

Contents lists available at ScienceDirect

# Journal of Membrane Science

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



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# Effect of humic-acid fouling on membrane distillation

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## ARTICLE INFO

Article history: Received 19 October 2015 Received in revised form 21 December 2015 Accepted 23 December 2015 Available online 6 January 2016

Keywords: Membrane distillation Fouling Humic acid Kelvin effect Vapor-pressure reduction

## ABSTRACT

Membrane distillation (MD) can concentrate non-volatile solutes or remove volatiles and dissolved gases from an aqueous feed. A microporous hydrophobic membrane provides a barrier between the hot feed and cold distillate. Although MD can operate at ambient pressure and moderate temperatures, use waste heat, and treat wastewater via an MD-bioreactor, it has problems such as temperature polarization, liquid weeping to the distillate side, and membrane fouling. Prior studies speculated that fouling can add a heat- or mass-transfer resistance, or cause a vapor-pressure reduction owing to the Kelvin effect, but did not isolate these effects. This study confirms that the vapor-pressure depression owing to the concave interface in the small pores of the fouling layer is a dominant cause of the 25–63% flux reduction observed for humic-acid fouling on PTFE and PVDF membranes. This study underscores the importance of selecting MD membranes based on their pore-size distribution rather than just their nominal diameter in order to maximize the contribution of Knudsen diffusion. It suggests the development of dual-layer membranes having a thin hydrophilic layer with relatively large pores overlying a hydrophobic layer with a typical MD membrane structure in order to mitigate the vapor-pressure reduction owing to membrane fouling.

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#### 1. Introduction

Membrane distillation (MD), which was first described in a patent by Bodell [1], is a technology for concentrating non-volatile solutes and impurities or removing volatile components and dissolved gases in a feed stream. It is based on evaporating the volatile materials in the feed at a temperature below the boiling point of the solution. For aqueous feed streams MD employs a hydrophobic microporous membrane that prevents liquid on the feed side from wicking through the membrane to the distillate side while maintaining reasonable vapor-diffusion rates. The driving force in MD is the difference in vapor pressure across the membrane. The latent heat-of-vaporization is supplied by the sensible heat in the hot feed stream. Comprehensive reviews of MD have been done by Lawson and Lloyd [2], Alklaibi and Lior [3], El-Bourawi et al. [4], Gryta et al. [5], Alkhudhiri et al. [6], Camacho et al. [7], and Warsinger et al. [8].

MD offers several advantages relative to other separation technologies. These include operation at atmospheric pressure and moderate temperatures that translate to reduced operating and

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capital construction costs, use of non-selective highly permeable membranes, complete rejection of particulates and nonvolatile solutes, reduced membrane fouling, and the ability to be integrated into a hybrid process such as the membrane distillation bioreactor (MDBR) [9]. As such, MD is particularly well-suited for wastewater treatment via MDBR technology and concentrating heat-sensitive solutions such as those containing biomaterials and fruit juices. Moreover, MD provides a possible use for low grade waste heat (T < 100 °C).

However, MD is confronted with several problems. These include temperature polarization that refers to a reduction in the temperature difference across the membrane owing to heattransfer resistances on the feed and distillate sides of the membrane [10], lower fluxes than conventional membrane processes such as nanofiltration and reverse osmosis [2], weeping or flow of liquid through the membrane [2], membrane scaling owing to sparingly soluble inorganic solutes [11], fouling owing to organic solutes, and biofouling [12-17]. Increasing the flux in MD translates directly to maintaining the maximum possible difference in vapor pressure across the membrane that in turn means minimizing temperature polarization. Phattaranawik et al. [18] have shown that temperature polarization can be mitigated by the use of a spacer mesh in the feed and distillate channels. The MD flux is also affected by membrane scaling and fouling that could add resistances to the heat transfer and mass transfer, and also can

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reduce the vapor pressure as has been shown recently by Chew et al. [19].

A comprehensive review of fouling and its control in MD has recently been published by Warsinger et al. [8] and Tijing et al. [20]. Fouling in MD occurs in the form of inorganic scaling, particulate or colloidal fouling, natural organic matter (NOM) fouling, and biofouling [2,11]. Scaling owing to the precipitation of inorganic solutes occurs only for nearly saturated feed solutions [21,22] such as in the continuous MD crystallization (CMDC) process [23]. NOM in the form of humic acids, proteins, aminosugars, polysaccharides and polyhydroxyaromatics, and biofouling caused by the growth of bacteria on a membrane can create a fouling layer with very small pores or free volume that can offer a resistance to both heat and mass transfer in MD and possibly can have other effects as well [11,19,24–26].

In their study of humic-acid fouling in MD, Srisurichan et al. [12] observed a 35% flux decline over 9 h. By comparing their MD experiments using distilled deionized (DI) water to those with humic-acid fouling, they inferred that the fouling layer offered an appreciable resistance to heat transfer. In his study of using MD to concentrate saline wastewater containing proteins and polysaccharides, Gryta [11] observed a 70% flux decline over 55 h that could not be explained by the added heat-transfer resistance owing to an estimated fouling-layer thickness of 90  $\mu$ m. Gryta speculated that the disparity between his predictions and observations might be caused by an added hydraulic resistance owing to the fouling layer. In their studies using an MDBR to treat a synthetic wastewater, Phattaranawik et al. [13] observed flux declines of over 80% in 5-7 days that could not be explained by any added resistance to heat transfer. They also speculated that the fouling layer might cause a hydraulic resistance to liquid-water transport to the membrane. In a similar study Goh et al. [15] observed a flux decline of 51% over 23 days owing to a biofouling layer whose thickness of 20 µm determined by confocal microscopy could not offer any significant heat-transfer resistance, but which they speculated could result in an added resistance to mass transfer. In an MD study using two sludges with different hydrophilicities, Goh et al. [16] observed a 60% flux decline over 180 h owing to a biofouling layer having a thickness of 7.4-15.1 µm determined by confocal microscopy that again could not contribute a significant heat-transfer resistance. Gravimetric experiments during evaporation of water from the two sludges indicated a significant vapor-pressure depression. In addition, determination of the pore-size distribution for the two sludges using evapoporometry [27–29], which is based on the relationship between pore diameter and vapor pressure dictated by the Kelvin equation [30,31], indicated average pore diameters for the two sludges ranging between 5 and 10 nm. Hence, they concluded that the large flux decline was due primarily to a vapor-pressure depression owing to the very small pores in the biofouling layer.

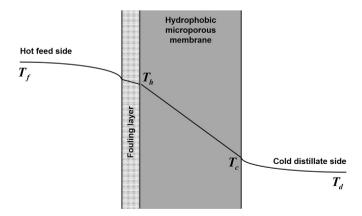
Chew et al. [19] subsequently adapted the MD model of Schofield et al. [10] to include both an added heat-transfer resistance and a vapor-pressure depression owing to the presence of a fouling layer with very small pores. Their model was able to explain the flux declines observed by Gryta [11], Phattaranawik et al. [13], and Goh et al. [15,16] based on a vapor-pressure depression owing to small pores whose average diameter ranged between 4 and 9 nm. In the absence of any comprehensive data set for MD fouling, Chew et al. [19] used their model to recast the data of Hanbury and Hotchkiess [32] for MD with a DI water feed, which correlated well with the original model of Schofield et al. [10], to show how these same data would be widely scattered if there were a vaporpressure depression owing to an average pore size of 10 nm. However, a problem with using the modified model of Schofield et al. [10] to correlate data for fouling in MD is that the parameters extracted from the correlation involve combinations of the heattransfer resistances of the fouling layer, feed side, distillate side, and the membrane in addition to the mass-transfer resistance of the membrane. The vapor-pressure-depression effect enters as an adjustable parameter in the ordinate involved in correlating the data. As such, it is difficult to conclude with any certainty what effects a fouling layer has on MD based on correlating the data via the modified correlation of Schofield et al. [10] developed by Chew et al. [19].

This brief review indicates that biofouling and fouling owing to organic matter in MD is not well-understood. It is questionable whether the marked flux decline observed by several investigators can be explained by any additional resistance to heat transfer offered by the thin fouling layers. It has been speculated that the unexplained flux decline might well be due to an additional masstransfer or hydraulic resistance in the fouling layer, although no definitive experimental studies have confirmed this. The speculation of Goh et al. [16] that the small pores in a biofouling layer might cause a vapor-pressure depression was shown to be possible in the modeling study of Chew et al. [19]. However, no data are available to confirm definitively that vapor-pressure depression in fact can occur during MD fouling. Whereas Srisurichan et al. [12] were able to explain the flux decline observed in their humic-acid fouling studies by incorporating a heat-transfer resistance owing to the fouling layer, this could have resulted equally well from a vapor-pressure depression that they did not consider. Hence, the objectives of this study were the following: to design an experiment that would permit isolating the effects of a fouling layer in MD; to determine if a fouling layer can offer any significant heator mass-transfer resistance in MD; to determine if a fouling layer can cause any vapor-pressure depression in MD; and to advance a strategy to mitigate the effects of fouling on MD.

### 2. Theoretical considerations

The water-vapor flux in MD is driven by the difference between the vapor pressure of the liquid adjacent to the membrane on the feed and distillate sides. These vapor pressures are determined by the temperatures on each side of the membrane, which as shown in Fig. 1, are controlled by the heat-transfer resistances on the feed side, distillate side, within the membrane, and by the presence of a fouling layer. The fouling layer also can introduce a hydraulic resistance to the transfer of liquid water through it and can cause a reduction in the vapor-pressure driving force owing to the small pores within it.

It is necessary to model this coupled mass- and heat-transfer process in order to design an experiment that will permit



**Fig. 1.** Schematic of the membrane-distillation process showing the temperature profile from the hot feed side across a hydrophilic fouling layer and the hydrophobic microporous membrane to the cold distillate side.

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