A NOVEL AIR BACKWASHING METHOD FOR REVERSING WETTING IN MEMBRANE DISTILLATION

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Abstract

The most critical failure mode of the desalination technology, membrane distillation, is wetting of the hydrophobic membrane, which allows the feed to leak through and contaminate the pure distillate. In order to reverse wetting that has begun to occur, a common technique used is allowing the full dryout of the membrane. However, this process is slow, and can cause considerable downtime as well as allow for salt deposits to stay on the surface of the membrane, which may cause further wetting. Here, a new method is proposed to reverse wetting: pushing pressurized air through the wetted membrane from the distillate side, thus forcing out wetted feed water, instead of merely allowing it to evaporate. To test its effectiveness, the liquid entry pressure (LEP) of the membranes was tested before and after the pressurized-air process. The results reveal that the air backwashing method was very effective, restoring LEP to nearly 85% of the original value, without requiring the long delay times associated with dryout. SEM images after the air-pressurizing process showed that the pressurized air did not significantly damage the membrane. It is therefore recommended that this new method be used in restoring the hydrophobicity of wetted membranes.

1. Introduction

Membrane distillation is a thermal desalination process that uses a temperature gradient across a hydrophobic membrane to evaporate water from a saline solution and condense it as pure water on the other side of the membrane [Warsinger, et al. (2015)]. Membrane distillation has proven uniquely advantageous for waters with high salinity, and it is thought to be superior to reverse osmosis in fouling resistance [Warsinger, et al. (2014)]. In membrane distillation, the two primary failure modes caused by foulants are flux decline caused by the blocking of membrane pores, and wetting, where saline feed water leaks through the membrane [Franken, et al. (1987), Goh, et al. (2013)]. Of these, wetting is by far the biggest concern, for it causes contamination of the very pure distillate, ruining the desalination process. Wetting of the MD membrane prevents the use of MD for more fouling-prone feed water [Gyra (2005); Gyra, *Calcium Sulfate,* (2008);

Gyra, *Fouling in direct contact,* (2008)] as recent evidence shows that fouling and wetting are inter-related phenomena in MD. Intermittent operation has also been a primary contributor to fouling, which occurs especially with solar-powered MD [Banat et al., *Desalination,* (2007); Banat et al., *Performance,* (2007); Guillen-Burriesa et al. (2013); Koschikowski (2003)]. The primary method to treat wetting is to shut down the MD system and allow the membrane to dry out [Warsinger et al. (2014)]. However, this method leaves salt crystals on the membrane surface and within the pore structure, as the feed solvent evaporates leaving its solutes behind [Gyra et al. (2009), Karakulski et al. (2005)]. This may play a role in the decrease in performance of membrane distillation systems, as the resistance to wetting declines over time. The ability to avoid wetting in MD is described by the liquid entry pressure (LEP). LEP is the static pressure required for the liquid feed to overcome the resistance forces (due to membrane's hydrophobicity) and penetrate the membrane's pores, thus causing wetting to occur. LEP is described in the modified Young-Laplace equation:

$$LEP = \frac{2B_g \sigma \cos\theta}{R_{\max}} \tag{1}$$

where B_g is a geometric factor for pore shape (B_g = 1 for cylindrical pores), σ is the surface tension of the feed solution, θ is the contact angle for the membrane and solution interface, and D_{max} is the maximum pore size (i.e., diameter) of the membrane.

Surface cleaning of the membrane is often used to help avoid MD membrane wetting, but even so, the membrane tends to become more readily wetted over time in many feed solutions with common foulants. To treat wetting, previous studies have suggested backwashing the membrane with a non-aqueous liquid that has a low surface energy, so it readily wets the membrane [McAlexander and Johnson (2003)]. McAlexander et al. found that this provided some restoration of membrane performance, but the heating and the liquid required will likely make this method costly to implement. Moreover, the removal of non-aqueous liquid may be incomplete, in addition to its possible interaction with the membrane material itself. Furthermore, this method requires significant downtime of operation (15 hours in lab tests), more than most dry-out trials which may require as little as several hours in lab conditions [McAlexander and Johnson (2003), Warsinger et al. (2013)]. These durations are much longer than the pressurized air method presented here, which may only need minutes. Also, backwashing with other liquids may not work for extremely hydrophobic membranes, as the surface energy of the liquid may not wet such membranes. In the present work, we present a new method for restoring the hydrophobicity of MD membranes: air backwashing with pressures exceeding LEP. The idea behind this is that the high-pressure air will be able to force out any water and foulants trapped within the membrane pores, allowing restoration of a dry state without allowing saline liquid to evaporate on and within the membrane. For this study, we tested the liquid entry pressure of the membranes to saline solutions. We then attempted to reverse wetting using dry out as well as air backwashing without allowing dry out. Upon retesting LEP, we determined the extent to which each method brought the membrane back to its pre-wetting performance. Trials were performed at several different salinities using aqueous NaCl solutions.

2. Methodology

The membrane used was a 0.2 μm PVFD Millipore Immobilon-PSQ part # ISEQ 000 10. Reagent grade NaCl was purchased from Sigma-Aldrich.

Liquid entry pressure was measured using a syringe-pump based LEP setup to pressurize a saline solution until the pressure was high enough for the solution to permeate through an MD membrane (**Figure 1**). This system has been described previously and is similar to a number of previous designs [del Carmen Garcà-Payo et al. (2000), Guo et al. (2015), He et al. (2011)]. To test LEP, a sample of a new unwetted membrane was held in a 13 mm syringe membrane holder (GE healthcare biosciences, Product Code 1980-001). The membrane was exposed to saline solution on one side and ambient air on the other side. A syringe pump (PHD 22/2000, Harvard apparatus) was used to push the saline solution against the membrane at constant volume increments to generate a stepwise increase in pressure against the membrane. There was a 12 second hold between each step in which constant pressure was maintained. The pressure difference across the membrane was monitored using a USB pressure transducer with a precision of ±0.3 kPa (P409, Omega). When the LEP was reached, the pressure readings had a negative slope during the pauses between steps indicating leakage of water through the membrane (see **Figure 4**).

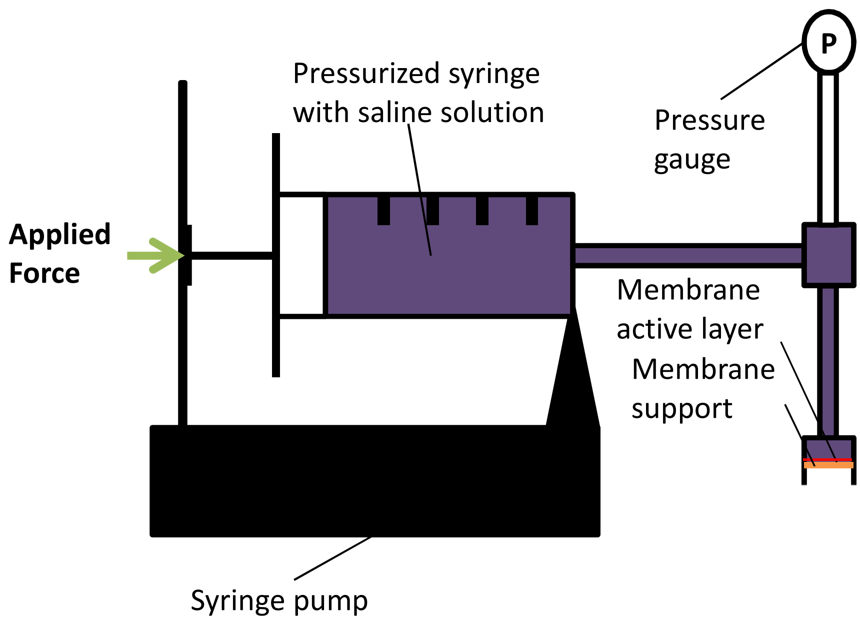

Figure 1. Liquid Entry Pressure (LEP) testing setup, using pressurized syringe, a saline solution, and a small membrane holder.

After the membrane had wetted due to surpassing the LEP, the membrane was removed from the system and either allowed to air-dry for a minimum of 24 hours or immediately had pressurized air forced back through it from the side opposite the one which had been in contact with the saline solution (**Figure 2**).

The pressurized air was forced though the membrane in 10 second pulses at approximately 450 kPa, a pressure well above the LEP of the membrane. Such high pressures may not be necessary, but should be more effective. The pressurized air line was coupled with the membrane holder using a custom design made by modifying the original GE membrane holder (**Figure 2**).

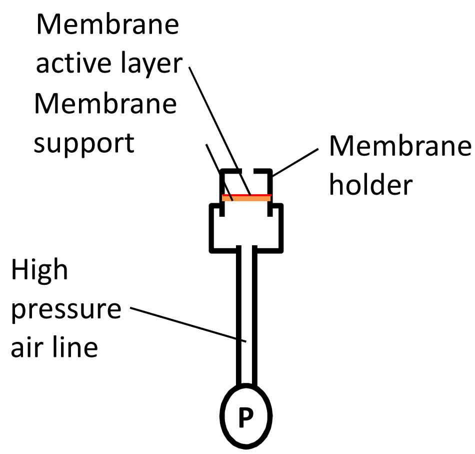

Figure 2. Setup for adding forced air into the gap side of the membrane holder.

3. Results and Discussion

This new method of injecting air into a previously wetted membrane substantially restored LEP. As seen in **Figure 3**, the forced air trials only exhibited a slight decrease from the membranes' original LEP of about 260 kPa (about 15% at low salinity). However, it was difficult to judge the comparative performance of the dryout case, as that membrane sample had a lower LEP and relatively large error bars. While both membranes were the same part number from Millipore, the most likely explanation is that the maximum pore size on the sample with lower LEP was 2-3 times larger, which can be caused by the imperfect process of phase inversion used to create the membrane.

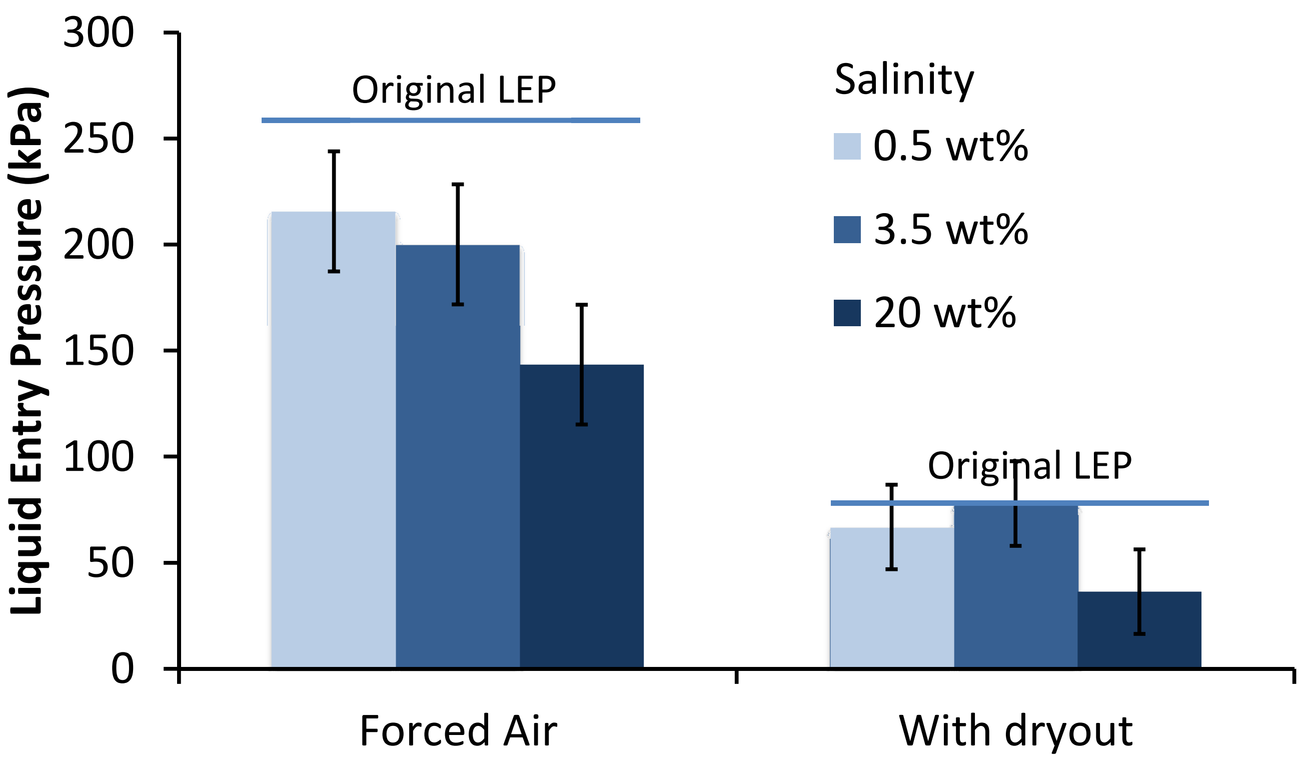

Figure 3 LEP Values for Experimental Conditions Using Different NaCl concentrations

Another important trend was the impact of salinity for the forced air trials. In more saline cases, the recovery of LEP was worse. This trend can be observed despite the large error bars, which are calculated from the standard deviation between trials for the LEP of previously unwetted membranes. Variation between data sets may be due to variation of the membrane itself, which may vary slightly in pore size, thickness, and porosity throughout its surface. The membrane circles that were cut out for the trials in each set of experiments (forced air and dryout sets) were taken from adjacent parts of the membrane, and so were more predictable and consistent between one another. The more saline conditions are expected to impair LEP more, as they leave more salt trapped in the membrane. A representative LEP trial is seen in **Figure 4**.

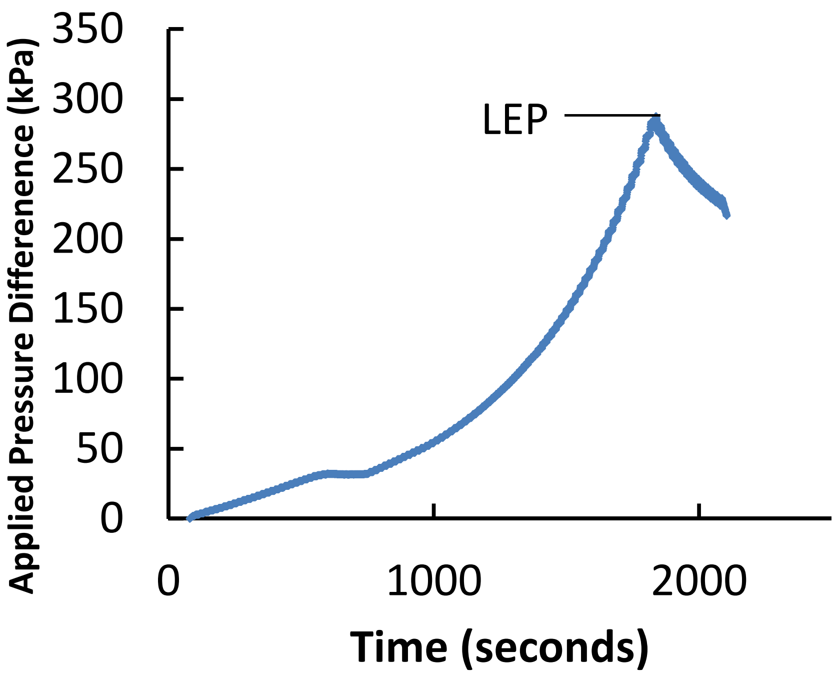

Figure 4 Applied pressure difference versus time, for the 3.5% w.t. NaCl trial after air cleaning had been performed.

Once LEP had been exceeded (**Figure 4**), a rapid drop in pressure behind the membrane was observed, as the slope of the line during the 12-second hold became and negative thus indicating that water rushed through the membrane. This peak pressure is the LEP of the membrane sample.

It is worth noting the wetting method here, using a rising pressure to exceed LEP, is different than the wetting that occurs from fouling during operation.

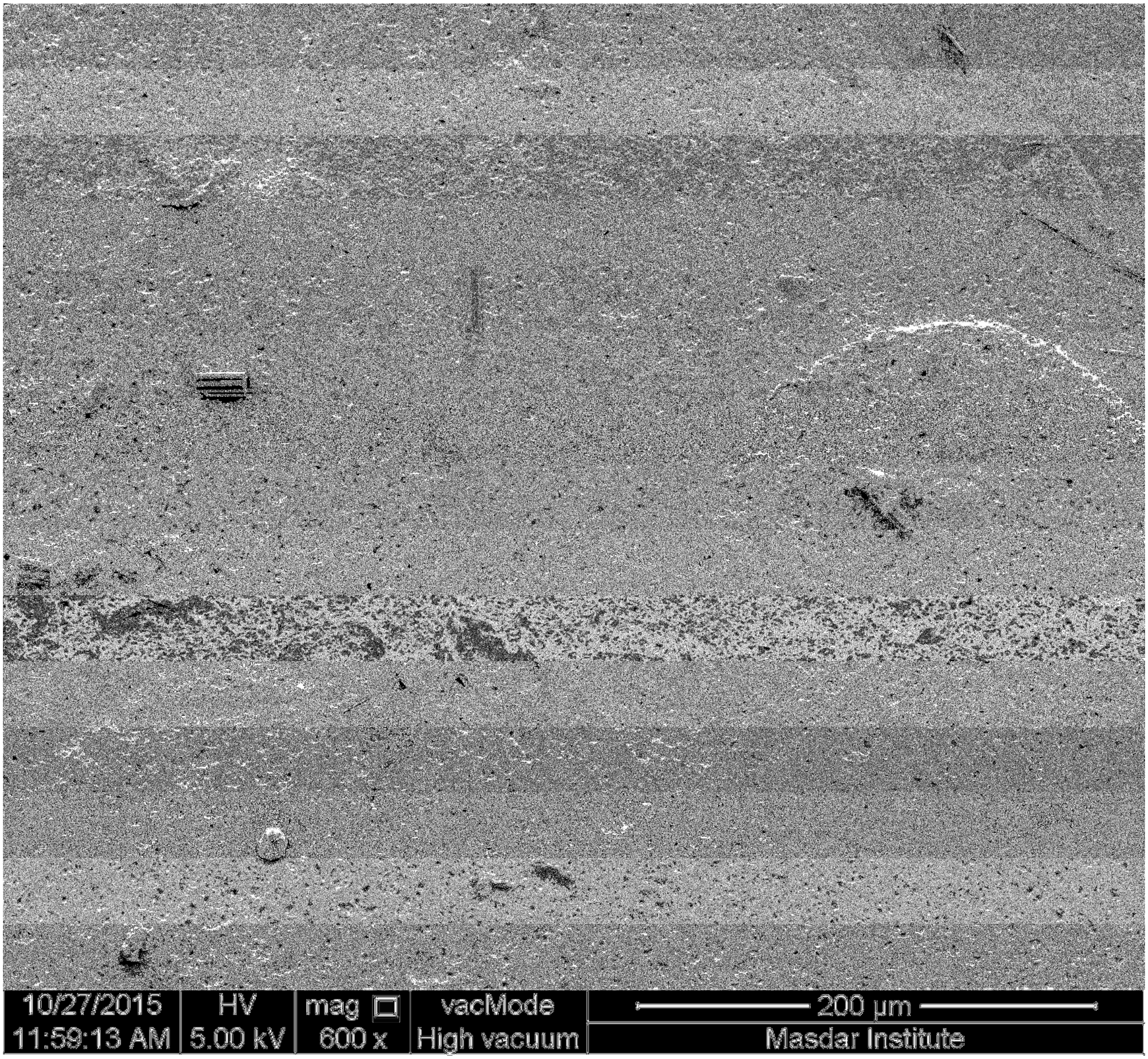

Figure 5 SEM image of feed side of membrane surface, in a trial with 3.5%w.t. NaCl and forced air cleaning

Membranes from both the forced air and dryout cases appeared similar. As seen in **Fig. 5**, the membrane shows superficial crystals and membrane deforming. The salt remaining on the surface did dry out afterwards, as the membrane had to be allowed to dry in both cases to examine under SEM. The crystals appear superficial, which may mean they are not likely to cause wetting. Circular patterns are evident from membrane deforming. Notably, the deforming occurred during the LEP testing part: the metal spacer that held the membrane has small circular holes, which are clearly visible. Both dryout cases and air recharging cases had this same pattern, and no difference seemed to be present from the addition of high pressure air.

4. Conclusions

Membranes tested with higher salinity solution experienced worse flux restoration for both the air backwashing and dryout cases. This indicates that salt deposition inside the membrane pores plays an important role in membrane wetting. The air backwashing method worked well, restoring LEP within up to 75% of the original value. An autopsy of the membrane with SEM indicated that while the LEP test itself warps the membrane, there did not appear to be large-scale deformation in the air backwashing approach. This comparison and the high LEP after air backwashing treatment implies that even at very high pressures, pressurized air does not significantly damage the membrane. As the air backwashing method was about two orders of magnitude faster than full drying out of the membrane, it requires less downtime than the currently used dryout method. Because of this, it could conceivably be done frequently to allow membranes to last longer and operate better with more adverse feed solutions.

5. Acknowledgement

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