total organic carbon	< 1		
Calcium	122		
Carbonate	< 10		
Free chlorine	< 0.1		
Chloride	19,314		
Iron	< 0.005		
Magnesium	< 5		
Potassium	10.5		
Sodium	11,800		
Sulfates	133		
STD	31,448.7		
pH	7.89		

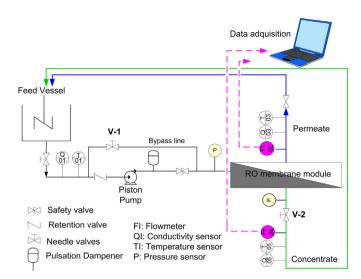


Fig. 1. RO membrane set-up system for cross-flow and constant pressure employed in the fouling experiments.

NaOH and 1.0 wt% disodium ethylenediaminetetraacetate (Na₂-EDTA) at pH 12 (for colloidal silica test). For the sodium alginate test, 0.1 wt% NaOH and 1.5 wt% Osmotech 2691 at pH 12 were used. Both the solutions mentioned above and 2.0 wt% citric acid at pH 2–3.5 were used for the combined fouling test. All chemicals used were reagent grade.

2.2. Spiral-wound RO membrane element

A standard polyamide thin-film composite (TFC) spiral-wound RO membrane element SW30-2521 (Dow[®] Water & Process Solutions) was employed in the fouling test. The effective surface area of the membrane is 1.2 m^2 (523 mm long and 61 mm in inner diameter).

2.3. Experimental set-up and fouling test

All the experiments were carried out on a modified version of a commercial cross-flow pilot-scale unit (Gunt Hamburg, CE530). Fig. 1 shows the experimental set-up used in the fouling tests.

The facility has the following instrumentation: (a) two differential pressure gauges controlled by two needle valves (V-1 and V-2) at the feed and reject sections, respectively, (b) two digital flowmeter sensors connected to a data acquisition system, (c) pneumatic level switches and (4) three digital conductivity and temperature sensors (WTW Tretracon 325) in the feed, permeate and rejection line.

The experimental protocol was divided into two parts, clean membrane characterization and fouling experiments.

2.3.1. Clean membrane characterization

Prior to the fouling experiments, deionized water was filtered through the virgin membrane to allow compaction and the removal of substances to preserve the membrane element. A baseline test was then performed over approximately 5 h to evaluate flow behavior without the presence of foulant. Characterization of the clean membrane was established as a reference point for the analysis of the results obtained in the fouling experiments, and also allowed consideration of the performance characteristics of the membrane under the study conditions.

2.3.2. Fouling and cleaning experiments

Typical fouling experiments for a constant pressure run include the following four steps (Fig. 2): (1) permeate flux of deionized water, (2) membrane setting, (3) foulant filtration and (4) membrane cleaning.

First, prior to each fouling test, deionized water was filtered through the membrane element for 1 h to obtain the membrane hydraulic resistance, $R_{\rm m}$.

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