Journal of Membrane Science 453 (2014) 636–642

Contents lists available at ScienceDirect

Journal of Membrane Science

journal homepage: www.elsevier.com/locate/memsci

Rejection and fate of trace organic compounds (TrOCs) during membrane distillation

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ARTICLE INFO

Article history:
Received 10 September 2013
Received in revised form
28 November 2013
Accepted 1 December 2013
Available online 10 December 2013

Keywords:
Trace organic compounds (TrOCs)
Direct contact membrane distillation (DCMD)
Volatility
Fate and transport
Hydrophobicity/hydrophilicity

ABSTRACT

In this study, we examined the feasibility of membrane distillation (MD) for removing trace organic compounds (TrOCs) during water and wastewater treatment. A set of 29 compounds was selected to represent major TrOC groups, including pharmaceuticals, steroid hormones, phytoestrogens, UV-filters, industrial chemicals, and pesticides that occur ubiquitously in municipal wastewater. Results reported here suggest that rejection and fate and transport of TrOCs during MD are governed by their volatility and, to a lesser extent, hydrophobicity. All TrOCs with $pK_H > 9$ (which can be classified as non-volatile) were well removed by MD. Among the 29 TrOCs investigated in this study, three compounds (i.e. 4-tert-octylphenol, 4-tert-butylphenol and benzophenone) possess moderate volatility ($pK_H < 9$) and therefore had the lowest rejection efficiencies of 54%, 73% and 66%, respectively. The results suggest that the rejection of TrOCs with $pK_H < 9$ may be governed by the interplay between their hydrophobicity and volatility. In addition, the fate and transport of the TrOCs during the MD process was also investigated. Hydrophilic TrOCs having negligible volatility were concentrated in the feed, while hydrophobic compounds with moderate volatility were substantially lost due to evaporation or adsorption. When MD treatment was integrated with a thermophilic membrane bioreactor (MBR), near complete removal (> 95%) of all 29 TrOCs investigated in this study was achieved despite their diverse physicochemical properties (i.e. hydrophobicity, persistency and volatility). The results suggest that MD could be a promising post-treatment to be used in conjunction with thermophilic MBR for TrOC removal.

1. Introduction

Membrane distillation (MD) is a low temperature distillation process that involves the transport of water in the vapour phase from a feed solution through a microporous and hydrophobic membrane to the distillate (product) side. Direct contact membrane distillation (DCMD) is probably the most widely studied MD system configuration due to its simple operation [1,2]. In DCMD, the feed solution is maintained at a higher temperature than the distillate; thus, creating a vapour pressure difference between the feed and distillate. The membrane separates the liquid phase of the feed and distillate streams but allows water vapour to transport freely through its dry micro porous pores. In MD, the membrane material must be hydrophobic to prevent wetting of the pores by liquid feed or distillate under standard operating conditions. Because mass transfer can occur only in the gas phase, MD can offer complete rejection of all non-volatile solutes such as inorganic salts and pathogenic micro-organisms. As a result, to date, much of the effort in MD research has focused on desalination applications [2–5].

Unlike pressure driven membrane processes, due to the absence of hydraulic pressure, MD is less susceptible to membrane fouling [3,6]. Even when a fouling layer does form on the membrane surface, it is expected to be less compacted and can be easily removed [3,7,8]. The low operating temperature of MD allows the utilisation of solar thermal or low grade heat as the energy source [1,2,9–13]. Given the advantages of high separation efficiency, low fouling propensity, and potentially low energy consumption (when low grade heat is readily available), MD can be used for a range of applications beyond those for brackish and seawater desalination. Several studies have explored the use of MD for food processing, such as whey protein recovery in dairy processing [8], polyphenolic antioxidants recovery from olive oil wastewater [14], and orange juice concentration [15], separation of fermentation broth [16] as well as treatment of wastewater from the textile [17] and petrochemical industries [10], and municipal water reuse [11,18].

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http://dx.doi.org/10.1016/j.memsci.2013.12.002

Despite the growing interest in using MD for treatment of a range of wastewaters, there is still a lack of understanding of the rejection mechanisms of trace organic compounds (TrOCs) by MD. TrOCs have been frequently detected in raw sewage and biologically treated effluent at concentrations ranging from several ng/L to several μg/L [19–23]. As a result, the removal of these TrOCs from secondary treated effluent by advanced treatment processes such as nanofiltration (NF), reverse osmosis (RO), oxidation and activated carbon adsorption has been extensively investigated in recent years [24–28]. Nevertheless, only a few studies have been conducted to elucidate the rejection of specific organic compounds by MD. Moreover, the available studies are mostly concerned with industrial chemicals such as benzene [29] and trichloroethylene [30] at an elevated feed concentration.

Given the concerns associated with human and environmental exposure to TrOCs, it is important to elucidate their fate and transport during MD, particularly in water reuse applications. Examples of these include the investigation by Cath et al. [18] and Cartinella et al. [31] to treat urine and hygiene wastewater by MD for water reuse in long term space missions and the novel membrane distillation bioreactor (MDBR) concept proposed by Phattaranawik et al. [11] and Goh et al. [32].

In this paper, we studied the rejection of a broad range of TrOCs by MD. The potential application of MD as a post-treatment for thermophilic MBR to enhance TrOC removal was also investigated. The transport and fate of TrOCs during MD treatment are discussed with respect to compound hydrophobicity and volatility (measured by the log D and the Henry's law constant, respectively). The results provide further insight with respect to TrOC rejection using MD, which is critical for further development of this technology for wastewater reclamation applications.

2. Materials and methods

2.1. Experimental system

The rejection of TrOCs by MD was evaluated using a hydrophobic microporous polytetrafloroethylene (PTFE) membrane (GE, Minnetonka, MN) and a laboratory-scale DCMD system [33]. According to the manufacturer, the average pore size and porosity of the MD membrane were 0.22 μm and 70%, respectively. The DCMD system (Fig. 1) comprised a membrane cell, a stainless steel feed tank, a glass distillate tank, two circulation pumps (Micropump Inc., USA), a temperature controller (Coleparmer, USA), and a heating element (Process Technology, USA). The membrane cell was made of acrylic glass, and a flow channel was engraved in each of the two acrylic glass blocks that make up the feed and permeate semi-cells. The length, width, and height of the each channel were 145, 95, and 3 mm, respectively. The feed solution was circulated from a stainless steel reservoir to the membrane cell and then returned back to the feed reservoir. A temperature sensor was placed immediately before the feed inlet to the membrane cell. The heating element and the temperature sensor were connected to a temperature control unit that was used to regulate the temperature of the feed solution. Another temperature sensor was installed immediately at the outlet of the distillate semi-cell. The temperature of the distillate was regulated using a chiller (AquaCooler, Australia) equipped with a stainless steel heat exchanging coil immersed directly in the distillate reservoir. Excess water was allowed to overflow from the distillate reservoir into a glass container, placed and continuously weighed on an analytical balance (Mettler Toledo, Switzerland). All pipework used in the DCMD test unit was covered with insulation foam to minimise heat loss. The feed and distillate tanks were covered with aluminium foil to minimise evaporation loss during the experiment. At the end of each experiment, the solution volume was measured again and the total volume loss was found to be less than 6%.

One set of MD experiments was conducted using a synthetic feed solution containing approximately 5 μg/L of each TrOC in Milli-Q water. In another set of experiments, effluent obtained from a thermophilic MBR system (Supplementary Data Fig. S1) was used as the feed solution to evaluate the feasibility of combining MD with MBR. The MBR and MD experiments were conducted separately. The MBR system consisted of a 5 L glass reactor immersed in a PID control water bath (Julabo, Germany), three peristaltic pumps (Masterflex L/S, USA) for feeding, recirculation and effluent extraction, and an external ceramic membrane module (NGK, Japan). The ceramic membrane had a nominal pore size of 1 μm and effective area of 0.09 m^2. Further details of this MBR system are available elsewhere [34].

2.2. Experimental protocol

In all MD experiments, the feed and distillate temperatures were 40 and 20 °C, respectively, and the cross flow velocity of the

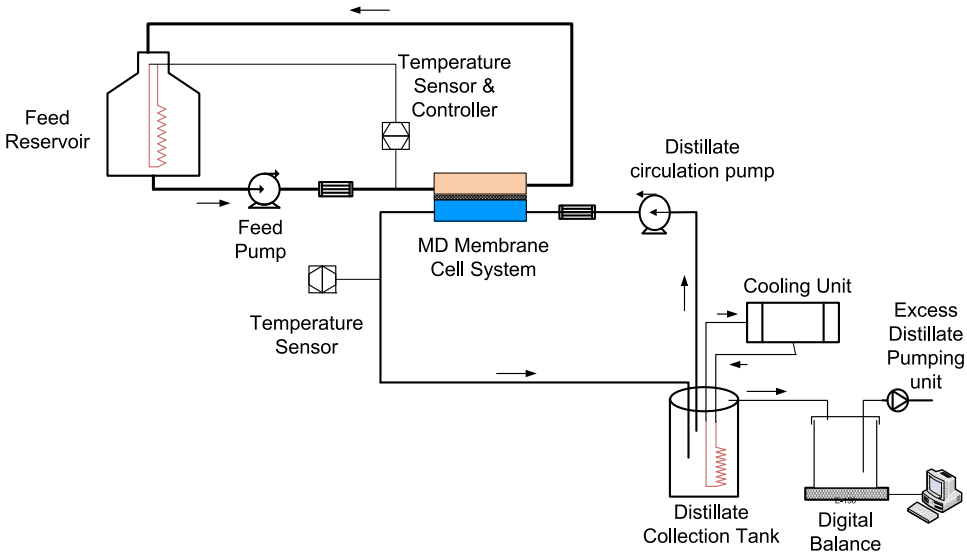

Fig. 1. Schematic diagram of the DCMD system.