Contents lists available at ScienceDirect

Desalination

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

Air gap membrane distillation for hypersaline brine concentration: Operational analysis of a full-scale module–New strategies for wetting mitigation

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ARTICLE INFO

Keywords: Membrane distillation Wetting mitigation Brine concentration Draw solution recovery

ABSTRACT

Membrane distillation (MD) is a thermal separation process which possesses a hydrophobic, microporous membrane as vapor space. A high potential application for MD is the concentration of hypersaline brines, such as e.g. reverse osmosis retentate or other saline effluents to be concentrated to a near saturation level with a Zero Liquid Discharge process chain. In order to further commercialize MD for these target applications, adapted MD module designs are required along with strategies for the mitigation of membrane wetting phenomena. This work presents the experimental results of pilot operation with an adapted Air Gap Membrane Distillation (AGMD) module for hypersaline brine concentration within a range of 0–240 g NaCl /kg solution. Key performance indicators such as flux, GOR and thermal efficiency are analyzed. A new strategy for wetting mitigation by active draining of the air gap channel by low pressure air blowing is tested and analyzed. Only small reductions in flux and GOR of 1.2% and 4.1% respectively, are caused by air sparging into the air gap channel. Wetting phenomena are significantly reduced by avoiding stagnant distillate in the air gap making the air blower a seemingly worth- while additional system component.

1. Introduction

Membrane distillation (MD) is a combined membrane and evaporation process which possesses a hydrophobic, microporous membrane as vapor space. Typical operation temperatures lie between 30 and 80 °C [1–3]. Membrane distillation utilizes a vapor pressure gradient across a hydrophobic membrane commonly induced by a temperature gradient. Low grade waste heat [4] can be used to apply the necessary thermal energy required to power the process. Many applications can be targeted with MD [5–12] but still require specific adaptations to the respective module design for a commercial break-through.

A high potential application for MD is concentrating hypersaline brines, e.g. reverse osmosis (RO) discharge or osmotic draw solutions for Forward Osmosis (FO). Brine disposal in desalination plants, power plants, dyeing plants etc. is a current major issue and governments are pushing towards solutions such as zero liquid discharge (ZLD). The United States of America had imposed a ZLD on saline discharge from power plants in the 1970's whereas China and India have followed suit in recent times [13]. In 2011, in the city of Tirupur in India, the government had to close down around 750 dyeing plants on account of salty wastewater discharge compliance [14]. Membrane distillation as a solution to the problem on brine discharge has been acknowledged by several authors in their studies on the concentration of different brines [15-20]. However, findings on full scale module pilot experience are scarce yet required urgently in order to drive a focused development of the technology for this application. Where most of the work conducted with pilot plants using real brines and field studies in the years 2007-2014 focuses on seawater or brackish water desalination [21–25], starting around 2014 more work was conducted on the use of MD for the concentration of other saline feed sources such as RO discharge from coal seam gas produced water [26] or simply higher concentrations of sodium chloride [27,28]. Further recent pilot scale studies on seawater desalination with full scale MD modules can be found with [29,30]. On a bench scale, more work is available that focuses on higher salt concentrations [31-36], however with drawbacks for result

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https://doi.org/10.1016/j.desal.2018.06.012

Received 6 February 2018; Received in revised form 20 May 2018; Accepted 28 June 2018 0011-9164/ © 2018 Elsevier B.V. All rights reserved.







transferability to a larger scale due to the respective selection of operational parameters such as driving force temperature difference and partially very small effective membrane areas e.g. 14.4 cm^2 [32]; the effects of which on the process are discussed in [20].

For MD to become a serious commercial option in the technological landscape of brine treatment, some barriers must yet be overcome. Besides the mentioned piloting experience in relevant environment the topic of membrane wetting, fouling and regeneration require further attention from the research and academic community [37,38]. The works that can be found on wetting phenomena in membrane distillation, a selection of which is given here [39–44], have shown distinctly that wetting and reduction of liquid entry pressure or hydrophobicity are issues that are affected by operational parameters common to membrane distillation. A vital observation regarding mechanisms of wetting is that liquid contact on both sides of the membrane inside an MD module or test cell set-up promotes the effect of unwanted wetting even when the system is in standstill. This can be observed well in the data collected from a long- term pilot site in Pozo Izquierdo, Gran Canary, Spain [23] where the distillate conductivity value showed an increase at every morning start- up operation but was diluted by the onset of permeate production in the first time-period after operation initiation. This effect is also discussed on a more fundamental level by [40] who argues that "water bridges" are formed via the pore across both sides of the membrane which allows the passage of salt into the distillate channel. Since Direct contact membrane distillation (DCMD) and permeate gap membrane distillation (PGMD) have constant water contact on both sides of the membrane, the chances of wetting occurring are higher in these channel variants according to theory. The MD module utilized on Gran Canary Island was a PGMD module optimized for seawater desalination [45]. In this application, a small amount of distillate contamination during the morning start- up was not problematic for the overall quality of the daily product water batch. However, it is assumed that higher the salt concentration of the feed solution. such as in an industrial brine concentration application, the more severe the contamination of the distillate product through membrane wetting is expected to become. Thus, for an operation of MD in hypersaline concentration applications, an AGMD module seems a more sensible option from a perspective of wetting mitigation and control. An air gap channel configuration gives the possibility to avoid liquid contact with the membrane on both sides and have the option of accessing the air gap for measures to reverse and or control wetting.

In this work, a full scale AGMD module with novel access points to the air gap is designed and then used in a long- term operation study with highly concentrated Sodium Chloride (NaCl) brine. Key performance indicators (KPI) such as flux, Gained Output Ratio (GOR) and thermal efficiency, are obtained from the study for a range of concentrations from tap water to \sim 250 g NaCl/kg (Conductivity 0.6 mS/cm to \sim 250 mS/cm). The air gap access points at the top of the module enable active draining of the channel with low pressurized air from an air blower. Another method of improved draining is investigated by increasing the number of distillate outlets at the bottom of the air gap; both with and without the additional assistance of low pressurized air. The effects of the new draining methods on the AGMD module's performance are evaluated by comparing the measured KPI's to a standard operation without improved draining. After selection of the most promising method, the effect of its application on distillate quality is studied within the 881 hour-long operation with hypersaline brine as feed. Distillate values for operation without improved draining are also obtained and compared.

This work was carried out within the EUROSTARS project ComFORMD (01QE1611), which is an implementation project for the coupling of a forward osmosis (FO) pilot system to a reverse osmosis system and an MD system. A NaCl solution was selected as draw solution to be concentrated by the MD loop of the hybrid FO-MD pilot plant. Results presenting data from the combined operation will be presented elsewhere as this study was conducted on the MD pilot in solitary



Fig. 1. DCMD (left) and AGMD (right) channel configuration variants.

operation.

2. Materials and methods

2.1. MD basics

Mass transfer in membrane distillation is described as a combination of molecular and Knudsen diffusive transport mechanisms. Heat transfer depends strongly on the selection of membrane material and spacer induced flow regime [46,47]. Heat and mass transfer in (air gap) membrane distillation has been well investigated [35,48] and will thus not be explained in detail here. A comprehensive reference for fundamentals can be found with [47]. For different applications and different energy supply scenarios, a variety of channel configurations have been established in MD. Two basic variants are displayed in Fig. 1.

The main difference between the two channel configurations Direct Contact Membrane Distillation (DCMD) and Air Gap Membrane Distillation (AGMD) is the additional air gap in AGMD between the hot feed solution and the cooling or permeate side. DCMD can also be used for membrane contactor processes and osmotic membrane distillation applications. It the most commonly studied and most simple variant of MD. However, AGMD enables internal heat recovery inside the module as any liquid can be used on the cooling side e.g. the feed solution itself. On the other hand, DCMD has lower overall heat and mass transfer resistances which come with higher fluxes at the cost of simultaneously higher conductive heat transfer which can result in cut backs to the thermal efficiency of the process. The air gap in AGMD created by the addition of an impermeable film towards the cooling side, provides a much higher insulation between the channels. This lowers the flux compared to DCMD but also reduces the conductive heat transfer across the combined membrane and gap. A comparison of the two variants is given by [49]. By applying vacuum to the air gap, diffusive mass transfer resistance can be reduced by a large fraction. The variant is then named vacuum Air Gap Membrane Distillation (v-AGMD). A further common variant is permeate gap membrane distillation (PGMD) in which the distillate accumulates in the distillate channel filling it with liquid [50].

The key performance indicators used for operational analysis in this work are as follows:

Distillate output \dot{m}_d , is defined as the amount of distillate produced in one hour of operation:

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