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Optimising the recovery of EDTA-2Na draw solution in forward osmosis through direct contact membrane distillation

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ABSTRACT

Ethylenediaminetetraacetic acid disodium (EDTA-2Na) has been demonstrated as an excellent draw solution in the forward osmosis (FO) process because of its high osmotic pressure together with low reverse salt flux but its application is hindered by difficulties in the recovery of draw solution. Hence, in this study, microporous hydrophobic membranes were used in direct contact membrane distillation (DCMD) to concentrate the diluted EDTA-2Na draw solution. The MD was found to require lower operating pressures than do all other widely applied pressure-driven membrane processes, particularly in RO. This study systematically investigated the effect of different polytetrafluoroethylene membranes under various cross flow velocities of 2.67–14.67 cm/s, feed temperatures of 35–60 °C, and distillate temperatures of 10–20 °C in DCMD process for regeneration of diluted EDTA-2Na. The results revealed that DCMD system could achieve a salinity rejection rate exceeding 99.99%; furthermore, the conductivity of the permeate distillate was consistently below 6.4 μS/cm for all of the EDTA-2Na feed concentrations. More importantly, the water flux slightly decreased from 8.27 to 7.04 L/m^2 h when the concentration of the EDTA-2Na feed increased from 0.1 to 0.5 M, corresponding to increased osmolality from 300 to 1411 mOsm/kg, indicating that water flux in DCMD is not significantly influenced by the osmotic pressure gradient across the membrane. This study demonstrated that MD could be an effective method for EDTA-2Na recovery in FO–MD systems and could economically utilize the wasted heat from industrial sources.

1. Introduction

The increasing demand for water in growing numbers of municipal centers together with diminishing freshwater supplies in arid areas has become one of the greatest challenges of the current century. Hence, exploration of advanced technologies is essential for maintaining drinking water supplies. Among suggested technologies, forward osmosis (FO) is the most effective option for improving water sustainability because (1) osmotic pressure is used as the driving force instead of hydraulic pressure, (2) FO membranes exhibit high rejection for a wide range of contaminants, and (3) the membranes have a low fouling tendency [1–4]. These apparent advantages have recently influenced numerous scientists to explore the FO technique, and promising results have been shown in a variety of fields such as desalination, sludge dewatering, wastewater treatment, protein concentration, food processing, and algae concentration [3–14]. However, FO cannot be applied alone; it must be combined with another process to recover the diluted draw solution for reuse. In FO, the energy consumption for recovering diluted draw solution and maintaining water quality in the permeate stream should be considered carefully. Hence, regeneration of diluted draw solution is one of the most notable problems in FO, particularly in drinking water production when high-quality water is required.

Reverse osmosis (RO) combined with FO in FO–RO systems have been widely used for regeneration of draw solution. For example, Holloway et al. [15] employed an FO–RO hybrid system for a specific application using monovalent salt (NaCl) as the draw solute. Although the diluted draw solution could be completely recovered through the RO process with a salt rejection of 99.9%, this system required high hydraulic pressure (>30 bar), subsequently consuming high energy (3 kW h/m^3) and causing high fouling potential. Compared with using RO, using nanofiltration (NF) to regenerate diluted draw solutes is relatively rare. High molecular weight and highly charged valence of draw solutions

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[i.e., ethylenediaminetetraacetic acid disodium (EDTA-2Na), EDTA-$ZnNa_2$, and PAA-Na] have been considered for NF recovery [16,17]. In our previous study, a hybrid FO–NF process was designed for dewatering high-nutrient sludge and recovering diluted draw solution (i.e., EDTA-2Na) with minimum energy and low fouling [17]. However, the recovery of the diluted draw solution by using an NF membrane was incomplete (salt rejection: 93%), which resulted in low water quality and required draw solute replenishment. In addition, magnetic fields (MFs) have also been investigated for the regeneration of magnetic nanoparticles (MNPs) draw solution in numerous studies. Because they do not require hydraulic pressure, FO–MFs hybrid system may be the most economic among other systems employing draw solute regeneration processes such as FO–RO and FO–NF. Lin et al. [18] proposed the regeneration of MNPs by MFs separator; however, this method caused the agglomeration of MNPs, which significantly decreased their osmotic pressures and therefore the reduced water flux. Ultrasonication technology was suggested to prevent this problem; however, this potentially weakens the magnetic properties of MNPs and thus reduces regeneration efficiency and reusability. Furthermore, synthetic draw solutes such as thermoresponsive copolymer, ferric and cobaltous hydroacid complexes, 2-methylimidazole-based compounds, and switchable polarity solvents have been used, showing promise in terms of negligible reverse solute fluxes or low energy consumption during regeneration [19–22]. Nevertheless, most novel draw solutes undergo complicated synthesis procedures and high viscosity. In 2016, Long et al. used gluconate salts and carboxyethyl amine sodium salts are as potential draw solutions for FO applications [23,24].

Recently, membrane distillation (MD) has been used as an excellent alternative process for regenerating diluted draw solution and producing high-purity distillate, enabling operation by means of low-energy sources such as waste heat as well as solar and geothermal power. MD is a thermal separation process employing hydrophobic microporous membranes sandwiched between cold and warm solutions. MD has an effective chemical resistance; there is no need to pretreat MD membranes. Moreover, the driving force of MD varies with membrane operations because of transmembrane temperature differences that are attributable to the partial vapor pressure differences across the membrane [25]. The previous studies on FO-MD hybrid system [20,22,26–29] demonstrated that a stable water flux was obtained using a laboratory-scale FO–MD system operating continuously with high water recovery, whereas MD reconcentrated the draw solution successfully and produced high quality water. However, no studies have systematically investigated the use of MD membranes to regenerate diluted EDTA-2Na draw solution in FO. Hence, in this study, diluted EDTA-2Na draw solution was recovered in direct contact MD (DCMD) by using different cross flow velocities, membranes, temperatures, and feed concentrations.

2. Materials and methods

2.1. Material and membranes

Laboratory-grade EDTA-2Na with a purity of 99.0% was purchased from Sigma-Aldrich Co., Germany. Three types of polytetrafluoroethylene (PTFE) MD membranes with different pore sizes were provided by Ray-E Creative Co., Ltd., Taiwan. The characteristics of the three types of MD membranes are shown in Table 1.

Table 1
Characteristics of the MD membranes.

Membrane	Pore size	Contact angle
PTFE	0.1 μm	126 ± 5°
PTFE	0.45 μm	114 ± 4°
PTFE	1.0 μm	126 ± 2°

2.2. DCMD experiment setup

The diluted draw solution was recovered through a laboratory-scale crossflow MD membrane cell (Ray-E Creative Co., Ltd., Taiwan), as shown in Fig. 1. The membrane cell was composed of acrylic and comprised two semi-cells. Each semi-cell had a flow channel with a depth, width, and length of 0.3, 10, and 10 cm, respectively. The effective membrane area for mass transfer was 100 cm^2. A peristaltic pump (Baoding Longer Precision Pump Co., Ltd., Taiwan) with two pump heads was employed to circulate the feed and distillate solutions through each semi-cell at cross flow velocities of 2.67–14.67 cm/s. The temperature of the feed and distillate tank was controlled using two water baths (D-606, Deng Yng, Taiwan). In the DCMD process, the EDTA-2Na solution (1 L) as feed was continuously pumped at a controlled temperature from a feed reservoir to the membrane cell, subsequently returning to the reservoir. DI water was used for the initial distillate stream. The distillate was circulated from a 1-L reservoir through the distillate membrane semi-cell and back to the reservoir. An analytical balance was applied to weigh the excess distillate flow to determine the water flux.

Theoretical water flux (J_w^T) across the MD membrane is the product of a mass transfer coefficient (K_m, L/Pa m^2 h) and the difference in water vapor pressure between the vapor–liquid interfaces formed at each side of the membrane (ΔP, Pa).

$$J_w^T = K_m \Delta P \quad (1)$$

The mass transfer coefficient is a function of membrane properties and operating conditions, comprising feed and distillate temperatures, pressures, and circulated flow rates [25,30]. In the experiment, water flux was calculated from the weight changes of the MD permeate by using Eq. (2).

$$J_w = \frac{\Delta V}{A \Delta t} \quad (2)$$

where ΔV is the total increase in the volume of the permeate water (L) collected over a predetermined period Δt (measured in hours) and A is the effective MD membrane area (measured in square meters).

The salinity can be obtained from the following equation:

$$R = \left(1 - \frac{C_p}{C_{Fi}}\right) 100\% \quad (3)$$

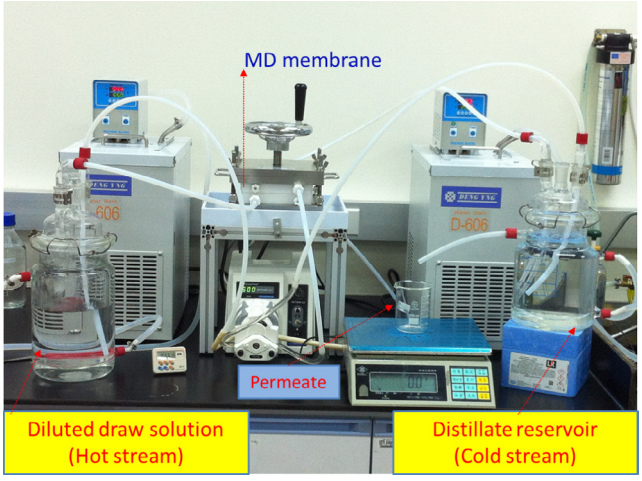

Fig. 1. Laboratory-scale DCMD system for EDTA-2Na draw solution recovery.

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