



## Transport phenomena and fouling in vacuum enhanced direct contact membrane distillation: Experimental and modelling



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### ABSTRACT

The application of vacuum to direct contact membrane distillation (vacuum enhanced direct contact membrane distillation, V-DCMD) removed condensable gasses and reduced partial pressure in the membrane pores, achieving 37.6% higher flux than DCMD at the same feed temperature. Transfer mechanism and temperature distribution profile in V-DCMD were studied. The empirical flux decline (EFD) model represented fouling profiles of V-DCMD. In a continuous V-DCMD operation with moderate temperature (55 °C) and permeate pressure (300 mbar) for treating wastewater ROC, a flux of  $16.0 \pm 0.3$  L/m<sup>2</sup> h and high quality distillate were achieved with water flushing, showing the suitability of V-DCMD for ROC treatment.

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### 1. Introduction

Membrane distillation (MD) is a thermal membrane process based on the principle of vapor–liquid equilibrium for molecular separation, in which only vapor molecules are transported through the pores of a hydrophobic membrane and needs a supply of latent heat of vaporization for the phase change from liquid to vapor [1–4]. The driving force for an MD process is transmembrane vapor pressure difference, primarily established through temperature gradient between the liquid–vapor interfaces. MD possesses the potential to concentrate solutions to their saturation point with minimal flux decline, compared to convention pressure driven membrane processes [1,2]. On the basis of allowing only vapor through the membrane, theoretically MD operation can achieve 99% rejection of all non-volatile contaminants and ions to produce high quality distillate. At the same time, the low thermal requirement for MD process can be met by alternative energy sources such as industrial waste heat, solar energy and geothermal energy [2,5]. In view of these promising benefits, MD has been regarded as a viable alternative concentrate treatment technology [3,4].

Presently, many wastewater reclamation plants (WRPs) around the world are progressively using reverse osmosis (RO) technology

as a final polishing step due to its capability to maintain a high grade water standard [1,6]. However, a substantially large volume of wastewater RO concentrate (ROC) is produced, generally comprising 20–25% of the feed stream volume [7]. For instance, two WRPs for biologically treated wastewater in New South Wales (Homebush bay and St. Marys WRPs) are using RO technology as a final treatment process. On a daily basis, around 2000 kL of water is treated by RO in Homebush bay WRP, resulting in 300 kL/day ROC while substantially larger volume of ROC (7000 kL/day) is produced from St. Marys WRP [8].

It has been well established that conventional methods such as coagulation and granular activated carbon as well as advanced technologies such as, ozonation, electrochemical oxidation and photocatalysis are effective for treating ROC contaminants, specifically to reduce dissolved organics carbons as well as selective micro-pollutants prior to discharge from WRPs [1,6,7]. Nonetheless, these operations are unable to reduce the ROC volume and its inorganic contents.

In this regard, MD offers the possibility to concentrate pre-treated ROC wastewater, while producing good quality distillate, making it a sustainable ROC treatment technology. Furthermore, the low salinity content of ROC wastewater (1–3 g/L), would enable MD to concentrate ROC to a high level. However, the aspect of scaling by the main ions present in ROC wastewater such as Ca, Na, SO<sub>4</sub>, and Cl must be given due consideration to

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## Nomenclature

J	permeate flux (kg/m <sup>2</sup> ·h)	t	time (s)
C	mass transfer coefficient	r	membrane pore diameter (m)
P	vapor pressure (Pa)	η	water vapor viscosity (kg/m/s)
T	temperature (°C)	ε	membrane porosity
D	diffusion coefficient (m <sup>2</sup> /s)	δ	membrane thickness (m)
D <sub>WA</sub>	molecular diffusivity coefficient (m <sup>2</sup> /s)	τ	membrane pore tortuosity
M	molecular mass of water (kg/kmol)	k <sub>0</sub>	flux decline potential
R	universal gas constant (kJ/kmol/K)	k <sub>1</sub>	rate constant
P <sub>total</sub>	total pressure inside the pore (Pa)	d	flux decline kinetic constant
P <sub>α</sub>	air pressure (Pa)	γ <sub>w</sub>	activity coefficient of water
P <sub>avg</sub>	average pressure of the membrane (Pa)	x <sub>w</sub>	mole fraction of water
P <sub>w</sub>	vapor pressure of pure water (Pa)		
P <sub>reference</sub>	reference pressure (Pa)	<i>Subscript</i>	
α	dimensionless coefficient ratio	f	feed side
Q	heat flux (kJ/m <sup>2</sup> ·h)	p	permeate side
h	heat transfer coefficient (W/m <sup>2</sup> ·K)	u	flow velocity (m/s)
A	thermal conductivity (kW/m·K)	E	molecular diffusion
L	membrane module length (m)	P	poiseuille flow diffusion
ρ	fluid density (kg/m <sup>3</sup> )	K	Knudsen diffusion
C <sub>p</sub>	specific heat capacity of fluid	m	membrane surface
λ	latent heat of water	x	transversal (x-direction)
R <sub>m</sub>	membrane resistance (Pa m <sup>2</sup> h/L)	z	axial (z-direction)

establish the feasibility of MD operation for ROC wastewater treatment.

Direct contact membrane distillation (DCMD) is the most commonly used MD configuration due to its application simplicity [9]. In general, high feed temperature is used to achieve higher water production (water flux) in DCMD system [10]. At a high feed temperature, higher heat and mass transfer occur from the feed side, across the boundary layer and membrane, and to the permeate side. Large quantity of heat is used to vaporize the molecules at the membrane surface. It results in a significant conduction and latent heat-loss resulting in large temperature difference between the bulk solution and membrane surface. This phenomenon is known as temperature polarization. Alternatively, a larger vapor driving pressure in DCMD can be created by incorporating a vacuum on the permeate side, even at relatively low feed temperature ranges. This is referred to as vacuum enhanced DCMD configuration (V-DCMD). The advantages of V-DCMD have been highlighted in previous literatures [11,12]. Cath et al. [11] demonstrated that the V-DCMD system achieved a 15% permeate flux increment (compared to DCMD) with the reduction of permeate side pressure (increased vacuum) from 108 kPa to 94 kPa. Importantly, their study highlighted that on an economic aspect, the incorporation of vacuum did not incur significant additional cost due to the low pressure-gradient on the pump. Similarly, Naidu et al. [12] established the enhanced permeate flux performance of DCMD by 30% with reduced permeate pressure from 1000 mbar to 300 mbar using vacuum pump. Conversely, that study reported a higher flux decline and fouling with V-DCMD compared to DCMD [12].

Overall, there are still limited researches on an in-depth understanding on the performance of a V-DCMD in comparison to DCMD. Generally, the mass transfer across the membrane for DCMD system consist of Knudsen diffusion, molecular diffusion, surface diffusion, and viscous flow [13]. In this present study, experimental data of V-DCMD and DCMD performance were used to estimate the transport mechanism and related resistance based on the empirical models and resistance coefficient values. An alpha coefficient was incorporated to represent the mass transfer scenario of V-DCMD. At the same time, a 2-D dynamic model

temperature profile along the membrane module was used to compare the temperature profile of the DCMD and V-DCMD configuration. Further, the fouling profile was represented by coefficient values based on empirical fouling model. A continuous experimental operation of V-DCMD with membrane water flushing was carried out to verify the effectiveness of this configuration for ROC wastewater treatment. Membrane fouling pattern and its reversibility on membranes were analysed using scanning electron microscope - energy-dispersive spectroscopy (SEM-EDX).

## 2. Materials and methods

### 2.1. Experimental set-up

Experiments for DCMD and V-DCMD were carried out with the same effective membrane area of 0.0168 m<sup>2</sup> (0.21 m × 0.08 m). The dimensions of the membrane cell channel were 21.0 cm (length), 8.0 cm (width), and 0.4 cm (height). The membrane cell was designed to hold a flat sheet membrane securely in the membrane cell under moderate pressure gradients without the need for physical supports such as spacers (Fig. 1). A polytetrafluoroethylene (PTFE) flat-sheet hydrophobic membrane (General Electrics, US) was used in this study. The porosity, normalized pore size, and membrane thickness as provided by the supplier were 70–80%, 0.22 μm, and 179 μm, respectively. The DCMD baseline study (with deionized (DI) water) was carried out at different feed temperatures ranging from 50 to 65 °C. Meanwhile the V-DCMD operation was carried out at different vacuum pressures (300–1000 mbar) at a feed temperature of 55 °C. For all operating conditions, flow rate in both feed and permeate sides was 1.1 L/min, which corresponds to a cross-flow velocity of 0.06 m/s.

### 2.2. Feed solution

The baseline experiments were carried out with 1.5 L of DI water. To study the performance of DCMD and V-DCMD for the treatment of ROC wastewater, a synthetic solution (1.5 L) was used. The synthetic solution comprised of 600 mg Na/L,

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