Science of the Total Environment 557–558 (2016) 44–50

Contents lists available at ScienceDirect

Science of the Total Environment

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

Exploring high charge of phosphate as new draw solute in a forward osmosis–membrane distillation hybrid system for concentrating high-nutrient sludge

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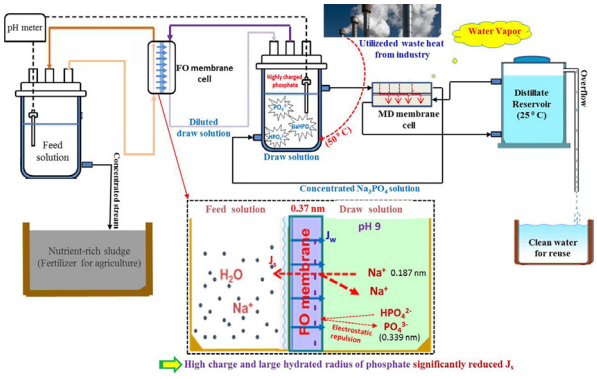
HIGHLIGHTS

- High charge of phosphate was employed as the draw solute in an FO–MD process.
- At pH 9, Na_3PO_4 draw solution obtained the lowest specific reverse salt flux.
- After 15 h, sludge concentration was concentrated up 6 times in the FO mode.
- Draw solution was regenerated using polytetrafluoroethylene with 100% rejection.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 15 February 2016
Received in revised form 4 March 2016
Accepted 4 March 2016
Available online xxxx

Editor: D. Barcelo

Keywords:
Forward osmosis
Membrane distillation
Draw solution
High charge
Complexion

ABSTRACT

For the first time, a high charge of phosphate was used as the draw solute in a forward osmosis–membrane distillation (FO–MD) hybrid system for concentrating high-nutrient sludge. A high water flux (12.5 L/m^2 h) and a low reverse salt flux (0.84 g/m^2) were simultaneously achieved at pH 9 by using 0.1 M Na_3PO_4 as the draw solute and deionized water as the feed solution in the FO process. The specific reverse salt flux of 0.1 M Na_3PO_4 (J_s/J_w = 0.07 g/L) was considerably less than that of 0.1 M NaCl (J_s/J_w = 0.37 g/L) because the complexion between Na^+ and HPO_4^{2-} at pH 9 led to the reduction of free Na^+ ions, which subsequently reduced the reverse salt diffusion substantially. Moreover, for a feed solution with an initial sludge concentration of 3500 mg/L, the sludge concentration could be concentrated to 19,800 and 22,000 mg/L in the pressure-retarded osmosis (PRO) and FO membrane orientations, respectively, after 15 h of operation. Four types of MD membranes were selected for draw solution recovery; of these, a polytetrafluoroethylene membrane with a pore size of 0.45 μm was the most effective in achieving a high water flux (10.28 L/m^2 h) and high salt rejection (approximately 100%) in a diluted Na_3PO_4 draw solution.

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http://dx.doi.org/10.1016/j.scitotenv.2016.03.025
0048-9697/

1. Introduction

A large amount of sludge with a high-water-nutrient content, especially nitrogen and phosphorus, is often produced in wastewater treatment plants (Bourioug et al., 2015). Consequently, sludge dewatering is obligatory for such treatment plants and accounts for approximately 50%–60% of the total operating cost of the entire wastewater treatment plant (Appels et al., 2008; Rai et al., 2004). Although conventional sludge dewatering techniques such as clarification, gravity thickening are simple, the clarification is difficult to meet stringent regulations for effluent water quality and the sludge retention time is long and an unpleasant odor is emitted in gravity thickening process (Kim et al., 2010; Wang et al., 2008). Furthermore, the high-nutrient-rich centrate obtained from the stabilization of digested sludge increases the influent nitrogen loading by 15%–20% and phosphorus loading by 8% after combining with raw wastewater (Fux et al., 2002; Wild et al., 1997). This is a major challenge because the conventional biological process can reduce the biochemical oxygen demand considerably, but it does not reliably achieve the target discharge limits for nitrogen and phosphorus.

To solve the problems of conventional sludge dewatering technologies and shorten sludge treatment processes, Wang et al. (2008) used polyvinylidene fluoride membranes (microporous membranes) for performing sludge thickening and digestion during 1 cycle (2 days). This sludge reduction system is actually a membrane bioreactor (MBR) offering the advantages of a small footprint, high pollutant removal efficiency, and considerably low sludge volume. However, the main limitations of the dewatering process are the high amount of energy required for operation and quick membrane fouling. In contrast to the conventional MBR, several researchers have shown that forward osmosis (FO) performs better in membrane fouling control (Achilli et al., 2009; Cornelissen et al., 2008; Lay et al., 2011). In FO, natural osmosis is the driving force for separation, and therefore FO is expected to (1) have low energy requirements for operation; (2) achieve a high rejection of many pollutants; and (3) exhibit less fouling than pressure-driven membrane processes (Cornelissen et al., 2008; Nguyen et al., 2013). These apparent benefits have recently attracted many researchers to explore the FO technique, and promising results have been reported in a variety of fields such as wastewater treatment (Holloway et al., 2014; Nguyen et al., 2015b; Nguyen et al., 2015c; Nguyen et al., 2016), sludge dewatering (Hau et al., 2014; Nguyen et al., 2013), food processing (Petrotos and Lazarides, 2001), and the concentration of algae (Buckwalter et al., 2013). However, the major challenge in creating a marketable FO technology is the lack of an ideal draw solution that can achieve a high water flux, low reverse salt flux, and easy recovery (Nguyen et al., 2015b).

Many different draw solutes have been studied over the past several decades. Achilli et al. (2010) used monovalent salts (e.g., NaCl, KCl, and NH_4Cl) as the draw solutes for specific FO applications. Although these monovalent salts could generate high osmotic pressure and be reconcentrated to high concentrations through the reverse osmosis process, the draw solution recovery involved high cost and high reverse salt flux. McGinnis and Elimelech (2007) observed that a thermolyte solution containing ammonia carbon could be employed as a novel draw solute in the FO process with a cost-effective solute recovery system, whereas the high reverse salt flux of NH_4HCO_3 was crucial. To overcome the reverse salt diffusion of monovalent salts, Tan and Ng (2010) investigated this issue using divalent salts ($MgSO_4$ and $CaCl_2$) as the draw solutes in FO process. Results showed that FO membrane could maintain at over 99.4% for all divalent salts, subsequently reduced replenishment cost.

The synthesis of magnetic nanoparticles (MNPs) could provide another approach for designing an innovative draw solute without salt leakage for the FO process. However, particle aggregation during draw solution recycling could decrease their osmotic pressure as well as water flux considerably (Bai et al., 2011; Ge et al., 2011; Ling et al., 2010). Thus, insufficient reuse of MNPs in the FO process is a major obstacle hindering the widespread use of MNPs. Very recently considerable progress has been made in developing synthetic materials for use as draw solutes, hydroacid complexes, polyelectrolytes, polymer hydrogels, and stimuli-responsive polymers (Ge et al., 2012a, 2014; Li et al., 2013; Zhao et al., 2014). Although the regeneration and reverse leakage of these innovative synthetic draw solutes have been improved, problems of poor repeatability, insufficient water flux, inability to generate high osmotic pressures at low viscosities, and a complex synthesis process persist [3]. In our previous study, high charge of EDTA was used as the draw solute in a hybrid FO–nanofiltration (NF) process for dewatering high-nutrient-containing sludge (Hau et al., 2014). Although EDTA sodium salt could obtain a low reverse salt flux, the viscosity of organic draw solutions (i.e. EDTA) increased quickly at high concentration, which prevent permeate water flux through FO membrane, and the recovery of the diluted draw solution by using an NF membrane was incomplete (salt rejection of 93%), which motivated the author to carry out this work.

Based on our research, this approach is the first to use high charge of phosphate as the draw solute in an FO–membrane distillation (FO–MD) hybrid system for concentrating high-nutrient sludge. The current work aims to evaluate feasibility of applying FO–MD system on sludge dewatering for conventional treatment to simultaneously improve the effluent water quality. In this study, the performance of Na_3PO_4 salt as the draw solute in an FO process was symmetrically investigated for concentrating high-nutrient sludge. First, the effect of the draw solution pH and concentration on the FO performance was evaluated using deionized (DI) water as the feed solution. Next, the FO performance in terms of sludge dewatering was examined using high-nutrient sludge as the feed solution. Finally, to determine the most appropriate membrane, the diluted Na_3PO_4 draw solution was recovered after FO tests by using four types of MD membranes.

2. Materials and methods

2.1. Feed and draw solutions

FO experiments were conducted using DI water and high-nutrient sludge as the feed solutions. Activated sludge was collected from the municipal wastewater treatment plant in Taipei, Taiwan, and the mixed liquor suspended solid (MLSS) concentration was 3500 mg/L. A solution containing D-glucose, NH_4Cl, and K_2HPO_4 was added to the raw activated sludge before its use as a feed solution to prepare a synthetic sludge with high nutrient concentration containing 300 ± 5 mg/L of dissolved organic carbon (DOC), 100 ± 2 mg/L of NH_4^+–N, and 100 ± 3 mg/L of PO_4^{3-}–P (Table 1). Different concentrations (0.05, 0.1, 0.2, 0.3, and 0.4 M) of the draw solution were prepared using laboratory-grade $Na_3PO_4 \cdot 12H_2O$ (Merck Co. Ltd., Germany). Because the formation of highly charged species of phosphate depends strongly on the pH, Na_3PO_4 solutions with different pH values (11, 10, 9, 8, 7, and 5) were obtained from the Na_3PO_4 solution with original pH 12 by using H_3PO_4 solution.

2.2. FO and MD membranes

Commercial thin film composite (TFC) FO membranes (OsMem™ TFC-ES Membrane 130424; HTI, USA) were used in all the FO experiments. Three types of polytetrafluoroethylene (PTFE) MD membranes with different pore sizes (0.1, 0.45, and 1 μm) were provided by Ray-E Creative Co., Ltd., Taiwan, and a polypropylene (PP) MD membrane was provided by Klean Filter, Inc., Taiwan. Characteristics of the FO

Table 1
Characteristics of synthetic high-nutrient sludge as feed solution.

MLSS (mg/L)	DOC (mg/L)	pH	NH_4^+–N (mg/L)	PO_4^{3-}–P (mg/L)
3500 ± 24	300 ± 5	7.4 ± 0.3	100 ± 2	100 ± 3
Na (mg/L)	Ca (mg/L)	Zn (mg/L)	Cu (mg/L)	Ni (mg/L)
126 ± 2	100 ± 3	3.48 ± 0.15	0.99 ± 0.06	0.18 ± 0.04