

OSMOTICALLY-INDUCED CLEANING OF FOULED REVERSE OSMOSIS MEMBRANES IN DESALINATION

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ABSTRACT

To counteract the effects of fouling, desalination plant operators ultimately resort to chemical cleaning, which incurs system downtime, risk of membrane damage, and the generation of chemical waste. Driven by these limitations, chemical-free cleaning has emerged as a potential alternative. Recent works have demonstrated the viability of osmotically-induced cleaning (OIC), where a membrane effectively undergoes osmotic backwashing. In light of conflicting reports, however, a deeper analysis and understanding of the underlying mechanisms involved is necessary. In this work, the effectiveness of OIC is examined, underlying mechanisms are elucidated, and potential applicability in the presence of spacers is investigated. Sodium alginate is adopted as a model organic foulant and commercial RO membranes are operated to emulate fouling conditions in practice. Video footage is presented to document the cleaning procedure and refine our understanding, while important observations are noted. Experimental results demonstrate effective cleaning, demonstrating a flux recovery of 10-30% depending on experimental conditions, while membrane integrity is preserved. To investigate the effect of OIC on biofilms, a real biofilm of *Cobetia marina* is exposed to solutions of higher salinity. The results indicate that while exposure to higher salinities does impact the biofilm spread and bacterial count, the method is far from completely eliminating the biofilm or preventing its regrowth once operation is resumed.

Keywords: Reverse osmosis, Membrane fouling, Chemical-free cleaning, Osmotically-induced foulant detachment, Osmotic backwashing.



I. INTRODUCTION

Although four billion people are already experiencing water scarcity, demand for fresh drinking water is only expected to intensify, fueled by a rapidly growing population, climate change, economic expansion, and an expanding middle class [1]. A staggering report by the United Nations [2] indicated water consumption has already been increasing by one percent annually over the past decades, placing half the world population in 2050 at risk of water scarcity. To meet an ever-growing demand at the heart of the energy-water-food nexus, desalination has become increasingly important.

In spite of being the most energy efficient desalination technology [3] [4], reverse osmosis (RO) is plagued by fouling. Fouling compromises membrane performance, leading to a lower permeate flux and quality. To keep fouling in check, desalination plants typically resort to chemical cleaning to restore performance. Chemical cleaning, however, presents a series of challenges, ranging from the lack of a universal cleaning agent to chemical waste, system downtime, and membrane damage [5].

In light of these limitations, chemical-free membrane cleaning methods have emerged as a promising solution. Chemical-free cleaning employs physical mechanisms in which mechanical forces are introduced to disrupt the foulant layer and encourage its detachment. In contrast to their chemical counterparts, chemical-free methods do not generate chemical waste and could be employed continuously, reducing overall system downtime. Recent works by Tow et al. [6] and Liberman [7] demonstrate the potential of osmotically-induced cleaning (OIC), a chemical-free process in which a membrane undergoes osmotic backwash, to restore performance.

II. MOTIVATION AND RESEARCH QUESTION

Unlike other cleaning methods, the OIC process involves controlling the net driving pressure (NDP) across the membrane such that the osmotic pressure difference drives the cleaning. More specifically, the osmotic pressure on the membrane feed side is allowed to exceed that of the permeate while the applied pressure difference is held below the osmotic pressure difference. The end result is an osmotically-induced backwash of the membrane as the permeate backflows from the permeate to the feed side by osmosis. Osmotically-induced cleaning may be achieved by: (i) lowering the applied pressure below the osmotic pressure difference across the membrane [6], (ii) introducing a solution of considerably higher salinity relative to the feed [7], or (iii) implementing a combination of the two.

Alginate, an extensively studied model organic foulant, has been selected herein given its gel-forming properties and similarity to extracellular polymeric substances (EPS) encountered in fouling. During OIC, multiple mechanisms taking place are responsible for the observed cleaning. Being hydrophilic, for example, an alginate foulant film has been reported to experience isotropic swelling when exposed to a change in ionic composition. Consequently, this swelling induces wrinkling in the foulant layer, which is experiencing in-plane compression due to applied pressure, in an attempt to relieve the stresses experienced [6]. Coupled with the shear forces induced by the flow, the swelling encountered brings about the eventual detachment of the foulant layer.

While lowering the osmotic pressure difference, furthermore, the backflow of permeate water exerts a drag force on the foulant surface, ultimately facilitating its removal. In addition to swelling and backflow induced cleaning, ion-exchange has also been reported as another mechanism aiding in breaking the gel network [8]. As a divalent ion, the presence of calcium is necessary in enabling



the crosslinking across the different alginate molecules forming the foulant gel layer. When a higher salinity solution is introduced, an ion-exchange process whereby calcium is replaced by sodium can break down the network of connected foulant molecules [8]. Figure 1 illustrates the cleaning process for a fouled RO membrane. By controlling the net driving pressure, permeate backflow is induced, leading to foulant swelling and wrinkling in addition to potential gel break-down through ion-exchange if a solution of different ionic composition is introduced. These mechanisms combined bring about foulant detachment and cleaning of the membrane surface.

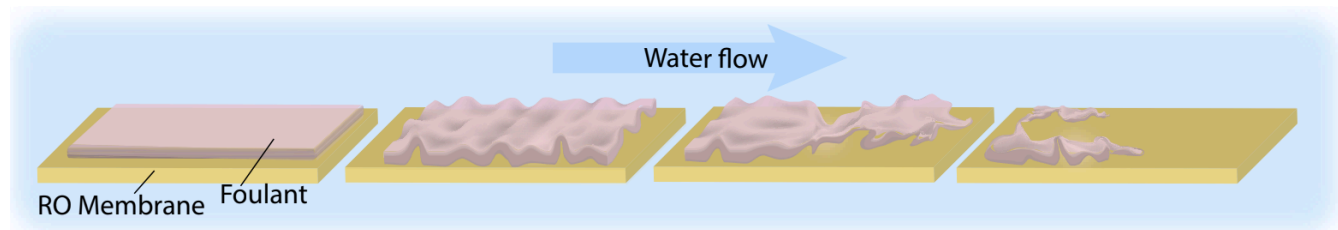


Figure 1 - Schematic illustrating osmotically-induced cleaning (OIC) for a fouled RO membrane.

Despite its potential, conflicting reports in the literature make it unclear whether and how OIC may be integrated with commercially existing RO modules for desalination. Although the work conducted by Lee and Elimelech [8], for example, demonstrated remarkable flux recoveries and cleaning efficiencies, the study focused on wastewater reclamation as opposed to desalination and featured no spacers in the feed channel. Similarly, the work by Qin et al. [9] focused on wastewater reclamation, while the work by Semiat et al. [10] investigated applicability to scaling. Other studies, furthermore, either did not demonstrate large-scale viability [6] or briefly offered a preliminary pilot demonstration without a detailed account or in-situ visualization [7].

In contrast, a study conducted by Farooque et al. [11] on commercial spiral-wound modules for seawater desalination indicates OIC is ineffective at mitigating fouling for SWRO in practice. The conflicting accounts underscore the need for further investigation into the applicability of OIC. In addition, another study by Liberman [7] suggests OIC, involving solutions with high salinity, can help mitigate biofouling by inducing plasmolysis in bacteria. Experimental data to support this hypothesis have yet to be published in connection with desalination applications.

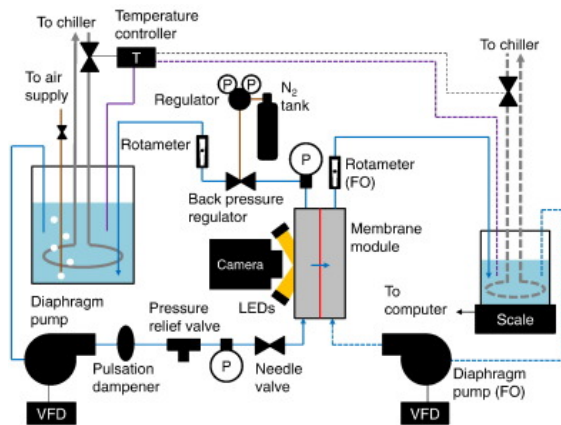
In this work, we investigate the applicability of OIC to cleaning organically fouled commercial RO membranes, featuring spacers, in brackish water desalination. The cleaning is recorded in-situ, its effectiveness is evaluated, and potential cleaning mechanisms are suggested. Experimental results demonstrate flux recoveries between 10-30% depending on experimental conditions while membrane integrity is preserved. The presence of spacers is shown to dramatically influence cleaning effectiveness. Furthermore, the effect of OIC on biofouling is further examined by exposing a real biofilm of *Cobetia marina* to solutions of higher salinity. Our results indicate that while OIC does impact the biofilm spread and the total bacterial count on the membrane surface, it does not completely eliminate the biofilm nor does it prevent its regrowth upon normal operation.

II. EXPERIMENTAL METHODS

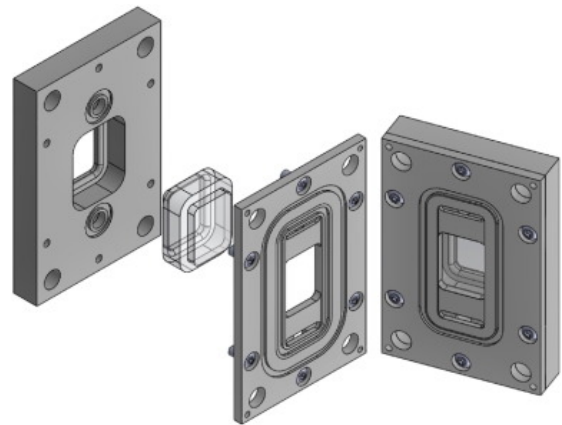
An experimental setup similar to that of Tow et al. [6] was employed to visualize the fouling and cleaning via OIC of alginate films, as given in Figure 1(a). A positive displacement pump, controlled by a variable frequency drive (VFD), was employed to maintain flow in the system. A pulsation dampener was installed to limit the effect of pulsations generated by the pump. A backpressure regulator, connected to compressed nitrogen, was selected to maintain the system pressure, measured by pressure transducers. The temperature, feed pressure and volumetric flow rate were all controlled while permeate flux was monitored using a digital scale. High rejection thin-film composite polymeric membranes (Dow FILMTEC SW30HR) with a pure water permeability (PWP) of 0.833 LMH/bar were housed in a custom-built plate and frame module, as depicted in Figure 1(b).

A feed solution with sodium chloride at a concentration of 10,000 mg/kg, sodium alginate at a concentration of 1000 mg/kg and 5 mM of calcium chloride was used in all experiments. Methylene blue (1% wt.) was adopted as a cationic dye for the foulant to improve visibility while video footage was captured to record the fouling and cleaning behavior through a transparent window (Figure 1b). Fine feed spacers (Sterlitech 17 mil diamond) were placed in the feed channel during spacer experiments, and the flux was initiated at 25 LMH for all experiments with applied pressure set to 32 bars. Crossflow velocities were maintained at 8.5 cm/s during fouling as would be typical in normal RO operation, and the temperature was maintained at 20 °C throughout the experiments with the aid of a chiller.

To initiate OIC, the applied pressure was relieved to match ambient conditions, allowing for a reversal in the NDP as the osmotic pressure difference across the membrane becomes the dominant contributor. During cleaning, crossflow velocities of 4.3 cm/s and 8.5 cm/s were selected to analyze the cleaning performances under different conditions for a duration of 30 minutes. Assuming the solution to be primarily sodium chloride, the osmotic pressure difference across the membrane for the conditions observed was calculated using Pitzer’s model to be approximately 7.5 bar for all experiments during OIC.



(a) Schematic of the flow setup



(b) Test cell with transparent window

Figure 2: Schematic diagram of the experimental flat-sheet setup for in-situ fouling and cleaning visualization [6].

In addition to the crossflow experiments, an experiment on real biofilms was conducted to elucidate the effect exposing the microbes to solutions of differing salinity has on their survival. Specifically, *Cobetia marina* (basonym, *Halomonas marina*) (ATTC 4741) was chosen to be the representative microbe due to its commercial availability and its use in marine biofilm research. Marine broth (37.4 g/L) was prepared by dissolving the powder form (2216, Difco, ATTC, USA) in filtered and sterilized DI water. *C. Marina* was stored in a frozen glycerol stock. To prepare the culture, a single drop from the glycerol stock was inoculated in the marine broth (50 ml, for *Cobetia marina*) and grown overnight (~12 h) with agitation at 25 °C (*Cobetia marina*), and the concentration of the cells produced by this method was 2.94×10^8 cells/mL.

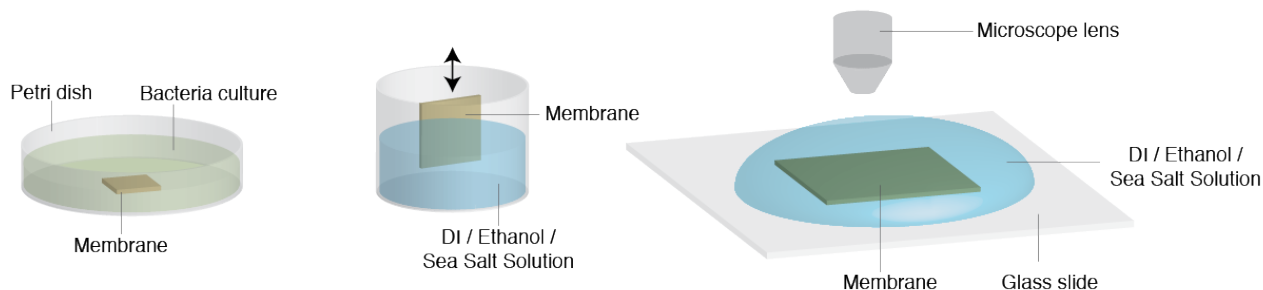


Figure 3: Procedure for real biofilm experiments.

For these experiments, the membranes were cut into 1 cm x 1 cm coupons and first sterilized with ethanol, followed by rinsing with sterilized DI water. The membranes were then glued to the bottom of a petri dish and the bacteria culture was added, such that the active layer was fully exposed to the culture as shown in Figure 3. The setup was then left on a lab shaker to allow for the formation of the biofilm. After approximately 8 hours, the dish was removed from the lab shaker. SYTO 13, a fluorescent nucleic acid, was added to the culture at a concentration of 0.3 $\mu\text{L}/\text{mL}$. The dish was then left in a dark location for 15 min to allow for the staining of all live bacteria in the culture and the biofilm.

Thereafter, each membrane sample was carefully removed from the dish and dipped into a beaker containing a) DI water, b) 3%, c) 10%, d) 20% sea salt solution or e) ethanol, to remove cells that were not attached. The membrane was then placed on a glass slide and covered completely by the sterilized solution that it was rinsed with and left for 10 min. The membrane was then observed under a fluorescent confocal microscope.

III. RESULTS

Shown in Figure 4 are the test results collected from the crossflow experiments. As the results demonstrate, membrane flux declines during fouling as alginate is deposited on the membrane surface, a process affected by the flow rate and the presence of membrane spacers. In these experiments, membrane cleaning was initiated when the normalized flux reached 50% to provide a preliminary assessment of cleaning effectiveness at high fouling rates. The membrane module underwent OIC for a duration of thirty minutes by the relieving pressure while maintaining the crossflow velocity indicated.

Achieving flux recoveries between 10-30% depending on experimental conditions, OIC is shown to be a promising alternative to traditional chemical cleaning methods. For all experiments conducted,

the membrane rejection was maintained above 97% before and after cleaning, indicating membrane integrity was preserved during the cleaning procedure. The mechanism underlying OIC could be observed from the videos recorded. Video stills at representative moments of the process are shown in Figure 5. As depicted, it can be observed that the OIC process appears very efficient at the removal of the foulant layer when there are no feed spacers. Partial delamination was observed followed by the removal of the layer by the shear forces induced by the flow.

Counter-intuitively, OIC with low crossflow velocity demonstrated better cleaning, a result recorded experimentally (Figure 4) and captured on video (Figure 5). At high crossflow velocity, the forces on the delaminated portion of the foulant contributes to the fracturing of the film, whereas low crossflow velocity allows the entire foulant layer to delaminate completely first. In the presence of spacers, nonetheless, swelling of the foulant layer was also observed, albeit taking almost double the amount of time for partial delamination of the film. In this case, the OIC process was shown to be ineffective as the foulant layer has to break into smaller pieces first before it is removed by the flow. It can be seen that the presence of spacers acts as an impediment to cleaning, providing a potential justification to the discrepancies encountered in the literature and underscoring the importance of including spacers in future studies on OIC.

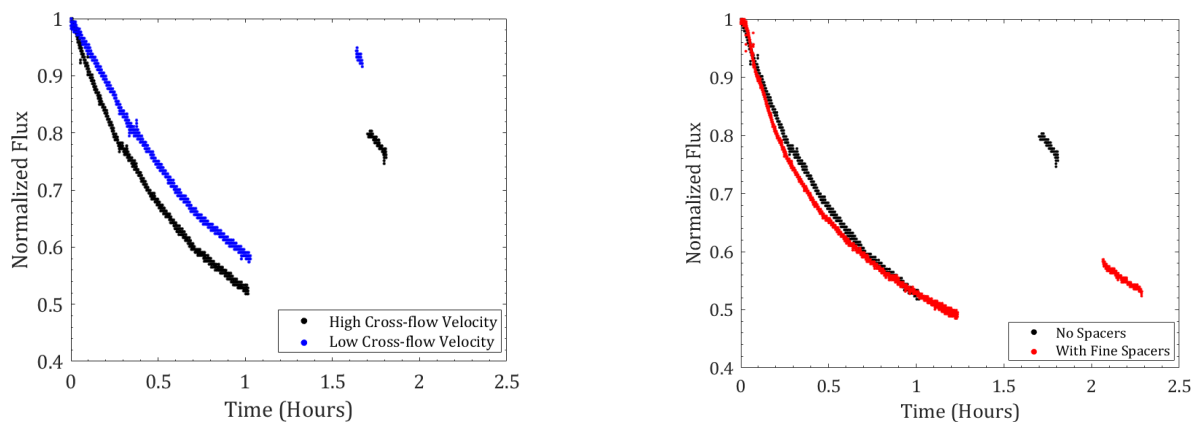


Figure 4: Normalized flux fouling and cleaning performance comparisons: (a) between spacer-less feed channels under low and high crossflow velocities; and (b) between spacer-less and spacer-filled feed channels under high crossflow velocities.

Apart from crossflow experiments, Figure 6 summarizes the results obtained for the experiments conducted on the real biofilm exposed to high salinities. The figure shows the confocal microscopy images of the membrane surfaces after being rinsed and soaked in five different solutions. The green fluorescence indicates live bacteria that are still left on the surface. The images measure 1024 x 1024 pixels, and each pixel was classified as bacterial or background using the threshold method. The percentages of surface coverage were then given by the number of bacteria pixels divided by total pixels.

To the best of the authors' knowledge, Figure 6 is the first to present microscopic images of a biofilm before and after exposure to solutions of varying salinities for cleaning in a desalination context. As shown in the figure, it can be noted that the coverage of the bacteria remains very high even with 10% sea salt solution. The 20% sea salt solution gets the coverage down to 61% of the membrane, but it is demonstrated that most of the bacteria are still alive. The decrease in coverage with increasing salinity is due to both, the decrease in concentration of live bacteria counts on the surface and the shrinkage of the cells as they lose water to the solution. For comparison, the treatment of the sample in ethanol

mostly gave images that were almost completely dark and the image shown in Figure 6e shows a remnant colony of live bacteria. The results indicate that while introducing solutions of higher salinity through OIC does impact the biofilm, it is not nearly sufficient to completely eliminate the bacteria or prevent their regrowth once operation is resumed.



Figure 5: Video stills of the swelling process with: (a) no spacer and high crossflow; (b) no spacer and low crossflow; and (c) fine spacer and high crossflow. The dark blue solid represents the foulant that formed on the surface of the membrane.

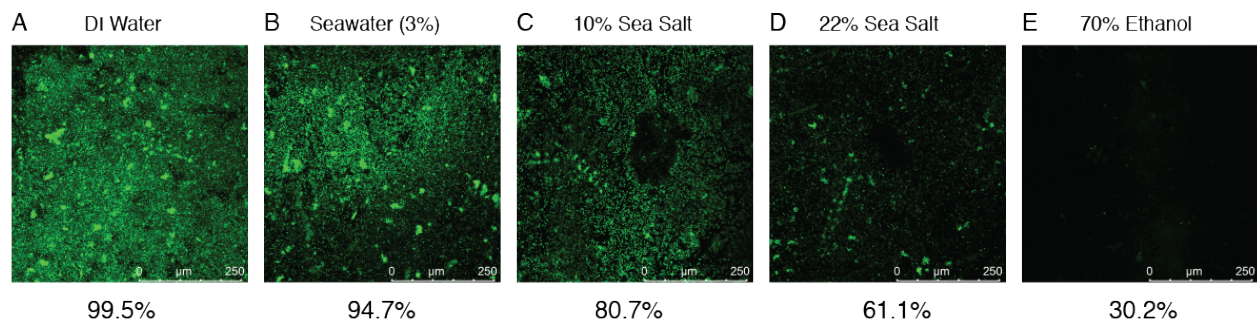


Figure 6: Confocal microscopy images of the membrane surfaces that were rinsed and soaked in (a) DI Water, (b) 3%, (c) 10%, (d) 22% sea salt solution. An image, showing some remnant live bacteria on the surface after soaking the membrane surface in 70% ethanol solution, is added for reference. The percentages at the bottom of the images were derived from counting the pixels that exceed a threshold limit, and indicate how much of the area is covered by bacteria.

IV. CONCLUSIONS

In this work, the potential of osmotically-induced membrane cleaning (OIC) in desalination has been explored. Commercial RO membranes were adopted along with a brackish feed water stream, and alginate was introduced as a model organic foulant. Experimental results demonstrate the viability of the method on a coupon-scale flat sheet setup. Flux recoveries between 10-30% were recorded, and the salt rejection remained constant after cleaning, indicating membrane integrity was preserved. In spite of its potential, the cleaning effectiveness of OIC dropped dramatically in the presence of spacers, acting as an impediment to membrane cleaning. Interestingly, experimental results recorded and video footage captured suggest that membrane cleaning by OIC was more effective at lower crossflow velocities as higher velocities caused film fracturing. Towards the end, experiments conducted on a real biofilm exposed to high salinity cleaning solutions indicate that while OIC could affect the biofilm, it does not completely eliminate it or prevent its regrowth once normal operation is resumed.

IV. ACKNOWLEDGEMENTS

This research is supported by the Abdul Latif Jameel Water and Food Systems Lab (J-WAFS) at the Massachusetts Institute of Technology (MIT). Omar Labban acknowledges the funding received under the J-WAFS Rasikbhai L. Meswani Fellowship for Water Solutions and the Den Hartog Travel Award in Mechanics at MIT. Grace Goon acknowledges the financial support provided by a postgraduate fellowship from DSO National Laboratories Singapore. Zi Hao Foo acknowledges the funding provided by the CN Yang Scholars Programme Office, Nanyang Technological University (NTU). The authors acknowledge Lara Ketonen for contributing towards figure design.



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