

# Application of forward osmosis (FO) under ultrasonication on sludge thickening of waste activated sludge

Nguyen Cong Nguyen, Hau Thi Nguyen, Shiao-Shing Chen, Nhat Thien Nguyen and Chi-Wang Li

## ABSTRACT

Forward osmosis (FO) is an emerging process for dewatering solid–liquid stream which has the potential to be innovative and sustainable. However, the applications have still been hindered by low water flux and membrane fouling when activated sludge is used as the feed solution due to bound water from microbial cells. Hence, a novel strategy was designed to increase sludge thickening and reduce membrane fouling in the FO process under ultrasonic condition. The results from the ultrasound/FO hybrid system showed that the sludge concentration reached up to 20,400 and 28,400 mg/L from initial sludge concentrations of 3000 and 8000 mg/L with frequency of 40 kHz after 22 hours, while the system without ultrasound had to spend 26 hours to achieve the same sludge concentration. This identifies that the presence of ultrasound strongly affected sludge structure as well as sludge thickening of the FO process. Furthermore, the ultrasound/FO hybrid system could achieve  $\text{NH}_4^+\text{-N}$  removal efficiency of 96%,  $\text{PO}_4^{3-}\text{-P}$  of 98% and dissolved organic carbon (DOC) of 99%. The overall performance demonstrates that the proposed ultrasound/FO system using seawater as a draw solution is promising for sludge thickening application.

**Key words** | draw solution, forward osmosis, seawater, sludge thickening, ultrasonication

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## INTRODUCTION

Biological treatment process generates large amount of sludge which includes a lot of water, microorganisms and extracellular polymeric substances (EPS). Therefore, the sludge dewatering process is necessary and accounts for approximately 50–60% of the total operating cost of the whole water treatment plant (Rai *et al.* 2004; Appels *et al.* 2008). Basically, activated sludge usually has a poor dewaterability and chemicals are added to sludge, such as alum, iron (III) chloride, and polyelectrolyte to improve dewaterability. Besides, physicochemical processes have been investigated such as electrolysis technology (Yuan *et al.* 2011), microwave, alkali pretreatment (Chang *et al.* 2011) and integrating chemical conditions with filter press (Zhai *et al.* 2012). Although these methods have high dewatering potentials, their main limitations are high operation cost and secondary environmental pollution. Meanwhile, forward osmosis (FO) is known as an excellent technology for sludge dewatering with low fouling and minimum operating cost (Cath *et al.* 2006; Holloway *et al.* 2007; Cornelissen *et al.* 2008; Mi & Elimelech 2010; Hau *et al.* 2014). Typically, FO is a process

of water driven by osmotic pressure difference across the semi-permeable membrane without the aid of external energy. Theoretical water flux ( $J_w$ ) across the FO membrane is the product of the membrane permeability coefficient ( $A_w$ ) and the osmotic pressure driving force ( $p$ ):

$$J_w = A_w (p_{\text{Draw solution}} - p_{\text{Feed solution}}) \quad (1)$$

More recently, Zhu *et al.* (2012) used FO membrane on dewatering of activated sludge. After 19 days of operation, the mixed liquor suspended solids (MLSS) concentration reached 39 g/L from an initial amount of 7 g/L, indicating a good thickening efficiency. In addition, our previous works (Nguyen *et al.* 2013) investigated dewaterability of sludge with various biomass loadings by FO performance. However, dewatering time of sludge is still an important key, and this may be only solved when the bound water from microbial cells is released into aqueous phase to enhance dewatering of FO membrane. Ideally, the ultrasonic technique is an advanced method to achieve a better

dewaterability and decrease sludge moisture content. Previous investigations indicated that long ultrasonication time, low-frequency ultrasound (20–40 kHz) and high ultrasonic intensity (0.44 W/ml) are more effective for sludge disintegration and dewatering (Dewil *et al.* 2006; Huan *et al.* 2009). Therefore, the combination of ultrasonication and FO might be feasible for reducing sludge thickening time.

However, up to now, no research has studied the ultrasound/FO hybrid system using seawater as draw solution on sludge thickening. This study focused on evaluating feasibility of applying ultrasound/FO hybrid system to improve dewaterability of sludge and the critical objectives are to compare FO only and ultrasound/FO for the following: (1) effect of operational conditions on water flux; (2) rejection of nutrient and organic compounds; (3) FO membrane fouling during the experiment; and (4) the proposed technology fitting on conventional biological treatment.

## MATERIALS AND METHODS

### Materials

The flat-sheet cellulose triacetate (CTA) FO membranes used in this study were supplied by Hydration Technology Innovations (HTIs OsMem™ CTA Membrane, Albany, OR, USA) with size of 15 × 22 cm for each piece, and membrane orientation of active layer facing the feed solution was used. The FO membrane thickness was approximately 50 μm and the membrane was recorded to be negatively charged in typical feed waters (Cath *et al.* 2006; Achilli *et al.* 2010; Lutchmiah *et al.* 2014). The raw waste sludge samples were collected from secondary sedimentation in the New Taipei City wastewater treatment plant, where its duty is to treat domestic and industrial wastewater for New Taipei City in Taiwan. Glucose, NH<sub>4</sub>Cl and K<sub>2</sub>HPO<sub>4</sub> solution were then added to produce synthetic sludge with NH<sub>4</sub><sup>+</sup>-N of 100 mg/L, PO<sub>4</sub><sup>3-</sup>-P of 100 mg/L, dissolved organic carbon (DOC) of 200 mg/L and various MLSS concentrations of 3000 and 8000 mg/L, as presented in Table 1. Besides, draw solution was prepared by dissolving

NaCl in the deionized (DI) water to achieve concentration of NaCl of 36 ± 1 g/L, a typical concentration of seawater.

### Experimental setup

The FO experiment setups for sludge thickening are shown in Figure 1. The flat sheet was rolled as a tube configuration with total effective membrane surface area of 106 cm<sup>2</sup> placed into the reactor tank with a total volume of 3.0 L. In addition, an air supply system was used to simultaneously supply the dissolved oxygen concentration of 2 mg/L in reactor and prevent membrane fouling. The peristaltic pumps (Master Flux L/S Drive, Model 7518-00) were used to circulate the draw solution flow. High salt concentration (NaCl 20%) was added into draw solution tank (5 L) every two hours to keep concentration of the draw solution always stable at 36 ± 1 mg/L. Water flux,  $J_w$  (L/m<sup>2</sup> h) through the membrane was determined by measuring the weight changes of the draw solution container every two hours using the scale based on Equation (2).

$$J_w = \frac{m \cdot 10}{d \cdot A t} \quad (2)$$

where  $m$  is total increase mass of the draw solution container (g),  $d$  is the density of the draw solution (g/mL),  $A$  is the effective FO membrane area (cm<sup>2</sup>) and  $t$  is the time (h).

The ultrasonic equipment consisted of a generator, a converter and a sonotrode, supplied by Alpha Omega Ultrasonics Corporation, USA. The frequency of ultrasound is set at 40 kHz and turned on five minutes per eight hours of sludge thickening time in FO. The reverse solute flux,  $J_s$  (g/m<sup>2</sup> h) from the draw solution side into the feed solution side was calculated based on the conductivity change over time of the feed solution according to Equation (3).

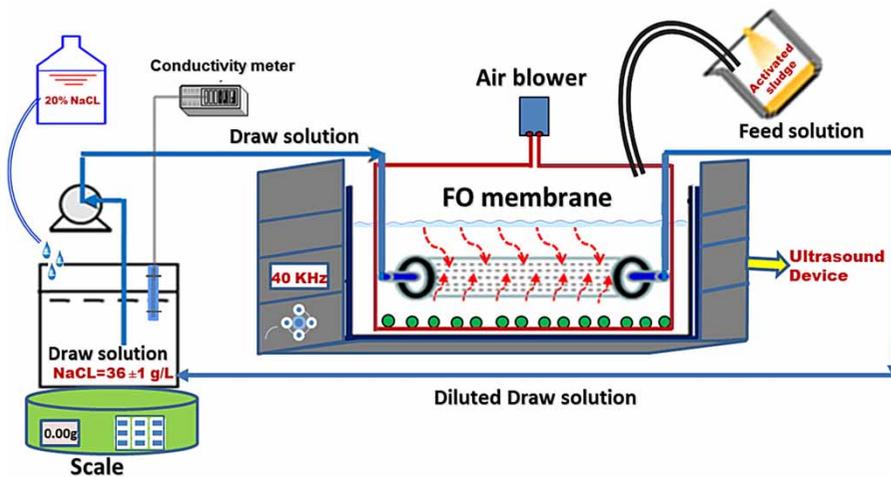
$$J_s = \frac{V_t \cdot C_t - V_0 \cdot C_0}{A \cdot t} \quad (3)$$

where  $C_t$  and  $V_t$  are the concentration (g/L) and volume (L) of the feed solution measured at time  $t$  and  $C_0$  and  $V_0$  are the initial concentration (g/L) and volume (L) of feed solution.

**Table 1** | Initial sludge properties with various mixed liquor suspended solids concentrations

Initial MLSS concentration (mg/L)	Osmolality (mOsm/kg)	TDS (mg/L)	EC (μs/cm)	Initial concentration (mg/L)		
				NH <sub>4</sub> -N	PO <sub>4</sub> <sup>3-</sup> -P	DOC
3000 ± 28	11 ± 1	328 ± 3	657 ± 5	100 ± 2	100 ± 1	200 ± 3
8000 ± 52	18 ± 2	575 ± 3	1018 ± 7	100 ± 1	100 ± 1	200 ± 3

TDS: total dissolved solids; EC: electrical conductivity.



**Figure 1** | Schematic illustration of the sludge thickening ultrasound/FO system.

Following the time, sludge samples in the reactor tank are collected to measure MLSS concentration and conductivity, while DOC,  $\text{NH}_4^+\text{-N}$  and  $\text{PO}_4^{5-}\text{-P}$  were measured in the diluted draw solution.

### Analytical methods

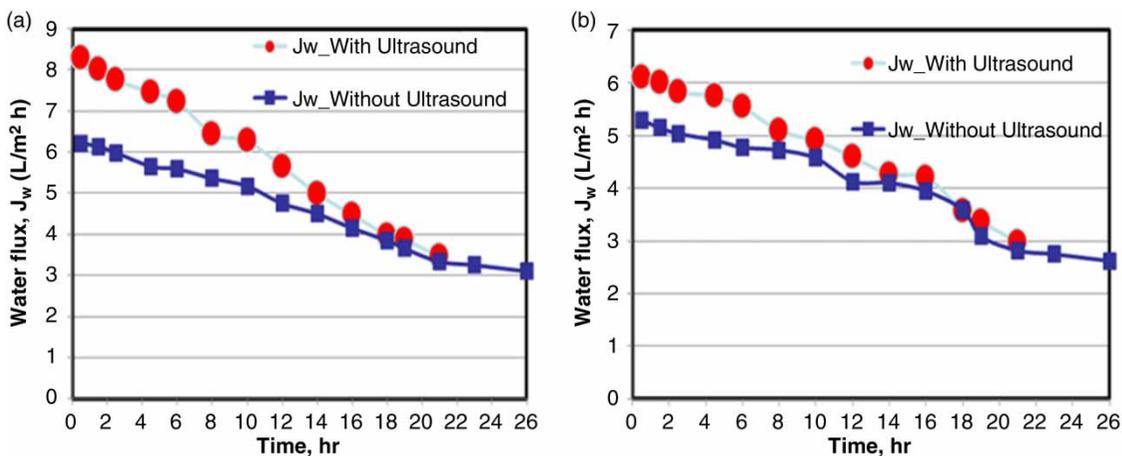
The concentration of  $\text{PO}_4^{5-}\text{-P}$  and  $\text{NH}_4^+\text{-N}$  were analyzed by a UV-Vis spectrophotometer (HACH Model DR-4000, Japan). Total phosphorus (TP) was measured using methods NIEA W437.51C according to Taiwanese Environmental Protection Agency standard methods. DOC samples were prepared by filter paper with pore size of  $0.45\ \mu\text{m}$  then measured by Aurora 1010C TOC Analyzer was purchased from O.I. Analytical Corporation in USA. The concentrations of MLSS was analyzed using methods 2540 D described in

*Standard Methods for the Examination of Water and Wastewater* (APHA 2005). The conductivity value was monitored by conductivity meter (Sension156, Hach, China). The osmolality of solutions was measured using an osmometer (Model 3320, Advanced Instruments, Inc., USA).

## RESULTS AND DISCUSSION

### Variation in water fluxes and sludge concentrations

Figures 2(a) and 2(b) show that the water flux was reduced with ultrasound (average water flux decreased from  $5.99$  to  $4.71\ \text{L/m}^2\ \text{h}$ ) and without ultrasound (average water flux decreased from  $4.68$  to  $4.10\ \text{L/m}^2\ \text{h}$ ) as initial sludge concentration was increased from  $3000$  to  $8000\ \text{mg/L}$ . The



**Figure 2** | Variations in water fluxes versus operating time: (a) initial sludge concentration of  $3000\ \text{mg/L}$  and (b) initial sludge concentration of  $8000\ \text{mg/L}$  (feed solution: activated sludge; draw solution:  $\text{NaCl}\ 36 \pm 1\ \text{mg/L}$ ; membrane orientation: active layer facing the feed solution and flow rate of draw solution:  $280\ \text{mL/min}$ ).

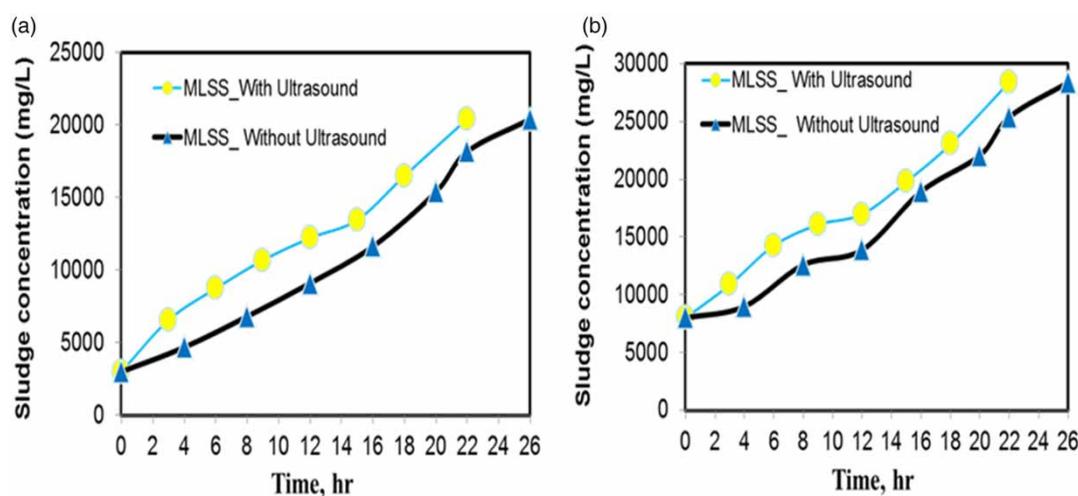
reason is due to the effect of membrane fouling and lower osmotic pressure gradient. Clearly, the higher sludge concentration was used, the higher fouling and lower osmotic pressure gradient were recorded according to Equation (1). In fact, other researchers also have found that lower sludge concentration shows better permeability than that of higher sludge concentration (Zhang et al. 2012). Moreover, when ultrasound device was applied, water flux was higher than that without ultrasound. This phenomenon can be attributed to the occurrence of ultrasonic disruption of sludge since ultrasonic irradiation has a remarkable effect on sludge solubilization. Besides, the ultrasonic process leads to floc size reduction, which permits the release of organic substances and bound water into the liquid phase. Moreover, ultrasound was turned on five minutes per eight hours of sludge which also reduced membrane fouling. As the experiment progressed, the water flux was decreased quickly versus time for ultrasound and without ultrasound due to two main reasons: (i) deposition of sludge cake layer on the FO membrane surface resulting in inhibited water transport through membrane; and (ii) transportation

of reverse salt into the feed solution leading to reduced osmotic pressure gradient.

Figures 3(a) and 3(b) show that the MLSS concentration had similar increasing trends during the thickening process. The results recorded that the final MLSS concentration reached 20,400 and 28,400 mg/L after 26 hours without ultrasound from initial sludge concentration of 3000 and 8000 mg/L, respectively. However, when ultrasound was used for the experiment, the same concentration sludge was achieved after 22 hours, indicating sludge dewaterability was increased significantly.

### Nutrient and organic removal efficiency on sludge thickening in FO

Notably, an excellent advantage of FO membrane is the high rejection of nutrient and organic compounds to achieve superior effluent water quality. Rejections were calculated using initial concentrations in the feed solution and final concentrations in the diluted draw solution. The results from Table 2 indicate that removal efficiency of nutrient and

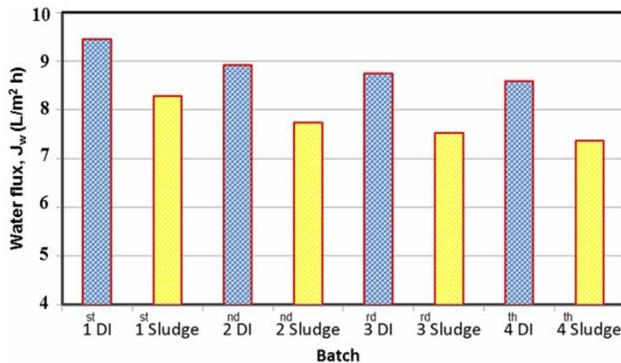


**Figure 3** | Variations in sludge concentrations versus operating time: (a) initial sludge concentration of 3000 mg/L and (b) initial sludge concentration of 8000 mg/L (feed solution: activated sludge; draw solution: NaCl 36 ± 1 mg/L; membrane orientation: active layer facing the feed solution and flow rate of draw solution: 280 mL/min).

**Table 2** |  $\text{NH}_4^+\text{-N}$ ,  $\text{PO}_4^{3-}\text{-P}$ , TP and DOC rejections in FO experiments with ultrasound and without ultrasound

Operating condition	Initial sludge concentration (mg/L)	Rejection (%)			
		$\text{NH}_4^+\text{-N}$	$\text{PO}_4^{3-}\text{-P}$	TP	DOC
Without ultrasound	3000 ± 28	96.36	98.07	98.12	99.06
	8000 ± 52	96.92	98.32	98.36	99.21
With ultrasound	3000 ± 28	96.06	98.03	98.11	98.94
	8000 ± 52	96.32	98.12	98.25	99.13

organic compounds were higher than 96% in all experiments. The removal efficiency of nutrients and organic compounds depended on the sludge properties, especially sludge concentration. Apparently, as MLSS concentrations of the feed solution were increased from 3000 to 8000 mg/L, rejection of both nutrient and DOC was increased for both experiments. One of the possible reasons was that the increasing MLSS concentrations in feed solution contributed to form thicker cake layer of membrane fouling, which enhanced nutrient and organic compound removal efficiency.

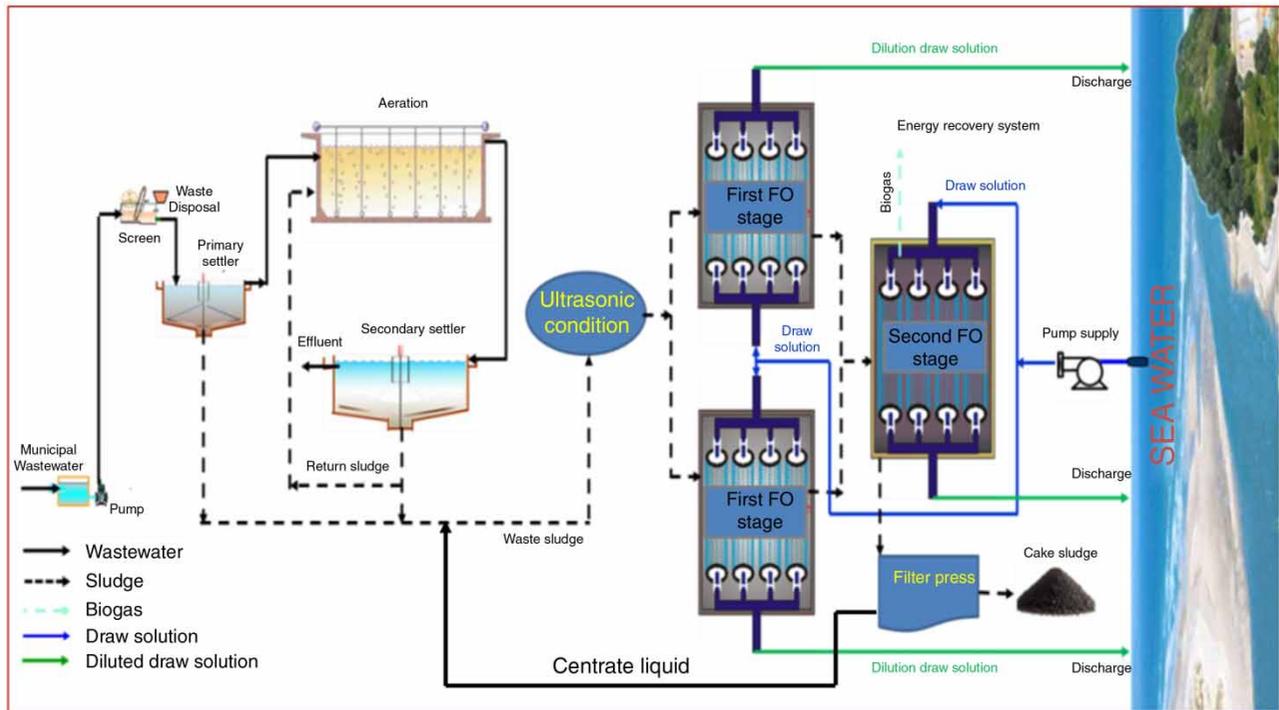


**Figure 4** | Water flux as a function of the number of FO membrane operations during experiment batch (feed solution: activated sludge of MLSS = 3000 mg/L and DI water; draw solution: NaCl  $36 \pm 1$  mg/L; frequency: 40 kHz and flow rate of draw solution: 280 mL/min).

In all FO tests, DOC removal efficiencies were achieved up to 99%, which is higher than those obtained in conventional membrane bioreactors (MBRs) where removals up to 95% are typical (Achilli *et al.* 2009). More specially, in almost of the FO experiments,  $\text{PO}_4^{3-}\text{-P}$  removal efficiency (about 98%) was higher than  $\text{NH}_4^+\text{-N}$  (96%) due to both effects of hydrated radius and charged repulse. According to Kiriukhin & Collins (2002) and Lide & Frederikse (1995), the hydrated radius of ammonium was 0.104 nm with a diffusion coefficient in water of  $1.96 \times 10^{-9} \text{ m}^2/\text{s}$  whereas the hydrated radius of phosphate was 0.339 nm with a diffusion coefficient in water of  $0.44 \times 10^{-9} \text{ m}^2/\text{s}$ . Obviously, larger hydrated radius and lower diffusion coefficient could have a higher removal efficiency. Moreover, as pH of activated sludge always remained at 7.2 and the FO CTA membrane is negatively charged at  $\text{pH} > 5$  (Xie *et al.* 2012),  $\text{NH}_4^+\text{-N}$  ions were easy to attach on the surface of the membrane and into water flux by electrostatic attractions, whereas the CTA membrane rejected  $\text{PO}_4^{3-}\text{-P}$  ions by electrostatic repulsions.

#### FO membrane fouling experiments

After every experiment, membrane was washed in 10 minutes with DI water and it was reused to check the effect of



**Figure 5** | The proposed technology fitting on traditional wastewater treatment plant.

membrane fouling and concentration polarization on water flux. Figure 4 shows that the water flux as a function of the number of FO membrane operation using NaCl  $36 \pm 1$  mg/L as draw solution and activated sludge and DI water as feed solution under conditions of ultrasound. The result shows that water flux decreased around 4.0% and 4.5% for DI water and activated sludge after the second batch since a thin cake layer of sludge attached on FO membrane surface and NaCl may also attach in the support layer of membrane caused water flux decline. However, from the third and fourth batch, the decrease in water flux was not significant because these layers of membrane fouling were removed by washing with DI water.

### The proposed technology fitting on traditional wastewater treatment plant

The proposed FO thickening technology can be fitted into conventional biological treatment processes, as illustrated in Figure 5, where two stage FO systems are designed to incorporate with the traditional system. Firstly, FO membrane is designed to replace thickening process of traditional wastewater treatment plant as presented in stage I, which significantly improves the final effluent with  $\text{NH}_4^+$ -N removal efficiency of 96%,  $\text{PO}_4^{3-}$ -P removal efficiency of 98% and DOC removal efficiency of 99%. Secondly, the high biomass concentration from first FO stage is continuously dewatered by stage II of anaerobic FO to produce biogas for energy recovery system. For clarification, a significant sludge thickening performance was achieved when seawater was used as the draw solution and this design appears to be potentially inexpensive for wastewater treatment plant built near coastal areas but easily discharge diluted draw solution back to the sea.

### CONCLUSIONS

This study investigated the effect of ultrasound on sludge thickening in FO performance system. The water flux from ultrasound/FO hybrid system was always higher than that without ultrasound, indicating that bound water from microbial cells was released into liquid phase due to wave fluctuation of ultrasound to enhance thickening in FO. The results from the ultrasound/FO hybrid system showed that sludge concentration reached 20,400 and 28,400 mg/L from initial sludge concentration of 3000 and 8000 mg/L with frequency of 40 kHz after 22 hours, while the system

without ultrasound had to spend 26 hours to achieve the same sludge concentration.

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