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Application of an innovative draw solute in forward osmosis (FO) processes

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Abstract:

More attention is being paid to forward osmosis (FO) as a breakthrough technology to tackle environmental pollution due to advantages such as high contaminant removal and low energy consumption. Nevertheless, FO applications remain limited by the lack of ideal draw solutes that simultaneously achieve high permeate flux and low salt leakage flux. Therefore, this paper aims to increase water flux while maintaining low reverse salt diffusion by using a low concentration of a highly charged organic EDTA compound coupled with inorganic NaCl salt as a novel draw solute. Results of the FO performance revealed that a draw solute of 0.3 M EDTA-2Na mixed with 0.6 M NaCl yielded a higher water flux (J_w=8.82 l/m^2 h) when compared to 0.9 M NaCl only (J_w=7.61 l/m^2 h). Moreover, the FO-membrane distillation system produced good quality drinking water with a total dissolved solid (TDS) of <5 mg/l from the permeate stream originating from influent brackish water with a TDS of 6000 mg/l. The analysis results from scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM-EDX) images observed a cake layer of NaCl on the FO membrane surface.

Keywords: **brackish water, distillation, forward osmosis, membrane draw solute, reverse salt flux.**

Classification number: **2.3**

Introduction

Nowadays, climate change has significantly impacted the life of people especially as rising sea levels lead to an increase in salinity intrusion of many water sources such as the Vietnamese Mekong river delta and the Cau Do river [1]. Saltwater intrusion has caused a lot of problems for drinking water treatment plants using coagulation-sedimentation-filtration-disinfection technology because these traditional technologies are not equipped to treat brackish water. Therefore, water scarcity by salinity intrusion is an increasingly serious global problem and clean water production through the treatment of brackish water is receiving more attention as a solution to freshwater shortages. Among suggested technologies, reverse osmosis (RO) is the most widely employed technology for brackish desalination [2, 3]. However, RO processes consume large amounts of energy because they require high pressure. The limitations of RO membrane technology include severe membrane fouling, scaling, and low water recovery. Furthermore, the discharge of RO concentrate streams are damaging to the environment [4-6]. Hence, it is essential and urgent to investigate a sustainable and environmentally friendly water production technology.

Forward osmosis is a green technology for clean water production [7-10]. Unlike RO process, FO uses natural osmotic pressure to draw permeate water from a feed solution to a draw solution. The semipermeable membrane allows clean water to pass through but rejects solutes [8, 11-13]. Therefore, the FO process can be operated at low or negligible pressure. Many studies have reported that FO is a feasible process because of its low energy consumption, low fouling propensity, and high rejection of various contaminants [11, 14-16]. Although FO has been widely used in brackish desalination and wastewater reclamation [7-9], exploring an appropriate draw solute remains a particularly crucial and major challenge. High permeate stream, minimal salt leakage, nontoxicity, as well as efficient

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regeneration, should be maintained in an ideal draw solute [6, 17-19].

Over the past few years, many categories of materials such as inorganic salts and organic solutes have been studied and evaluated as potential optimized draw solutions [17, 20-23]. These draw solutions are easily recovered using an RO membrane, are inexpensive, and produce a relatively high permeate stream. However, both problems of severe reverse solute fluxes during the FO process and high energy consumption during the regeneration process hinder their application. Furthermore, some new draw solutes such as polydiallyldimethylammonium chloride [24], 2-methylimidazole-based compounds [25], and hexavalent phosphazene salts [26] have shown promising results with low salt leakage and easy recovery. Despite these advantages, most of these synthetic draw solutes produce a lower water flux than conventional draw solutions. In our group's previous research, EDTA-2Na was explored as a draw solution for a hybrid FO-nanofiltration (NF) process [27]. These draw solutions, with large molecular size and high osmotic pressure, have received considerable attention [28, 29] because they performed much better than others in terms of relatively high water flux and low reverse solute flux. However, the high viscosity and limitation of the solubility of EDTA-2Na at high concentration remain as challenges for FO applications, which is the main reason for the author to conduct this research.

To the best of our knowledge, a mixed draw solute of highly charged organic EDTA with highly soluble NaCl salt has not yet been used in an FO-membrane distillation (FO-MD) system to simultaneously achieve a high water flux and maintain a low reverse salt flux. Therefore, this study aims to do the following: (1) assess the influence of various draw solute concentrations on FO system; (2) evaluate the efficiency of the application of NaCl mixed with EDTA-2Na as draw solute in FO/MD for desalinating brackish water; and (3) investigate the water quality from the permeate stream of an MD system.

Materials and methods

FO and MD membranes

In this study, we used cellulose triacetate with an embedded support cartridge-type (CTA.ES) FO membrane provided from Hydration Technology Innovations (American). Its characteristics are shown in Table 1. We used an MD membrane, namely, a polytetrafluoroethylene (PTFE) membrane, with 0.45 µm pore sizes and 114±4° contact angle, delivered by the Ray.E.Creative Taiwanese Company. Before use in the MD process, the membrane was washed with clean water and dried at room temperature.

Table 1. Characteristics of CTA.ES FO membrane.

CTA.ES FO membrane	Value
Size for each piece	15×22 cm^2
Contact angle	60-80°
pH range	2-8
Salt rejection	95-99%
Post treatment	Soak in DI water

Preparation of feed solution and draw solution

We used EDTA-2Na with purity >99% from Sigma-Aldrich Co., Ltd., (Germany) and NaCl salt with purity >99% from Vietnamese salt Co., Ltd., (Vietnam). The draw solutions were prepared using a combination of 0.3 M EDTA-2Na and different concentrations of NaCl. To all draw solutions, NaOH solution was added to adjust to pH 8 and they were stirred for 1 d before being used in the FO process. DI water was used as feed solution in FO to test water flux and reserve salt flux. Synthetic brackish water served as the feed solution for the FO desalination process. The synthetic brackish water was made with a total dissolved solid concentration of 6000 mg/l by mixing NaCl salt into DI water.

FO-MD process

All FO-MD experiments were conducted with a lab-scale FO-MD setup, as illustrated in Fig. 1. The CTA.ES FO membrane was used for all FO experiments. The FO cell was composed of two semi-cells, each of which was engraved to form a rectangular flow channel with length×width×height of 9.2×4.5×0.2 cm, respectively. The membrane coupons were inserted in the membrane cell such that the active layer faced the feed solution (FO mode) and the flow rate of the feed and draw solutions were both fixed at 500 ml/min. Furthermore, 0.3 M EDTA-2Na mixed with different concentrations of NaCl (0.1, 0.2, 0.4, 0.6 and 0.8 M) were prepared as the draw solutions. Synthetic brackish water served as the feed solution for FO desalination process. We prepared 1 l for the feed solution and draw solution, and then placed it on a scale (BW12KH, Shimadzu, Japan) to monitor weight variations versus time. Then, the mass change of the feed solution was converted to volume change based on the density of the feed solution.

The experimental water flux (J_w, l/m^2 h) was calculated according to the volume variation in the feed tank with time:

$$J_w = \frac{\Delta V}{A\Delta t} \quad (1)$$

where A is the membrane area (m^2) and ΔV is the increased water volume (l) of draw solution obtained in a time interval Δt (h).

The reverse salt flux of the draw solution (J_s, g/m^2 h) was determined from the amount of salt accumulated in the feed tank:

$$J_s = \frac{V_t.C_t - V_0.C_0}{A.t} \quad (2)$$

where C_t and C_0 are the concentration of the feed solution measured at time t (h) and initial time (t=0 h), respectively, and V_t and V_0 are the volume of the feed solution at time t (h) and initial time (t=0 h), respectively.

Following the FO tests, the MD process was conducted to recover the diluted draw solution using an MD cell module (Sterlitech, USA). The FO membrane module was produced from acrylic material and composed of two semicells with a flow channel 0.2 cm deep, 4.5 cm wide, and 9.2 cm long. We pumped and circulated the distillate and feed through each semicell with a velocity of 500 ml/min. Moreover, 0.3 M EDTA-2Na mixed with 0.6 M NaCl was used as the hot feed solution and was temperature controlled at 55±0.5°C, whereas cold DI water used as the original distillate was maintained at 25±0.5°C. The feed solution and distillate were continuously pumped from their reservoirs through each semicell membrane and then back to the reservoirs. The permeate water from the distillate tank that overflowed into the clean water tank was weighted by a digital weighing scale. Using Eq. (1), the water flux was calculated from the volume changes of the MD permeate.

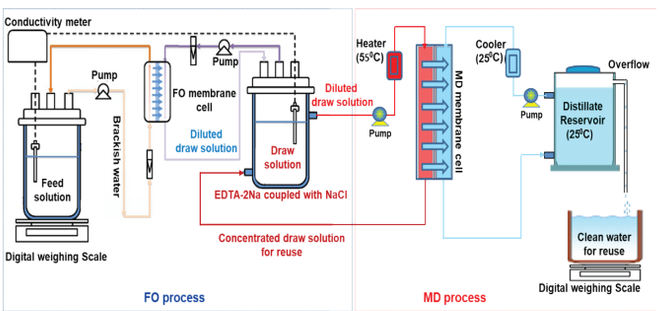

Fig. 1. Illustration of FO-MD system for desalinating brackish water.

Rejection of TDS can be calculated by the equation:

$$R = (1 - \frac{C_P}{C_F}).100\% \quad (3)$$

where R is the TDS rejection, C_P (mg/l) is the TDS concentration in the permeate, and C_{Fi} (mg/l) is the initial feed concentration.

Analytical methods

Viscosity was obtained using a Viscometer from Japanese Company and conductivity was determined by a conductivity meter (China). Furthermore, we used CAM 100 (Opto-Mechatronics P Ltd., India) to measure the membrane contact angle. Membrane fouling was detected through scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM-EDX). Osmolality was measured using an Osmometer (Model 3320, Advanced Instruments, Inc., USA) on the basis of the freezing-point depression method. The concentration of permeation solutions was analysed using a total organic carbon (TOC) analyser from Japanese Shimadzu Company.

Results and discussion

The influence of draw solution concentrations on water flux and reverse salt flux

Figure 2 presents the variations in the water flux and reverse salt flux for different NaCl concentrations (from 0.1 to 0.8 M) coupled with 0.3 M EDTA-2Na as the draw solutes. The FO experiments were conducted in the membrane orientation of the active layer facing the feed solution of the DI water. The water flux gradually increased from 5.73 to 9.24 l/m^2 h when the concentration of NaCl increased from 0.1 to 0.8 M due to the increase in the osmolality (from 896 to 1584 mOsm/kg) in the draw solution (Fig. 3). Clearly, the increase in FO water flux was not linear with increasing osmolality of the draw solution. This non-linearity could be explained by the rise in viscosity from 1.41 to 1.79 cp (Fig. 3) when the draw solution concentration rose from 0.1 to 0.8 M NaCl, which led to the prevention of permeable water through the FO membrane. Moreover, the reverse salt flux rose from 1.42 to 2.95 g/m^2 h as NaCl concentrations increased from 0.1 to 0.8 M into the 0.3 M EDTA-2Na draw solution, as shown in Fig. 2.

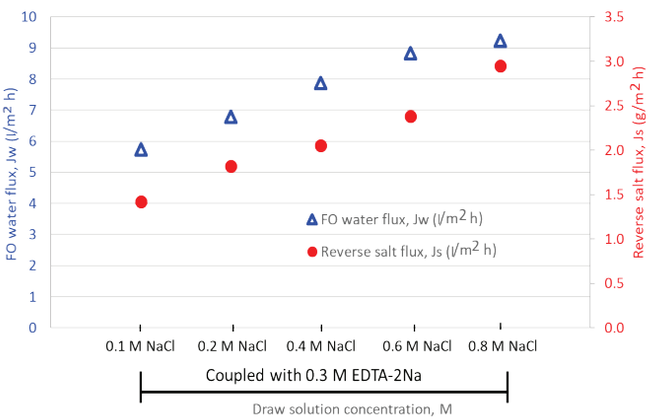

Fig. 2. Variation of reverse salt and water flux using 0.3 M EDTA-2Na mixed with various NaCl concentrations as draw solution.

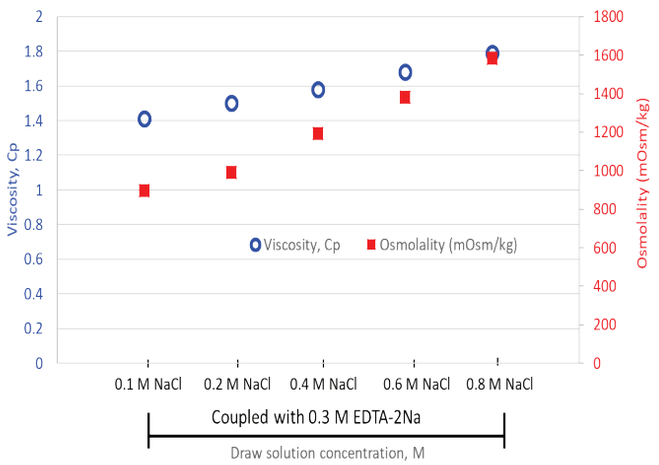

Fig. 3. Variation of osmolality and viscosity using 0.3 M EDTA-2Na mixed with various NaCl concentrations as draw solution.

Meanwhile, the reverse salt fluxes of 0.4 M-1.1 M NaCl as the only draw solution quickly increased from 3.18 to 5.93 g/m^2 h, which is much higher than the measured value of EDTA-2Na mixed with NaCl as the draw solutions (Fig. 4). The reason for the different reverse salt flux of the two kinds of draw solution can be explained by the influence of the complexation and highly charged compounds present in the EDTA-2Na and NaCl mixed draw solution. For instance, we observed 14.4% of Na[EDTA]$^{3-}$complexion and 81% of trivalent compound of H[EDTA]$^{3-}$(complexion and charge formation are observed by Mineql+ software) when coupling 0.3 M EDTA-2Na into NaCl, which resulted in a reduced reverse salt flux [18, 19, 30]. This is the most noteworthy aspect of using the mixed highly charged draw solution of EDTA-2Na into NaCl.

As seen in Fig. 2, between the 0.6 M NaCl concentration coupled with 0.3 M EDTA-2Na (J_w=8.82 l/m^2 h) and 0.8 M NaCl coupled with 0.3 M EDTA-2Na (J_w=9.24 l/m^2 h), the difference in water flux was negligible, however, the reverse salt flux of 0.8 M NaCl coupled with 0.3 M EDTA-2Na (J_s=3.01 g/m^2 h) was much higher than that of 0.6 M NaCl coupled with 0.3 M EDTA-2Na (J_s=2.38 g/m^2 h). These results revealed that 0.6 M NaCl coupled with 0.3 M EDTA-2Na proved to be the optimum concentration of a draw solution in the FO process for simultaneously obtaining low reverse salt flux (J_s=2.38 g/m^2 h) and high water flux (J_w=8.82 l/m^2 h).

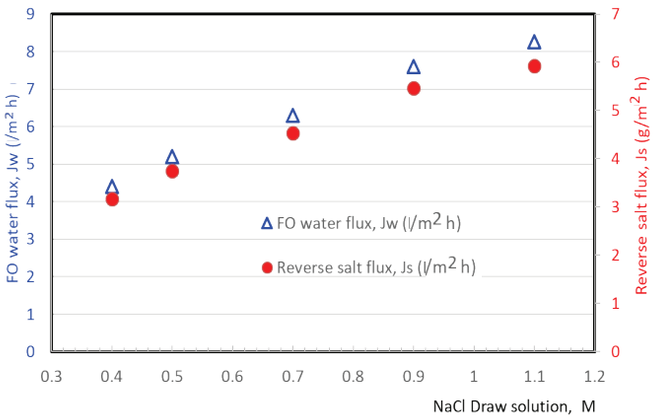

Fig. 4. Variation of water flux and reverse salt flux using pure NaCl with various NaCl concentrations as draw solution.

Application of EDTA-2Na mixed with NaCl as the draw solute in FO/MD for desalinating the brackish water

Concerning the optimum FO performance among the various NaCl concentrations, 0.6 M NaCl mixed with 0.3 M EDTA-2Na was selected as the draw solution for studying brackish desalination through the FO-MD hybrid system. Fig. 5 depicts the water flux of the FO process for desalinating the synthetic brackish water of 6000 mg/l TDS concentration. The FO water flux decreased from 6.78 l/m^2 h (over the first two hours) to 6.01 l/m^2 h (over the last ten hours). After the 10-h operation, the FO water flux was reduced by 10.32%. A possible reason for the decline in FO permeate flux is the increase in TDS of the feed solution (from 6000 mg/l to 8100 mg/l), which caused an increase in the osmotic pressure of the feed solution, which then reduced the net driving force across the FO membrane.

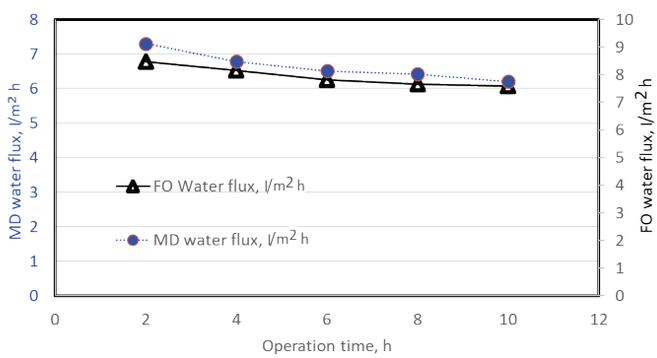

Fig. 5. Variation of FO water flux and MD water flux for the desalination of brackish water versus operation time (0.3 M EDTA-2Na coupled with 0.6 M NaCl as draw solute, MD membrane: PTFE 0.45 µm; hot stream: 55±0.5°C; distillate stream: 25±0.5°C).

Moreover, the reduction in water flux can be attributed to the concentration polarization (CP) effect because of the salt accumulation (NaCl) on the active layer of the FO membrane. In fact, salt accumulation on the FO membrane surface can be observed in Fig. 6A with 6000 mg/l of NaCl as the feed solution. There were some cake layers of NaCl attached to the FO membrane that were identified by EDS through the appearance of peaks indicating the elements Na and Cl (Fig. 6B). This result agrees with that reported by Alnaizy, et al. [22], who showed that the water flux decreased as feed concentration was increased due to a reduced osmotic pressure gradient between feed and draw solution and increased concentration polarization phenomenon.

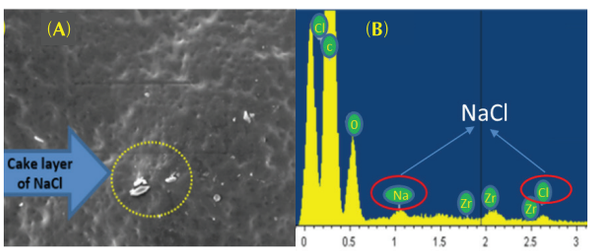

Fig. 6. (A) SEM picture and (B) EDS graph of a used membrane (0.3 M EDTA-2Na coupled with 0.6 M NaCl as draw solute, 6000 mg NaCl/l as feed solution, draw solution facing the support layer, pH of 8, temperature of 25±0.5°C).

As can be seen from Fig. 5, 0.45 µm PTFE was used as an MD membrane for diluted draw solution recovery and the slight decrease in MD water flux from 9.12 to 7.75 l/m^2 h can be attributed to membrane fouling. This outcome agrees with Elzahaby, et al. [31], who observed that a high feed tank temperature could lead to membrane fouling and reduce its performance.

Water quality from permeate stream of MD system

The recovery of the diluted draw solute (0.3 M EDTA-2Na coupled with 0.6 M NaCl) from FO was induced by the MD process to reuse the draw solution and separate the clean water under the conditions of a hot stream of 55±0.5°C and a distillate stream of 25±0.5°C. Table 2 illustrates the variation of TOC and TDS in the permeate stream from the MD system using the 0.45 µm PTFE MD membrane during 10-h operation. The results showed that the 0.45 µm PTFE efficiently removed almost all ions (more than 99.9%) from the diluted draw solution during the 10-h operation. The overall high salt rejection observed here can be largely attributed to the MD process in which only water vapour is transported through the membrane pores. The concentration of TOC in the permeate stream had a slight increasing trend versus operating time (from 0.48 to 0.93 mg/l), however, the TOC concentration is still lower than that of the drinking water standard. In addition, the TDS concentration in the permeate stream of the MD system slightly rose from 0.95 to 4.54 mg/l after the 10-h operation, which was lower than that of the National Technical Regulation on drinking water quality (QCVN 01:2009/BYT with TDS<1000 mg/l). This result demonstrated that the FO-MD hybrid system can produce high quality drinking water from brackish water.

Table 2. Water quality from permeate stream of MD system for desalinating of synthetic brackish water.

Operating time, h	2	4	6	8	10
TOC in permeate stream, mg/l	0.48	0.59	0.72	0.84	0.93
TDS in permeate stream, mg/l	0.95	1.16	1.47	2.48	4.54
TDS rejection, %	99.99	99.99	99.98	99.96	99.92

Experimental condition: MD membrane: PTFE 0.45 µm; hot stream: 55±0.5°C; distillate stream: 25±0.5°C; Feed and distillate velocity: 500 ml/min; diluted draw solute as feed: 0.3 M EDTA-2Na coupled with 0.6 M NaCl.

Conclusions

The highly charged organic EDTA-2Na compound coupled with highly soluble inorganic NaCl salt creates a suitable draw solute for simultaneously achieving a low reverse salt flux and high water flux in the FO process. The results revealed that the relatively high water flux of 8.82 l/m^2 h and low reverse salt flux of 2.38 g/m^2 h were obtained when 0.3 M EDTA-2Na coupled with 0.6 M NaCl was used as the draw solute and DI water served as the feed solution. Moreover, 0.3 M EDTA-2Na was mixed with 0.6 M NaCl in the brackish desalination process, yielding average water fluxes of 6.35 l/m^2 h when brackish water with a TDS of 6000 mg/l was used as the feed solution in the FO process. Notably, a diluted draw solution was successfully recovered using a 0.45 µm PTFE MD membrane, with an average water flux of 8.30 l/m^2 h and more than 99.9% rejection of salinity.

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COMPETING INTERESTS

The authors declare that there is no conflict of interest regarding the publication of this article.

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