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# Influence of module orientation and geometry in the membrane distillation of oily seawater

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A B S T R A C T

To improve the mechanistic understanding for advancing the design and engineering of the membrane distillation (MD) modules, the objective of the current study was to investigate via both experiments and simulations the impact of (i) module orientation, (ii) module geometry, and (iii) an oily feed on the permeate flux and pore wetting propensity of direct contact membrane distillation (DCMD). Three module orientations and four feed channel geometries were investigated via experiments and simulations for oily feeds. Two key highlights emanated from this study. Firstly, module orientation mattered for DCMD, particularly in view of the formation of natural thermal convective currents and when the particle density of the particulate foulants varied. Particulate foulants with density much lesser and greater than water only deposited when the membrane was oriented respectively atop and beneath the feed. Secondly, the lack of consideration of convection currents, oil coalescence and the corresponding cake-enhanced temperature polarization in the simulations caused disagreement with the experimental results, which underscores the importance of these factors. This highlights that the optimization of MD modules particularly for treating oily feeds requires more mechanistic studies, especially in view of the thermal gradients, rather than relying on analogy with pressure-driven filtration processes.

## 1. Introduction

Membrane distillation (MD) involves distillation through a microporous hydrophobic membrane, which acts as a physical interface between the hot feed and cool permeate. It is a promising low-cost, energy-saving alternative (based on use of waste-heat) to conventional separation processes like distillation and reverse osmosis that is gaining much traction, as evidenced in more than ten reviews in the past five years [1–15]. When MD was first described in a patent by Bodell [16] in 1968, it did not immediately become popular for water treatment due in part to the lack of suitable membranes which need to be hydrophobic yet highly permeable to vapors, able to withstand the thermal operating conditions, and are cost-effective [17]. As membrane fabrication advanced, commercial hydrophobic membranes such as polypropylene, PVDF and PTFE used for microfiltration has become viable for MD [17–20], which has improved the potential of MD for treating a multitude of feeds. The key advantages of MD include high rejection of solutes, operation at lower pressures because the osmotic pressure difference does not need to be overcome, amenability to make use of waste heat, among others [3]. Despite MD being an attractive green technology, two of the primary issues that plague MD and that are areas of active research are low fluxes and pore-wetting which compromises permeate quality [21].

The treatment of oily wastewater via membrane-based filtration processes has not flourished due to the challenges associated with sustaining the flux and rejection rates [22,23]. However, in view of the large amounts of oily wastewater from the three main contributing industries of oil and gas, palm oil and mining [22–24], improved membrane separation processes could have a role in cost-effectively treating these streams to mitigate the environmental impact associated with their disposal. It is notable that, despite MD being a promising green technology, studies on treating oily feeds via MD are scarce. A recent study on produced water treatment using MD suggested that MD can only be considered for treating low concentrations of oil (500 ppm) and oils with higher proportion of hydrocarbon [25]. Furthermore, another study on shale gas produced water treatment using MD suggests that pre-treatment of oil and grease is mandatory prior to MD application to improve stability, quantity and quality of permeate [26]. Yet another study indicated that pore-wetting in MD is not due to the oil itself, but to the interactions between salt, surfactant and the membrane [27]. The focus of this study is on furthering the understanding of MD in treating such oily feeds.

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Advances in MD have focused on membrane development (e.g., membranes customized to have excellent anti-wetting properties, confer high flux, withstand high temperature, resistant to fouling and scaling, and improved thermal transfer efficiency) and processes designed to save energy through system hybridization [28]. The design and orientation of MD modules, despite being an economical alternative to changing the hydrodynamic conditions, have unfortunately garnered little attention [29]. A recent review on computational fluid dynamics (CFD) in MD showed the importance of coupling both heat and mass transfer across and near the membrane surface [30]. The hydrodynamic conditions affecting foulant deposition add even more complexities. It should be noted that, while RO modules are standardized, MD modules have yet to be optimized for better performance. One recent study by Warsinger et al. [31] examined the effect of module orientation on the efficiency of air gap MD (AGMD) in treating saline solutions without colloidal foulants. They concluded that module orientation, because of its effect on droplet flow and film thickness on the condenser surface in AGMD systems, can be optimized to improve flux by up to 40%. In an analogous work on module design for microfiltration (MF), Zamani et al. [32] evaluated a simple tapered feed channel with channel height increasing from the entrance. This design, known as flow-field mitigation of membrane fouling (FMMF), was able to mitigate fouling during microfiltration via a transverse flow trajectory induced to counter the permeate drag towards the membrane. The critical flux of particulate foulants, namely, polystyrene, was shown to be much improved via FMMF.

In view of the gap in the knowledge base regarding the impact of module orientation, module geometry and particulate foulant density on the MD performance parameters of flux and membrane pore wetting, this study sought to bridge the gap via both experiments and simulations. The three questions addressed are described as follows. Firstly, does the module orientation matter in direct contact membrane distillation (DCMD) applications? Warsinger et al. [31] observed that optimizing the module orientation improved the flux of air gap membrane distillation (AGMD) by up to 40% due to hydrostatic effects, and further hypothesized that module orientation may not benefit DCMD. Secondly, with respect to the key feature of FMMF in countering the permeate drag in MF, is FMMF also feasible and beneficial in MD particularly when the 'particulate' foulant is less dense than water (as for oily feeds)? Thirdly, does the density of the particulate foulant, particularly since oil is less dense than water, affect the DCMD performance?

## 2. Experimental setup and simulation

### 2.1. Experimental study

An experimental study was first carried out to evaluate the efficacy of the flowfield mitigation of membrane fouling (FMMF) configuration in separating oil emulsions, which were significantly more buoyant compared to the polystyrene beads used in Zamani et al. [32], via membrane distillation (MD).

#### 2.1.1. Experimental setup

The schematic diagram of the experimental direct contact membrane distillation (DCMD) setup is shown in Fig. 1. The membrane module was made of acrylic, and had thermocouples (PT100 Resistance Temperature Detectors) inserted for the control of the feed and permeate temperatures at 65 and 14 °C, respectively. The permeate flow channel was fitted with a spacer mesh (specifications listed in Table A2) to reduce the heat-transfer resistance and provide mechanical support for the membrane, but a spacer was absent on the feed side to enable a more straightforward comparison with the simulations. A new piece of PVDF hydrophobic flat-sheet microfiltration membrane (Durapore GVHP; nominal pore diameter of 0.22 μm) was used for each experimental run with an active area of 0.00371 $m^2$ (53 mm by 70 mm). Three peristaltic pumps (Masterflex L/S Digital Drive) were used to drive the feed and permeate recirculating loops, and also the recycling line. The feed was continuously recirculated at 750 mL/min between the membrane module and the feed tank (a 2-L round bottom flask), which was constantly agitated with a magnetic stirrer and heated by a hot plate (Heidolph MR Hei-Tec), via Masterflex Norprene tubing. The permeate was continuously recirculated through Masterflex Tygon E-LFL tubing at 300 mL/min between the membrane module and the permeate tank (a 1-L acrylic tank with a spout). The permeate tank had a conductivity meter (Eutech Instruments Alpha Cond 500) inserted to monitor the permeate quality, was cooled by a recirculating chiller (Julabo ME) and overflowed into the overflow permeate tank (a 300 mL beaker) that sat on a mass balance (Mettler-Toledo ME4002) for the measurement of permeate flux. The mass and conductivity data were logged on a computer via a National Instruments Data Acquisition (NI-DAQ) module every 5 min. In order to maintain a constant feed concentration, between 0 and 82 mL of the permeate (depending on the flux) from the overflow permeate tank was recycled every 1 h throughout the experiment back to the feed tank by a peristaltic pump.

#### 2.1.2. Membrane module

The four geometries of the feed channel of the DCMD membrane module investigated, which were similar for both the experiments and simulations, are depicted in Fig. 2, while the permeate channels were constant at a uniform depth of 1 mm. Fig. 3 depicts the detailed dimensions of the channel, whose inlet was specially designed to minimize entrance effects, the influence of which was accounted for in the CFD simulations. The four feed channel geometries had different cross-sections in the *x-z* planes, namely, (a) a uniform channel depth of 1 mm; (b) a uniform channel depth of 2 mm; (c) the FMMF configuration, which has earlier been shown to more energy-efficiently mitigate fouling by polystyrene beads during microfiltration [32], with an inclination angle of 1.5° such that the inlet and outlet depths were 2 mm and 3.3 mm, respectively; and (d) the FMMF configuration with an inclination angle of 3° such that the inlet and outlet depths were 2 mm and 4.6 mm, respectively. While the uniform feed channel depths of 1 mm and 2 mm provided insights on varying channel depth and cross-flow velocity on the interactions between the membrane and oil droplets, the diverging channels shed light on the efficacy of flowfield alteration on mitigating membrane fouling and wetting. Each of these four geometries was investigated via both experiments and simulations for three different module orientations, namely, horizontal with feed at the bottom (i.e., membrane atop the feed), horizontal with feed on top (i.e., membrane beneath the feed), and vertical, as shown in Fig. 4.

#### 2.1.3. Materials

The initial feed solution was 35 g/L of sodium chloride (NaCl; Merck-Millipore CAS No. 7647-14-5) dissolved in DI water for the first 3 h, after which the oil-in-water emulsion was added. The oil used in this study was hexadecane (Sigma-Aldrich CAS No. 544-76-3) which was dyed with oil red (Sigma-Aldrich CAS No. 1320-06-5) at a concentration of 0.1 mg/mL to improve visual observation of oil coalescence and accumulation within the membrane module. Hexadecane was used due to its widespread use for studying membrane-filtration with feeds containing oil emulsions to mimic oily wastewater, as well as its representation as a foulant with density significantly lower than water. The oil emulsion was prepared by sonicating 1.3 mL of the dyed hexadecane in 48.7 mL of the feed solution (i.e., 35 g/L NaCl) using a Branson S-450D sonifier with 3/4 inch (19 mm) high gain horn at 70% amplitude for 2 min, resulting in a 20,000 ppm stock solution of oil emulsion with an average droplet size of 10 μm (measured by the Focused Beam Reflectance Measurement system (FBRM) Lasentec S400, PI-14/206).

#### 2.1.4. Experimental protocol

The following protocol was used for each experiment. Firstly, the membrane sheet was cut into dimensions of 70 mm by 100 mm and