Journal of Membrane Science 544 (2017) 221–233

Contents lists available at ScienceDirect

Journal of Membrane Science

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

Unpacking compaction: Effect of hydraulic pressure on alginate fouling

Emily W. Tow, John H. Lienhard V*

Rohsenow Kendall Heat Transfer Laboratory, Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

ARTICLE INFO

Keywords:
Forward osmosis
Foulant compaction
Fouling resistance
Organic fouling
Reverse osmosis

ABSTRACT

High pressure is often considered to be the cause of the high fouling propensity of reverse osmosis (RO) relative to forward osmosis (FO). Several experimental studies have shown that alginate fouling is more susceptible to cleaning in FO than in RO, but the theory that foulant compaction causes this disparity seems to be contradicted by the incompressibility of alginate hydrogels. In addition, the effect of hydraulic pressure on fouling in osmotic membrane desalination has never been experimentally isolated, because fixed-flux comparisons at different hydraulic pressures require different draw solution osmotic pressures. In this study, a new approach to isolating the effect of hydraulic pressure on alginate fouling and cleaning is introduced: operating FO with elevated but equal feed and draw hydraulic pressures. The same concentration of sodium chloride is used as the draw solution in all trials to eliminate possible effects of draw solution osmotic pressure on membrane fouling or cleaning. Theoretical modeling of the effect of alginate foulant compaction on flux reveals that foulant compaction should accelerate flux decline with low salinity feeds but retard flux decline at high salinity. However, in low-salinity alginate fouling trials, for which foulant compaction should accelerate flux decline, the measured flux decline rate was not affected by hydraulic pressure. Furthermore, when fouled membranes were cleaned by increasing the feed velocity and reducing the draw osmotic pressure, there was no apparent relationship between hydraulic pressure and cleaning effectiveness. Finally, in situ visualization of foulant removal during the cleaning process revealed no difference in foulant removal mechanisms between different hydraulic pressures. These findings demonstrate that alginate gel compaction by high feed hydraulic pressure does not occur and suggest that other explanations should be sought for FO's high fouling resistance relative to RO.

1. Introduction

Forward osmosis (FO) is often compared to reverse osmosis (RO) in terms of energy consumption and fouling propensity. After some debate [1,2], RO has been found to be more energy-efficient [3–6] but also more prone to irreversible fouling [7,8]. Although even FO can foul significantly (see, e.g., [9]), some researchers have postulated that the high feed hydraulic pressure used in RO exacerbates fouling. A number of recent reports, including Refs. [7,8,10–15], attribute differences between RO and FO membrane fouling to foulant compaction by high hydraulic pressure. The most compelling evidence comes from studies that show a marked difference in the effectiveness of physical cleaning between identical membranes fouled under identical hydrodynamic conditions at the same initial flux in RO and FO [7,8,14–16]. According to the theory that foulant cake density increases with feed hydraulic pressure, a less-compact cake layer forms in FO and should be easier to remove. However, the effects of pressure have never been experimentally isolated from other differences between FO and RO.

This study seeks to experimentally validate or invalidate the theory that high feed pressure compacts foulants. Previous studies of the effect of pressure on cleaning effectiveness in FO and RO are reviewed and the hypothetical effects of compaction on flux decline are modeled. As discussed further in Section 2, foulant compression is related only to feed hydraulic pressure and the pressure drop through the foulant layer and is not independently affected by the hydraulic pressure of the draw or permeate. Therefore, hydraulic pressure can be experimentally isolated as an independent variable by conducting FO fouling and cleaning trials with the feed and draw streams at elevated but equal hydraulic pressures (up to 40 bar), thus sidestepping the need to vary the draw solution concentration to maintain a fixed initial flux. Fig. 1 illustrates the approaches to examining the effect of pressure on fouling taken by this study and previous studies.

To determine whether foulant compaction by high pressure significantly affects membrane fouling and cleaning, flux decline and cleaning effectiveness were measured and video was recorded of the foulant removal process at different pressures. Feed hydraulic pressure was not found to significantly affect flux decline rate, cleaning effectiveness, or foulant removal mechanisms, indicating that foulant

---

* Corresponding author.
E-mail address: lienhard@mit.edu (J.H. Lienhard V).

http://dx.doi.org/10.1016/j.memsci.2017.09.010
Received 2 April 2017; Received in revised form 8 August 2017; Accepted 4 September 2017

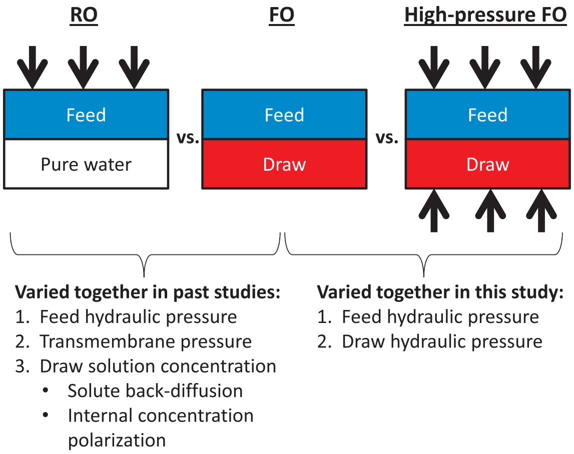

Fig. 1. Methods of isolating the effect of pressure on osmotic membrane fouling taken by past studies (Refs. [7,8,14,15]) and the present study. All studies varied feed hydraulic pressure, but other variables that could potentially affect fouling were also changed to avoid altering flux.

compaction by high feed hydraulic pressure does not explain the high fouling propensity of RO relative to FO.

1.1. *Definition of pressure terms*

For clarity, certain terms relating to pressure are defined as follows in the context of this study: Hydraulic pressure, P, is used to mean the gauge pressure relative to atmospheric pressure. Accordingly, feed hydraulic pressure, P_f, refers to the gauge pressure of the feed. Transmembrane pressure difference (TMP) is the difference in pressure across the membrane (including any fouling layer), $P_f - P_d$, where P_d is the gauge pressure on the back side of the membrane, whether the solution there is draw or permeate. When the draw or permeate pressure is atmospheric, as it is in RO and standard implementations of FO, feed hydraulic pressure is equal to TMP, and this distinction is unimportant. However, the present approach to testing the effect of feed hydraulic pressure on fouling propensity involves raising the hydraulic pressure of the draw solution in FO. As a result, feed hydraulic pressure is not necessarily equal to TMP in this study. The pressure drop across the foulant cake refers to the difference in hydraulic pressure between the feed solution and the feed-facing side of the membrane that results from resistance to water flow through the foulant layer. The potential effects of these various pressure differences on fouling are discussed in Section 2.

1.2. *Literature review: role of pressure in osmotic membrane fouling*

The theory that hydraulic pressure worsens fouling by compacting foulants stems from a plethora of experimental studies showing that FO fouls more slowly than RO and that the fouling layer in FO is easier to remove. The slower flux decline of FO at a given initial flux has been explained by the internal concentration polarization (ICP) self-compensation effect [17–19], which is unrelated to the system pressure. However, the lower effectiveness of cleaning fouled RO membranes is typically attributed to the high hydraulic pressure of the feed.

Multiple studies have compared fouling removal in osmotic membrane separation processes at different feed pressures and the same initial flux. Xie et al. [15] used a feed of 200 mg/L of alginate and 1 mM $CaCl_2$ and a glucose draw solution of varying concentration to compare RO, FO, and pressurized FO. Cellulose triacetate (CTA) FO membranes were used in all processes and cleaning was performed with DI water at high cross-flow velocity. Lee et al. [7] used a feed solution with 200 mg/L alginate, 1 mM $CaCl_2$, and an ionic strength of 50 mM, and cleaning was performed with the same feed at high velocity. CTA FO membranes were used with a draw solution of NaCl. Kim et al. [14] used CTA FO membranes with a feed of 100 mg/L alginate and 1 g/L of colloidal (approximately 100 nm) silica without calcium but with 50 mM ionic strength and an NaCl draw. A stack of permeate carriers were used as a feed spacer[1] and cleaning was performed at high cross-flow velocity with the same feed solution. Mi and Elimelech [8] used CTA FO membranes with a feed solution of 200 mg/L alginate, 50 mM NaCl, and 0.5 mM $CaCl_2$ and an NaCl draw. Cleaning was performed with a solution of 50 mM NaCl at high cross-flow velocity. Alginate, a polysaccharide that complexes with calcium to form a hydrogel [20,21], was used as a model foulant in all four studies.

Table 1 summarizes the experimental conditions and results of these four studies. Cleaning effectiveness (sometimes termed “cleaning efficiency”), which is defined as the fraction of flux lost due to fouling that is recovered by cleaning, is calculated from reported flux or normalized flux data except when cleaning effectiveness was reported. Although differences exist between the feed solutions, draw solutions, membranes, channel geometries, and cleaning methods used, all four studies varied pressure and draw concentration together to keep the initial flux fixed between trials.

Fig. 2 shows that, in this set of studies, cleaning effectiveness is not only negatively correlated with pressure but also positively correlated with draw concentration. Fig. 2a shows that, in each study, cleaning effectiveness decreased with increasing feed pressure. However, none of these studies truly isolated pressure as an independent variable because the concentration of the solution opposite the feed (called the “draw” in Fig. 2b, even in the case of a pure RO permeate) was also varied between these trials, as shown in Fig. 2b. Experiments in which both pressure and draw concentration are varied cannot distinguish between effects of feed hydraulic pressure, TMP, draw solute diffusion, and ICP, all of which differ between FO and RO and could potentially influence fouling, as will be discussed in Section 5.

Some studies have additionally explored the physical characteristics of fouling layers formed in FO and RO both in situ and ex situ. Mi and Elimelech [8] visually examined fouling layers formed in both processes, and found that FO fouling was more “soft and fluffy, indicating a loose structure.” Fouling layers created in FO and RO have also been imaged using confocal laser scanning microscopy (CLSM) to show that both alginate cakes [15] and biofilms [22] are thinner and more uniform in RO than in FO. Although this has been considered to be evidence for foulant compaction by high pressure, the ICP self-compensation effect [17] contributes to the larger foulant thickness in FO [19]. Furthermore, no justification has yet been given for why pressure should lead to a more uniform foulant layer. Ex situ measurements and images may also be affected by changes in the gel's ionic environment that occur after the fouled membrane is removed from the experimental apparatus. Changes in calcium and sodium ion concentration within the gel, such as could occur when it is rinsed or placed in a dye solution, can cause it to shrink or swell [23]. In situ visualization of FO and RO foulant layers has also been used to compare mechanisms of foulant removal [16]. Although previous studies suggested that the low pressure in FO led to a looser foulant layer that could more easily be broken up during cleaning [7,8], in situ observation of mechanical cleaning with reverse permeation revealed a similar progression of wrinkling, tearing, and peeling of full-thickness sheets of gel in both FO and RO [16].

Prior modeling has shown that foulant compaction by the high hydraulic pressure of the RO feed could be significant, but only for foulants with particular properties. Lay et al. [17] find the idea of compaction by high hydraulic pressure “contradictory to the well established critical flux concept,” [24] which implies that, “regardless of the type of driving force, the effect of membrane fouling should be comparable under similar flux and operational conditions.” However,

---

[1] At least in the high-pressure trial, but possibly in both trials.