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Simplified flux prediction in direct-contact membrane distillation using a membrane structural parameter

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HIGHLIGHTS

- Membrane structural parameters were evaluated for use in predicting water flux.
- Membrane constant containing non-coupled membrane properties was introduced.
- Membrane constant requires simple measurements and inexpensive analytical equipment.
- Membrane constant correlates well with experimental water flux.
- Comprehensive collection of MD membrane properties and water fluxes made available.

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ABSTRACT

A priori water flux prediction is desirable when conducting membrane distillation (MD) studies, however existing models are complicated with inconsistent mass transfer mechanism assumptions. To develop a simplified model that can be used to predict the relative magnitudes of water fluxes for a group of MD membranes, correlation analyses were performed between water flux and 28 structural parameters. Four parameters were found to be highly correlated with water flux: ε/δ, $\varepsilon/\tau\delta$, $1/\tau\delta$, and C_m. C_m is a newly introduced structural parameter that contains non-coupled membrane properties but still carries the physical meaning of a relationship between δ (thickness) and ε (porosity) and is determined by simple and reliable measurements using inexpensive analytical equipment. The correlation result between water flux and C_m suggests that C_m is a good structural parameter for MD flux prediction. The flux prediction errors for membranes with pore sizes from 0.1 to 0.9 μm were generally smaller for the model developed with C_m than for the dusty gas model. In addition to the new structural parameter and model, this study also makes available to the literature a detailed collection of MD membrane properties and their water flux values that will assist others in membrane selection, development, and application.

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1. Introduction

1.1. Membrane distillation

Membrane distillation (MD) is a thermally-driven process in which separation occurs through a phase change to produce clean water. The driving force in MD is the vapor pressure gradient, resulting from the temperature difference across the membrane. Among all types of MD, direct-contact MD (DCMD) is the most commonly used configuration in lab-scale research [1]. In DCMD, two solutions at different bulk temperatures are circulated on either side of a hydrophobic microporous membrane. Temperatures of the feed solution can range from 30 to 90 °C [2,3], which makes it feasible to be combined with low-grade heat sources. DCMD has been used to treat feedwaters with high fouling and scaling potentials, such as industrial wastewater [4,5], water from salt lakes [6], RO brines [7,8], and produced water from the oil and gas industry [9–11], because the driving force of DCMD does not decrease significantly with increasing water salinity. DCMD is also well suited to treat feedwaters with low fouling and scaling potentials where targeted removal or polishing is desired because DCMD achieves near 100% salt and organic rejection [12,13]. This includes treatment of impaired water containing endocrine disrupting compounds [14]; brackish water contaminated with fluoride [15]; groundwater with heavy metals [16]; and feedwaters with urine and hygiene wastewater [17]. In some DCMD applications (particularly with low fouling and scaling feedwaters) obtaining high water flux is desirable while in other applications (with high fouling and scaling feedwaters) it is not; thus, *a priori* water flux prediction is desirable for membrane selection. Because MD water flux is affected by membrane properties, feedwater properties,

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and operating conditions [18], if a group of MD membranes is operated using the same feedwater at specific operating conditions, only the membrane properties will affect the relative magnitude of water flux.

1.2. Existing mass transfer models for flux prediction in MD

Water flux (N_i) through an MD membrane is given as:

$$N_i = B\Delta P_i \tag{1}$$

where B is the membrane mass transfer coefficient and ΔP_i is the water vapor pressure gradient across the membrane. Here, subscript i is used to represent water vapor and subscript j will be used to represent air. Water vapor pressure (P_i) for both the feed stream and the distillate stream is expressed using the Antoine equation [19,20]:

$$P_i = \exp\left(23.328 - \frac{3841}{T-45}\right) \tag{2}$$

where T is the temperature of the respective stream. The dusty gas model is often used to estimate water flux in MD, where four mass transfer mechanisms (surface diffusion, Knudsen diffusion, molecular diffusion, and viscous flow) may occur; the thermal circuit representation is given in Fig. 1. The complete expression of the dusty gas model is complex, thus surface diffusion, which only occurs when membrane pore sizes are smaller than 0.02 μm [21], is typically not included so as to simplify MD flux prediction [2,22]. MD water flux without consideration of surface diffusion is given as:

$$N_i = N_i^D + N_i^V \tag{3}$$

where N_i^D and N_i^V are the diffusive (combined Knudsen and molecular) flux and viscous flux of water vapor, respectively. In its most general form, the dusty gas model applicable to MD is given by two equations [2]:

$$\frac{N_i^D}{\frac{2r\varepsilon}{3r\delta}\left(\frac{8RT_m}{\pi M_i}\right)0.5} + \sum_{j=1\neq i}^{n} \frac{P_j N_i^D - P_i N_j^D}{\frac{\varepsilon}{\tau\delta} PD_{ij}} = \frac{1}{RT_m}\Delta P_i \tag{4}$$

$$N_i^V = \frac{P_i}{8RT_m\mu}\frac{r^2\varepsilon}{\tau\delta}\Delta P \tag{5}$$

where r, ε, τ, and δ are the pore radius, porosity, tortuosity, and thickness of the membrane, respectively; R is the universal gas constant; T_m is the average temperature of the membrane; M_i is the molecular weight of water vapor; P_j is the air pressure inside the membrane pores; N_j^D is the diffusive flux of air; P is the total pressure; D_{ij} is the ordinary diffusion coefficient; μ is the fluid viscosity; and ΔP is the transmembrane pressure. Two equations for PD_{ij} are given in the literature [2,12,23]:

$$PD_{ij}\left(\mathrm{kPam^2/s}\right) = 4.46\times10^{-9}\times T_m^{2.334} \tag{6}$$

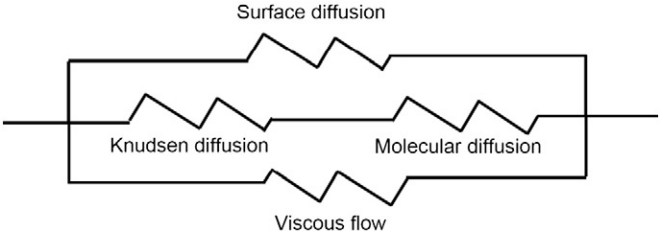

Fig. 1. Thermal circuit of the dusty gas model [2].

and [22,24]:

$$PD_{ij}\left(\mathrm{kPam^2/s}\right) = 1.895\times10^{-8}\times T_m^{2.072}. \tag{7}$$

Because of temperature polarization, the temperatures at the membrane surfaces (feed and distillate sides) are different from the bulk temperatures, thus the dynamic conditions inside the membrane module have to be considered (with hydraulic pressures, salinities, heat capacities, viscosities, and flow rates on both the feed and distillate sides, spacer properties if spacers are used, and membrane module dimensions) when determining the average membrane temperature (T_m) [19,25–27]. Also, because hydraulic pressures always exist in flowing streams, membrane compaction may occur during MD testing, resulting in modified membrane properties (r, ε, τ, and δ) [28,29]. Both temperature polarization and membrane compaction complicate the mass transfer equations. In seeking simplification of flux prediction, some investigations have assumed that viscous flow is negligible in DCMD due to the lack of a hydraulic pressure gradient [12,22]; in these cases, only Eq. (4) is used to predict water flux. Other investigations assert that viscous flow cannot be neglected, especially for membranes with large (e.g., >0.3 μm) pore sizes where the magnitude of the mean free path of water vapor in air is much smaller than the membrane pore size [19,30]. In these cases, a membrane pore size distribution instead of the average pore size has been used for mass transfer modeling [22,31,32]. The complicated model expressions and contradictory assumptions from the literature for the mass transfer mechanisms make prediction of water flux using the simplified dusty gas model cumbersome and ambiguous.

1.3. Existing membrane property parameters

If experimental operating conditions and solution chemistries are kept constant, then only the membrane properties will affect water flux. Considering this, further simplifications of the dusty gas model in the literature have used membrane property parameters (also referred to as membrane morphology parameters) to qualitatively analyze water flux. From Eqs. (4) and (5), membrane property parameters affecting water flux are $\varepsilon/\tau\delta$, $r\varepsilon/\tau\delta$ or $r^2\varepsilon/\tau\delta$ for molecular diffusion, Knudsen diffusion, and viscous flow, respectively. It is expected that membranes with greater $\varepsilon/\tau\delta$, $r\varepsilon/\tau\delta$ or $r^2\varepsilon/\tau\delta$ will have higher water fluxes [2,24, 33–36]. It is also generally agreed that higher water fluxes occur for MD membranes with higher porosity [33,37,38] or lower tortuosity [23,39].

It is unclear to what extent membrane pore size affects water flux since the role of membrane pore size is not the same in $\varepsilon/\tau\delta$, $r\varepsilon/\tau\delta$ and $r^2\varepsilon/\tau\delta$. Lawson et al. [37] found that water flux increased with increasing pore size. Mericq et al. [40] found that the Knudsen permeability of the membrane ($B \propto r\varepsilon/\tau\delta$; r included) strongly affected water flux. However, in a couple of observations, water flux was found to be highly sensitive to the characteristic parameter $\varepsilon/\tau\delta$ [27,34] and only slightly sensitive to pore size [27]. Ali et al. [36] also observed no dramatic increase of the water flux with increasing pore size, especially when the pore size was smaller than 0.3 μm.

Although thickness is generally included in the membrane property parameters, some studies discounted its role and utilized ε/τ, $r\varepsilon/\tau$, and $r^2\varepsilon/\tau$. Lawson et al. [37] found that flux increased as the membrane parameter $\tau\delta$ increased. Bonyadi and Chung [33] and El-Bourawi et al. [38] found that thickness was important because thinner membranes have reduced mass transfer resistance but they also found that flux did not monotonically increase with thickness reduction because of increased conductive heat loss through the membrane.

Although several membrane property parameters have been analyzed in the literature, there are contradictory observations about their effects on water flux (with the exception of porosity and tortuosity). Furthermore, only qualitative analyses between water flux and membrane property parameters were given; these enable the evaluation of

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