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Exploration of EDTA sodium salt as novel draw solution in forward osmosis process for dewatering of high nutrient sludge

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ABSTRACT

In this study, a hybrid forward osmosis–nanofiltration (FO/NF) process was designed for dewatering high nutrient containing sludge and recovering draw solution with minimum energy as well as low fouling. A novel draw solution – EDTA sodium salt – was also systematically studied for dewatering process. Results show that using EDTA sodium salt produced higher water flux and lower reverse salt flux when compared to conventional inorganic salt (NaCl) at pH 8. The final sludge concentration reached 32,000 mg/L after 16 h of operation. Moreover, nutrient compounds in sludge were successfully removed by the FO membrane with a removal efficiency of approximately 97% of NH_4^+–N, 90% of NO_3^-–N, 97% of NO_2^-–N and 99% of PO_4^{3-}–P, which was attributed to the multi-barrier layers of sludge forming on membrane surface and the steric effect of the FO membrane. The NF recovery of EDTA sodium salt indicated that all NF membranes performed well and TS-80 was the best among the tested membranes.

1. Introduction

Forward osmosis (FO) has been considered as a promising technology to solve environmental challenges with excellent advantages such as high rejection of many pollutants, without aid pressure and low membrane fouling. It not only reduces the stresses of wastewater treatment and seawater desalination but also promotes sludge dewatering meaningfully [1–6]. Unlike reverse osmosis (RO), FO is an engineering process utilizing natural osmosis whereby the water from the feed solution passes through a semi-permeable membrane to minimize operation cost and increase the lifetime of membrane [7]. Theoretically, the water flux (J_w) across the FO membrane is the product of the membrane permeability coefficient (A), the reflection coefficient (σ) and the osmotic pressure driving force ($\Delta\pi$) [8,9]:

$$J_w = A\sigma\,\Delta\pi \quad (1)$$

with

$$\pi = iCRT \quad (2)$$

where i is the Van't Hoff factor, C is the concentration of solute (mol/L), R is the universal gas constant (R=0.0831 L bar/(mol/°C)) and T is the absolute temperature (°C). However, the actual water flux is much lower than the theoretical water flux due to both internal concentration polarization and external concentration polarization effects [10–13].

Recently, new FO membranes have been synthesized to improve membrane permeability [14–16]. Chung and co-worker (2012) showed that the modified membranes could achieve 160% higher water flux compared with the traditional FO membranes, which was a good evidence to apply the FO technology in both academia and industry [10]. However, so far, the development of FO technology is still hindered by the unavailability of suitable draw solutions and the exploration of new draw solutes is vital for future FO applications. Basically, an ideal draw solution must simultaneously have the following characteristics: (i) high osmotic pressure; (ii) minimal reverse draw solute flux; and (iii) easy recovery. Previously, Holloway et al. [17]) used traditional inorganic salt (NaCl) as the draw solution in a continuous FO/RO process for concentration of anaerobic digester [17]. Although promising results have been recorded with high water flux (J_w=14 L/m^2 h at 23 ± 1 °C with 70 g/L NaCl draw solution) and 75% water recovery, the most critical drawback was high recovery energy of RO (4 kWh/m^3). As presented in our previous study [3], utilizing available seawater as the draw solution to sludge dewatering process without draw solution recovery requirement proved economically feasible. However, the high salt leakage from the seawater side into the feed solution side was the major obstacle to reuse sludge for agricultural purposes.

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To overcome the disadvantage mentioned above, Yong et al. [18] investigated the reverse salt flux of neutral draw solute glucose across an asymmetric FO membrane. Low reverse salt flux was obtained and diluted glucose could be recovered by the NF membrane. Nevertheless, the measured water flux for the glucose system (around 4.7 L/m^2 h with 1 M glucose) is one-half of the water flux measured for the NaCl system. Similarly, McGinnis and Elimelech [19] investigated a novel ammonia carbon dioxide forward osmosis desalination process and solute recovery was conducted by a single vacuum distillation column at low temperature (40 °C). The cost of the system was expected to be very competitive with an electrical power requirement of typically less than 0.25 kWh/m^3; however, the reverse salt flux of monovalent ions (NH_4^+) was a problem. In addition, synthesis of magnetic nanoparticles (MNPs) could provide a new way of designing draw solutions for the FO process [20–22]. It is noteworthy that the large particle size of the MNPs had a superior advantage in preventing salt leakage, whereas the observed particle aggregation was troublesome during recycling via a magnetic separator. Switchable polarity solvents (SPS) were also used as viable FO draw solutions [23]. With the presence of carbon dioxide, the tertiary amines were soluble in water and the removal of carbon dioxide from the SPS draw solution resulted in separation of the tertiary amine from the product water due to increased hydrophobicity of the SPS draw solution. However, polar to nonpolar switch was a complex process that depends on reaction chamber surface area, temperature, purge gas, and pressure. Chung and co-workers explored several new types of draw solutions for use in FO such as polyelectrolytes and 2-methylimidazole-based organic compounds where product water is recovered by application of pressure-driven UF process and membrane distillation process, respectively [24,25]. More recently, using copper sulfate as the draw solute in FO desalination has been considered as a good idea as diluted draw solution was recovered by metathesis precipitation without thermal or electrical energy [26,27]. However, copper sulfate, barium hydroxide, and sulfuric acid are all consumables and barium sulfate is produced as a waste. Hence, recovery of copper sulfate draw solution is costly and causes secondary environmental pollution. Stone et al. [28] used hexavalent phosphazene salts as draw solutes in forward osmosis. These salts created high osmotic pressure and might be suitable for seawater desalination.

Although many research efforts have been tested with various chemicals as solutes for draw solutions in the FO process, no work has been found using EDTA sodium salt as the draw solution. Thus, the objective of this study was to explore a novel draw solute – EDTA sodium salt – that could generate a high water flux with a minimal reverse salt diffusion. Fundamentally, EDTA exhibits high charged compounds in high pH conditions according to the following equations:

$$H_4EDTA \leftrightarrow H_3EDTA^- + H^+ \qquad pKa_1 = 1.99 \tag{3}$$

$$H_3EDTA^- \leftrightarrow H_2EDTA^{2-} + H^+ \qquad pKa_2 = 2.67 \tag{4}$$

$$H_2EDTA^{2-} \leftrightarrow HEDTA^{3-} + H^+ \qquad pKa_3 = 6.16 \tag{5}$$

$$HEDTA^{3-} \leftrightarrow EDTA^{4-} + H^+ \qquad pKa_4 = 10.26 \tag{6}$$

Clearly, at high pH, trivalent and tetravalent ions are much easier to remain at the draw solution side, which limits the diffusion of the counterion. More specifically, high charged compounds in draw solution will be easily regenerated with the aid of nanofiltration (NF). Furthermore, the safety of EDTA sodium salt for the environment and human is one of the advantages for its application in water treatment. In this study, the performance of EDTA sodium salt as draw solution in the FO process for dewatering high nutrient sludge was symmetrically investigated. Firstly, the effects of pH, cross-flow rate and draw solution concentration on the FO process were evaluated with deionized (DI) water as the feed solution with the active-layer-facing-draw-solution orientation. Next, simultaneous sludge dewatering and nutrient removal of FO were examined with activated sludge as the feed solution. Finally, the recovery of the diluted EDTA sodium draw solution after FO tests was also conducted with four kinds of NF membranes to investigate the separation mechanisms and determine the appropriate membrane.

2. Materials and methods

2.1. Materials

Commercial asymmetric cellulose triacetate (CTA) cartridge-type membranes (Hydration Technology Inc., Albany, OR, USA) were used for all FO experiments. The FO membrane thickness is approximately 50 μm with measured contact angle of 61° and the membrane has been recorded to be negatively charged in typical feed waters [29]. Characteristics of four NF membranes used in EDTA sodium recovery are shown in Table 1. Laboratory-grade EDTA–2Na (purity of 99.0%) was purchased from Sigma-Aldrich Co, Germany. D-glucose, NH_4Cl and K_2HPO_4 (Merck Co. Ltd., Germany) were used as organic and nutrient concentrations of activated sludge. Samples of activated sludge were first collected at the secondary sedimentation tank of new Taipei wastewater treatment plant (WWTP) in Taiwan with mixed liquor suspended solid (MLSS) concentration of 8000 mg/L. And then glucose, NH_4Cl and K_2HPO_4 solution was added to the raw sludge to produce synthetic sludge with dissolved organic carbon (DOC) of 550 mg/L, NH_4^+–N of 150 mg/L and PO_4^{3-}–P of 150 mg/L (Table 2). The calculation of concentration units for FO studies should be molality (mol/kg) instead of molarity (M, mol/L) [9]. However, in diluted solution, the difference of these two is small and thus molarity is used throughout the manuscript.

2.2. The production of draw solutions

Since the formation of high charged species of EDTA strongly depends on pH, the key point is the pH control of the EDTA draw solution. Hence, 4 M concentration of NaOH solution was prepared first, and for example, the pH of 0.3 M EDTA sodium salt draw

Table 1
Characteristics of nanofiltration membranes.

Membrane name	Manufacturer	Material	Molecular weight cut-off (MWCO)	Rejection	25 °C pH range
NF-XN45	TriSep	Polypiperazine amide	500	(95.0% $MgSO_4$)	2–11
NF-DL	GE Osmonics	Thin film composite			1–11
NF-TS80	TriSep	Polyamide	150	(99.0% $MgSO_4$)	2–11
NF-DK	GE Osmonics	Thin film composite	150–300	(96% $MgSO_4$)	2–10