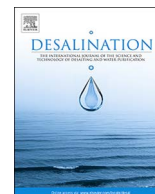




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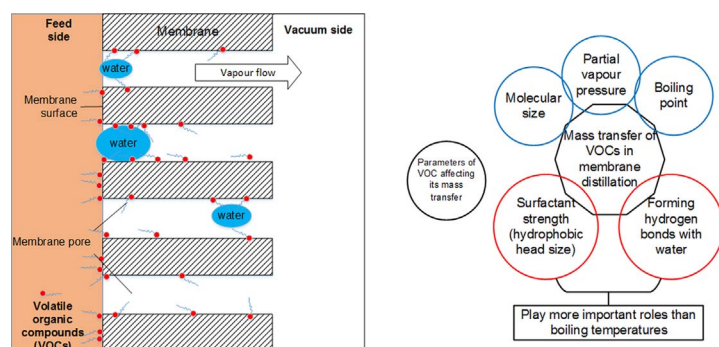
Effects of volatile organic compounds on water recovery from produced water via vacuum membrane distillation

Minwei Yao^a, Yun Chul Woo^a, Leonard D. Tijning^a, June-Seok Choi^b, Ho Kyong Shon^{a,*}

^a Centre for Technology in Water and Wastewater, School of Civil and Environmental Engineering, University of Technology Sydney (UTS), P. O. Box 123, 15 Broadway, NSW 2007, Australia

^b Environment and Plant Research Institute, Korea Institute of Civil Engineering and Building Technology (KICT), 283, Goyangdae-Ro, Ilsanseo-Gu, Goyang-Si, Gyeonggi-Do 411-712, Republic of Korea

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:

Membrane distillation
Gas produced water
Volatile organic compound
Wetting
Surfactant

ABSTRACT

Membrane distillation (MD) has great potentials to treat produced water in energy industries. However, volatile organic compounds (VOCs) existing in the produced water added in the fracking process can hinder the treatment process regarding two aspects: permeate quality and MD flux performance. To address this challenge, this study aims to systematically study the effects of the VOCs on the MD permeation performance and permeate quality, and the mechanism of its penetration. Acetic acid, ethylene glycol, isopropyl alcohol (IPA), and 2-Butoxyethanol (2-BE), which are commonly found in the produced water, were extensively investigated and their impacts were reviewed and compared. Among all the VOCs, 2-BE had the highest mass transfer despite its low vapour pressure and large molecule weight. Some of the VOCs had surfactant properties, which meant they could penetrate the membrane pores easily during MD process. In long-term operation, pore wetting started to appear as the salt rejection was dropping in the MD process, and flux was also decreasing. Based on the results, this study suggested that the strength of surfactant properties and intra-molecular hydrogen bonds between water molecules and VOCs are as significant as vapour pressure for the VOCs in terms of mass transfer efficiency in MD system.

* Corresponding author.

E-mail address: hokyong.shon-1@uts.edu.au (H.K. Shon).

<https://doi.org/10.1016/j.desal.2017.11.012>

Received 31 May 2017; Received in revised form 2 October 2017; Accepted 6 November 2017
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Table 1
Properties of VOCs and concentrations of ion components of the synthetic produced water used in this study.

Volatile organic compounds	Molecular weight [g mol ⁻¹]	Surface tension [mN m ⁻¹] at 20 °C	Log P	Boiling temperature [°C]	Vapour pressure at 20 °C [kPa]	Source/usage
Acetic acid	60.1	27.3	-0.17	118.1	1.6	Natural occurring
Isopropyl alcohol	60.1	21.4	0.05	82.6	4.1	Added as corrosion inhibitor in fracking process
2-Butoxyethanol	118.2	26.6	0.83	171	0.1	Added as surfactant and active solvent in fracking process
Ethylene glycol	62.1	48.4	-1.36	197.3	0.06	Added as friction reducer in fracking process
Water (for reference)	18.0	72.8	-	100	2.3	-

Ion components of RO brine from CSG produced water							
Ions	Na ⁺	HCO ₃ ⁻	Cl ⁻	Mg ²⁺	Si (SiO ₂)	CO ₃ ²⁻	SO ₄ ²⁻
Concentration (mg L ⁻¹)	6840	4740	7770	17	75	30	20

1. Introduction

Coal seam gas (CSG, coal bed methane) unconventional natural gas resources stored in coal seam layers at a depth of 300–1000 m while shale gas is usually buried beyond 1500 m [1]. Fast replacing coals and conventional natural gas, CSG has been developed into one of most important energy resources for Australian economies (especially for Queensland), which is being greatly exported as liquefied natural gas (LNG) to Asian countries. Similarly, the unconventional gas has become an important energy source in the United States, Canada, and some other countries as well. Although these gas is considered as a relatively “greener” resource as it produced much less carbon dioxide after burning, the processes of gas exploration still pose great risks on the local ecological system. One of the biggest challenges is produced water management [1,2]. To mine the gas out of the coal bed, large amounts of water is required to be removed from the coal to reduce the hydrostatic pressure. Formation water, flow back water, and water condensing from the gas phase compose produced water. Formation water is the major contributor to the produced water, naturally occurring and being stored in oil and gas reservoirs. The water is often of brackish to saline quality [1,3].

Management of the produced water is a major challenge in energy industry due to its large amounts, complex chemical compositions, and limited disposal options. In Australia, direct reinjection of the produced water back into the deep underground well is banned, a technique commonly practiced in the United States [4]. Therefore, currently most Australian exploration companies applied reverse osmosis (RO) on site to treat the produced water. The permeate water of RO is sold to local residents for irrigation [4,5]. However, RO is not efficient to treat concentrated salty water, so still large amounts of brine could only be stored in the onsite evaporation ponds as short or medium term solution.

Membrane distillation (MD) has strong potentials to further treat the brine or produced water as salt concentration in the feed had minimal effects on its flux performance. Around 95% recovery of RO brine from the gas produced water had been achieved through the RO-MD hybrid system [6–8]. Regarding salt removal, the quality of the permeate was decent as MD has theoretically 100% rejection rate for non-volatile solutes [9]. Also, the flux performance is less affected by the salinity of the feed. In terms of energy consumption, MD was found to be more efficient than the business-as-usual strategy (BAU) for treatment of the produced water. Tavakkoli et al. found that the operation cost of MD was roughly half of the BAU strategies when low-grade heat was available [10]. However, MD had degraded permeate quality when treating wastewater containing VOCs, dissolved gas, or some small organic particles [11]. Especially, when the VOCs are alcohol or having surfactant properties, membrane wetting will occur and

lead to further deterioration of the permeate quality as the feed water can pass through the membrane directly via the wetted pores [11–13]. Recently, some researchers studied the effect of the surfactant and oil existed in the produced water on the water recovery via MD and found that they had strong negative impact on the permeation flux and permeate quality [12,14]. However, none of researches have investigated the effect of VOCs in the produced water on the MD performance, which may share similar effects as the surfactant and oil.

It has been found that the compositions of the gas produced water are highly dependent on the geology [15]. In some mining wells, high concentrations of the volatile organic compounds (VOCs), mainly acetic acid, had been found in the produced water [16]; also, the produced water could contain large quantities of VOCs which had been artificially added into the pumping fluids for fracking, especially during initial stage of the mining [15]. A concentration of 1600 mg/L acetic acid had been detected in the flow-back water in some wells [17]. The concentration of the VOC in the RO brine can be further increased. Therefore, 2000 ppm was selected as the baseline concentration for all the VOCs tested in this study to explore their maximum potential impacts. Those VOCs can hinder the MD process regarding permeate quality and water permeation performance [18]. To address this challenge, this study aims to systematically study the effects of commonly found VOCs in the produced water and their mechanism of the membrane penetration. Here, acetic acid, ethylene glycol, isopropyl alcohol (IPA), and 2-Butoxyethanol (2-BE), which are commonly used in the fracking fluids or naturally occurring [15], had been extensively investigated regarding their roles in MD. A fundamental understanding of the effects of VOCs on water recovery from gas produced water via MD can help new membrane and system design, which can further promote the realization of its potential in the application.

2. Material and methods

2.1. Materials

Acetic acid was purchased from Chem-supply, Australia. IPA was bought from Merck group, Germany while 2-BE and ethylene glycol were obtained from Sigma-Aldrich, USA. All the VOCs were used as received and their properties are displayed in Table 1. To explore the maximum potential impacts by high concentrations of VOCs in the produced water, 2000 ppm of the tested VOC was added into de-ionized (DI) water and the solution was stirred for 15 min before MD experiment, individually. A second set of experiments using 50 ppm for each VOC was conducted to explore their effects on permeate quality in more general scenarios.

Various chemicals were purchased for the preparation of synthetic CSG RO brine water. Sodium chloride (NaCl) was purchased from Ajax

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