



## Organic fouling behavior in direct contact membrane distillation



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### HIGHLIGHTS

- Organic fouling development in DCMD was investigated with model organic compound.
- HA compound thermally disaggregated to LMW-HA organics.
- Membrane pore penetration was observed with the presence of LMW-HA organics.
- Fouling to fouling interaction was evident in mixed organic feed solution.

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### ABSTRACT

Organic fouling development in a direct contact membrane distillation system was investigated with synthetic model solutions of humic acid (HA), alginate acid (AA) and bovine serum albumin (BSA). A 40–50% flux decline was observed, with BSA and HA showing dominant fouling tendencies, while minimal fouling was observed with AA due to its hydrophilic nature. Based on the feed characterization analysis using liquid chromatography-organic carbon detector, the HA feed solution showed thermal disaggregation to low molecular weight-HA (LMW-HA) organics with increased feed temperature (50 °C and 70 °C). The HA fouled membrane autopsy analysis showed the penetration of organics through the membrane pores. In comparison, the BSA feed solution showed more significant deposits on the membrane surface (35.2% higher carbon mass compared to HA foulant) with less significant pore penetration. The mixed organic (MO) feed solution consisting of BSA, HA and AA altogether showed evidence of fouling to fouling interaction. The MO feed solution leads to a significant flux decline of 65% only at a later stage upon the disaggregation of LMW-HA organics that adhered on the membrane.

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### 1. Introduction

Membrane processes are widely used in the application of seawater desalination, wastewater reuse, and water treatment. Membrane fouling in pressure-driven membrane processes is inevitable, requiring the usage of chemical cleaning agents and pre-treatments which increases the cost of water production.

Membrane distillation (MD) is an alternative technology that integrates both thermal distillation and membrane processes using trans-membrane vapor pressure difference across a hydrophobic membrane [1]. The application of MD process has several advantages in comparison to pressure-driven membrane processes [1,2]. In MD, partial pressure would only be slightly reduced due to high salt concentration, as it is not subjected to osmotic pressure gradient [2]. As such, MD operation has the potential to operate up to high saturated levels of the feed solution.

A critical issue that still demands considerable research effort in MD is membrane fouling. This is specifically because, the feasibility of operating MD at saturated feed solution levels would be strongly dependent on the intensity of membrane fouling development. A number of factors attribute to membrane fouling such as natural organic matter (NOM), inorganic matters, particulates and colloidal matters. It has been acknowledged that NOM is one of the primary causes of flux decline in membrane organic fouling [3,4].

NOM includes a group of low molecular weight (LMW) to high molecular weight (HMW) organic compounds, quantified as a dissolved organic carbon (DOC). In surface water, ground water and seawater, humic substances are the major constituents of NOM, followed by carbohydrates (including polysaccharides), protein and a variety of acidic and LMW species [5,6].

Factors that influence membrane fouling have been analyzed extensively, namely the characteristics of the feed water, i.e. composition of NOM compound and the physiochemical condition of the feed water, MD operating parameters as well as the membrane characteristics. In terms of the influence of NOM compounds, it has been observed that fouling by non-humic alginate compound (polysaccharides) compared

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to humics is severe in high pressure membrane processes such as reverse osmosis (RO) and ultrafiltration (UF) process [3,4,7,8]. This has been associated with the compressible and deformable nature of alginate under pressure, causing initial pore blocking and gel layer formation. On the other hand, in low pressure processes such as forward osmosis (FO), it was observed that more severe permeate flux decline occurred with humic and alginate due to the higher adhesion capacity of these compounds on the membrane. Meanwhile, a much lower flux decline rate was observed in FO with bovine serum albumin (protein). This was primarily associated with structure deformation of the protein compound due to reverse salt intrusion from draw solution [9].

As a whole, membrane studies have highlighted that NOM rejection is controlled by size exclusion, electrostatic repulsion, physicochemical feed conditions as well as the interactions between the organic foulant and the membrane. MD fouling studies have not explored in detail these interactions. This is particularly important as the hydrophobic membrane has been deemed to be more susceptible to fouling adsorption than hydrophilic membrane [10–12]. It has actually been demonstrated that humic compounds adsorbed more favorably onto hydrophobic membranes [13]. Further, the thermal heating in MD may influence humic characteristics, whereby previous studies have observed humic disaggregation to lower molecular size humics and other organics under thermal condition [14,15]. However, MD studies have highlighted that fouling by humic substances was not severe as compared to pressure-driven membrane processes, primarily due to the vapor pressure operated application [10,16,17]. Instead, MD studies have indicated that feed water containing NOM compounds of proteins, amino sugars and polysaccharides showed strong tendency to deposit on the hydrophobic membrane [18,19]. The application of high feed temperature (60 °C and above) in MD operation intensified protein based NOM [18,20]. Previous studies have observed structural and surface charge changes in protein organics with increased temperature [21].

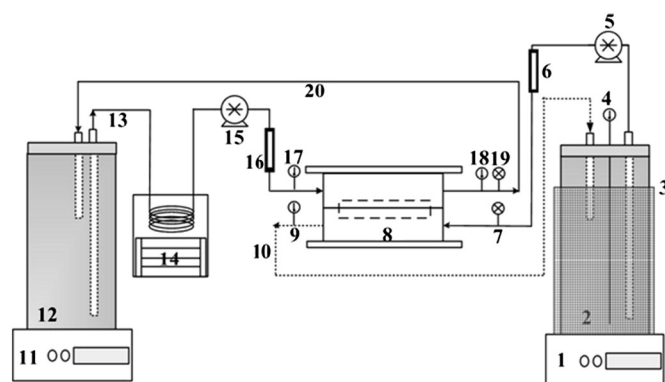
In most MD fouling investigations, membrane fouling is represented by the permeate flux decline [10,16,18]. Although membrane fouling is generally interpreted by flux decline, this approach is inadequate for characterizing fouling development in MD, especially due to the effect of temperature in the operation. Characterizing the foulant on the MD membrane would provide valuable guidance to the effective application of MD operation such as membrane cleaning as well as deciding the necessity for a pretreatment. A few recent studies have reported specific methods to represent more detailed organic foulant characteristics using Liquid Chromatography–Organic Carbon Detection (LC–OCD) [4,22]. The effectiveness of LC–OCD to characterize fouling material in membrane processes and to identify specific organic matters responsible for fouling in water and wastewater treatment has been demonstrated in a number of studies [23–25].

Hence, the objective of this study was to determine the dominant organic compound in MD operation based on the detailed analysis of the organic characteristics. For this purpose, a direct contact membrane distillation (DCMD) system was used with model solutions of humic acid (HA), alginic acid (AA) and bovine serum albumin (BSA). These represented the humic substances, polysaccharides and proteins respectively, which are the major organic components of natural water sources such as surface water, groundwater and seawater [5]. The fractions of organic compound in the feed and permeate solution as well as the membrane foulant was analyzed using LC–OCD. The morphology and composition of the deposit layer formed on the membrane surface were analyzed using scanning electron microscopy (SEM) while contact angle was used to determine the hydrophobicity of the membrane.

## 2. Materials and methods

### 2.1. DCMD set-up

A bench-scale DCMD system was used in this study as shown in Fig. 1. Details of the experimental set-up are available in another



**Fig. 1.** DCMD experimental set-up. (1) Feed electronic balance, (2) feed tank, (3) heating blanket, (4) feed tank temperature sensor, (5) feed pump, (6) feed flow meter gauge, (7) feed inlet pressure gauge, (8) membrane module, (9) feed outlet (brine) temperature sensor, (10) brine, (11) permeate electronic balance, (12) permeate tank, (13) cooling water, (14) cooling unit, (15) cooling pump, (16) cooling flow meter gauge, (17) cooling water inlet temperature sensor, (18) cooling water outlet temperature sensor, (19) cooling water outlet pressure gauge, and (20) permeate.

previous study [26]. Feed solution was heated at the set temperature in a sealed feed tank encased in an electric heating blanket. The temperature of the permeate side was regulated by a cooling unit. Initially, 2 L of feed solution and 2 L of cooling water (deionized (DI) water) were circulated into the membrane module by a peristaltic pump until a stable temperature is reached. The temperatures of the feed solution and cooling water were measured at the inlet and outlet of the membrane module with temperature sensors. The hydraulic pressures on the feed inlet and permeate side outlet were measured with pressure gauges. The feed and permeate tanks were placed on electronic balances to monitor the permeate production and feed reduction over time. Each experiment was carried out until the feed volume was reduced to 0.4 L. Accordingly the experimental duration ranged from 3.5 to 7.0 h based on the time required to achieve this feed volume reduction.

### 2.2. Membrane

A hydrophobic polytetrafluoroethylene (PTFE) flat sheet hydrophobic membrane (General Electric, US) was used in this study. The membrane support layer was made of polypropylene (PP). The effective membrane area was 0.0168 m<sup>2</sup> (0.21 m × 0.08 m). The membrane channel dimensions were 21.0 cm (length), 8.0 cm (width), and 0.4 cm (height). The porosity, average pore size and thickness of membrane provided by the supplier were 70–80%, 0.2 μm, and 179 μm, respectively.

### 2.3. Feed solution

Experiments were conducted with model organic compounds of alginate (AA) (A7003, CAS No. 9005-32-7, Sigma-Aldrich, St. Louis, MO), humic acid (HA) (53680, CAS No. 1415-93-6, Sigma-Aldrich, St. Louis, MO), and bovine serum albumin (BSA) (A2135, CAS No. 9048-46-8, Sigma-Aldrich, St. Louis, MO) to represent polysaccharides, humics, and proteins, respectively.

According to the manufacturer, the BSA MW was approximately 66 kDa. The sodium alginate extracted from brown seaweed had a MW ranging from 12 to 80 kDa [8]. Meanwhile the HA had a MW ranging from 4 to 20 kDa [27]. The organic compounds were received in powder form. The stock solutions were prepared by dissolving 350 mg C L<sup>-1</sup> organics in deionized (DI) water followed by filtration with 0.45 μm filter to exclude initial deposition of bulk suspended solids on the membrane. The initial concentration of the organic compounds (10 mg C L<sup>-1</sup>) was determined in terms of dissolved organic carbon concentration. For the mixed organic (MO) solutions, the three model

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