CHAPTER 14

Forward osmosis for nutrients recovery from wastewater

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14.1 Introduction

In the 21st century, the sustainable food supply has become one of the biggest global challenges, especially given the fast growth of global population and the requirement to balance the food production and agricultural impacts on the environment [1]. To satisfy the increasing food demand, the fertilizer production should be kept at an annually increased value of 4% [2]. The chemical fertilizers used in agriculture mainly consist of phosphate and ammonium. The current technologies to produce fertilizers need the non-renewable energy and finite mineral resources. Specifically, the supply of phosphate replies on the minable phosphate-bearing rocks, but the reserves of phosphate resources are estimated to be completely consumed in the coming few decades [2]. Moreover, the mining process may exert some serious impacts on the environment, such as the production (and inevitable release) of some by-products with rich heavy metals and radioactive elements [3]. On the other hand, the Haber-Bosch process (Eq. 14.1) is used to produce ammonia for fertilizer production [4, 5].

$$N_2 + 3H_2 \rightarrow 2NH_3 \qquad (14.1)$$

In the process, the nitrogen gas is obtained via fractional distillation of liquefied air while a steam reforming process results in the generation of hydrogen gas [4, 6]. The process requires high pressure and temperature with additional iron catalyst, in which substantial energy input is needed at around 35−50 MJ/kg·N [7]. Desloover et al. [8] reported that approximately 2% of the global power is used in producing

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ammonia at an industrial scale. Apart from this, the process also generates about 3.5 tonnes of carbon dioxide (CO_2)/kg·N. As a result of this, the global warm is aggravated while other gases such as nitrous oxide also contribute to the greenhouse effects [9, 10].

Although the direct land application of fertilizers can increase the productivity of agriculture, this also leads to the accumulation of nutrients (i.e., ammonium and phosphate) in the aquatic environment [11]. Rahman et al. [12] reported that 30% of nitrogen and 16% of phosphorus in fertilizers end up in wastewater. This will inevitably cause eutrophication in the aquatic environment [13-16]. Currently, traditional nutrient removal technologies include chemical phosphorus removal, nitrification-denitrification and aerobic activated sludge. However, removing nutrients in wastewater treatment requires a substantial amount of energy and chemical input whist the biological nutrient removal is subject to the reliability and stability of bacterial [5, 14]. For instance, 45 MJ/kg·N is needed while processing the nitrogen removal [17]. Furthermore, the gaseous nitrogen serving as the end-product of this process does not have any market value or contribution to the production of fertilizers. While removing nutrients from each liter of wastewater, 0.9 kg CO_2 is generated, which enhances the greenhouse effects [18]. In recent years, the removal of nutrients in wastewater treatment has to satisfy the increasingly strict controls of nutrients discharge standards [19, 20]. Overall, the disadvantages of nutrient removal such as recyclable resource-wasting, carbon emission, long process, instability treatment effect, and high energy consumption make nutrient removal from different sources unsustainable [21], particularly given the fact that wastewater has recently considered as a renewable resource [22, 23].

Recently, recovering nutrients from wastewater has attracted considerable attention because this option could supplement nutrients for fertilizer production to ensure food safety   [17] while reduces the uncertainties associated with minable phosphorus supply and high costs involved in producing ammonia used for fertilizer production [24, 25]. In addition, nutrient recovery could also bring benefits to wastewater treatment plants, such as a reduction in overall costs and environmental footprint, and struvite blockage prevention [26].

Currently, chemical precipitation is widely utilized to recover nutrients from wastewater, where struvite ($MgNH_4PO_4 \cdot 6H_2O$) and hydroxyapatite ($Ca_5(OH)(PO_4)_3$) are the most common products. The formation of such precipitates always needs the extra introduction of magnesium salts or calcium salts in wastewater treatment. Besides, high pH is needed for the chemical precipitation, which is achieved by additional alkaline chemicals or aeration stripping of CO_2. The chemical precipitation for the nutrient recovery has been widely investigated in a wide range of wastewater sources, such as domestic wastewater, anaerobically digested sludge and urine. Currently, the nutrient recovery via struvite precipitation has been implemented in several full-scale wastewater treatment plants in North America [27].

Nevertheless, the low concentration of nutrients in wastewater and the presence of foreign substances challenges the commercial implementation of chemical precipitation for recovering nutrients from wastewater. Specifically, the chemical precipitin's driving force and kinetics are subject to the concentrations of

phosphate and ammonium. Besides, Xie et al. [17] reported that the high phosphate concentration (> 100 mg·P/L) in the aquatic environment could achieve effective struvite precipitation. The possible reason for this is that low content of nutrients can prolong the precipitation reaction time and reduce the recovery efficiency, which decreases the economic feasibility of the nutrient recovery system. More importantly, it should be noted here that the typical concentrations of ammonium and phosphate in some real wastewater sources are indeed low, such as domestic wastewater. Consequently, it is necessary to concentrate nutrients in wastewater prior to conducting the chemical precipitation, which can significantly enhance the chemical precipitation potential and efficiency. On the other hand, the presence of toxic heavy metals as well as the emerging organic contaminants is another challenge for the nutrient recovery via chemical precipitation since the foreign matters compromise the purity and quality of the recovered product, and further commercial application of such products in agriculture. Several studies have detected 570 mg/kg of arsenic accumulated in the recovered struvite crystals [28, 29]. This finding may result in the application of such struvite being forbidden in direct land application for crops and plants.

As discussed above, it is needed to enrich nutrients and simultaneously separate foreign substances from the nutrients. Due to its high rejection rate and relatively low input of costs and energy, membrane technology is encouraged to couple with chemical precipitation for recovering nutrients from wastewater [30]. Among different membrane separation processes such as membrane distillation (MD), reverse osmosis (RO) and electrodialysis (ED), forward osmosis (FO) membrane processes seem to be the most promising ones [31, 32]. This is because the FO membrane has lower membrane fouling potential, higher fouling reversibility and smaller energy input compared to other membrane technologies [33, 34]. As a result of this, the FO process can be applied to recover nutrients in a wide range of wastewater sources, even in some complicated conditions. Apart from this, the FO process can be integrated with current wastewater treatment facilities.

In recent years, the FO process has been largely explored to enrich nutrients from a range of different feed solutions (e.g., domestic wastewater, urine, digested sludge and secondary treated effluent) at different operation conditions [17, 35-41]. It was reported that more than ten-fold concentrations of nutrients could be concentrated by the FO membrane in the feed side [38], which facilitates the further recovery of nutrients. Also, the integration of the FO-based system with chemical precipitation could improve the technical and economic feasibility of the nutrient recovery system, making the recovery system more accessible in real applications.

In this chapter, the nutrient recovery through FO-based technologies from wastewater is critically reviewed. Section 14.2 elaborates the FO membrane technology, including its principle and popular configurations. Section 14.3 introduces the recovery of ammonium and phosphate by the FO-based systems in wastewater treatment. This section involves the mechanisms of the recovery system and discusses the effects of influencing factors on this system. Section 14.4 discusses the key challenges associated with the FO-based systems for the nutrients recovery. Sections 14.5 delineates the possible future roadmap for the nutrients recovery using FO-based processes from wastewater. Sections 14.6 summarizes the

important insight into the FO-based systems for the nutrients recovery from wastewater and further development of the recovery systems.

14.2   Forward osmosis (FO) technology

In 1976, Moody and Kessler [42] firstly proposed the concept of FO, which has now developed into an energy-efficient and sustainable membrane technology. In the FO system, a semipermeable membrane is placed between the feed solution and draw solution. The difference in osmotic pressure between the feed side and draw side offers the driving force for the mass transport, in which the transfer across the FO membrane from a more dilute feed solution with lower osmotic pressure to a concentrated draw solution with higher osmotic pressure is achieved. As a result of this, water, solvents or other targeted substances could be extracted from the feed solution [43]. It could be seen that this osmotically-driven separation process does not need additional energy input and presents lower fouling propensity and higher fouling reversibility, which is attributed to the absence of hydraulic pressure. Nevertheless, the FO process cannot be used as a single unit in wastewater treatment because it does not have the ability to remove any contaminants. In this scenario, coupling the FO process with other technologies is common in wastewater treatment [44-47].

Currently, there are two main configurations of the FO process in wastewater treatment, namely the aerobic and anaerobic FO-based systems (Fig. 14.1). For the aerobic FO-based systems, the FO process always integrates with the bioreactor to form the aerobic osmotic membrane bioreactor (OMBR) [35, 36, 48-50] (Fig. 14.1a). On the other hand, the combination of FO technology and anaerobic bioreactor are also explored [51, 52] (Fig. 14.1b). In this case, the anaerobic OMBR (AnOMBR) is formed while directly adding anaerobic sludge into the FO reactor; alternatively, the FO system can receive the anaerobically digested effluent for further treatment. In these two configurations, a submerged FO module is utilized since the submerged configuration is beneficial for the concentration in wastewater treatment [53]. Besides, aeration supply is required in the aerobic FO-based system whilst the anaerobic FO-based system could produce the biogas which has significant potential as an energy resource [51, 52].

The treatment performance of the FO-based systems depends on various factors, including membrane materials, operation conditions and the type of applied biological treatment [54]. Since such FO configurations could retain the pollutants such as microbes, colloidal particles and organics in the feed solution, it is possible for the FO-based systems to obtain high removal efficiency of a wide range of contaminants [54]. Besides, the reserve salt flux from the draw side to the feed side may negatively affect the feed solution.

 # Insert Fig. 14.1 #

14.3   FO systems for nutrient recovery

Table 14.1 summarizes some FO-based systems for nutrients recovery. In the FO-based systems, ammonium and phosphate in the feed solution (wastewater)

can be enriched due to the superior rejection capability of FO membrane, which provides favorable conditions for recovering ammonium and phosphate (Fig. 14.2). Besides, the rejection of other metals such as magnesium and calcium ions is also beneficial for the nutrients recovery. Specifically, the feed solution containing rich nutrients improves the kinetics of chemical precipitation and reduces the number of chemicals (e.g., magnesium and calcium salts) needed for the precipitation. Apart from this, the reverse solute flux could enrich the content of salts (e.g., Mg^{2+} and Ca^{2+} ions) for the chemical precipitation while employing Mg- and/or Ca-based salts as the draw solute [39, 40].

 # Insert Fig. 14.2 #

As shown in Fig. 14.2, the draw solution will be gradually diluted due to the water transfer from the feed solution, which may reduce the osmotic pressure and thereby impair the FO performance. Simultaneously, the reverse salt flux may also result in the migration of partial draw solute from the draw side to the feed side [17]. Consequently, a new draw solute is needed; alternatively, reconcentration of the draw solution is needed to ensure enough osmotic pressure between the feed side and draw side. Some researchers proposed pressure-driven membrane processes through the RO membrane or thermally-driven membrane processes via the MD membrane to extract fresh water from the draw solute for re-enriching the draw solute [54, 61]. In this scenario, freshwater with high quality could also be produced [39, 62]. As an example, Nguyen et al. [63] used an FO-RO system to obtain 99.9% of phosphate and 92% of ammonium which were rejected in the feed side of the FO process. Hancock et al. [64] examined this concept at a pilot scale, in which > 99% of phosphate was enriched in the feed solution and the quality of water permeated from the RO could reach the primary drinking water standards of the Environmental Protection Agency (EPA). It should be noted here that the FO-MD system could use solar energy for heating the MD process [65], which enhances the economic feasibility of the FO-based system in terms of recovering nutrients and clean water [39, 62]. Moreover, seawater can also be used as the draw solution, especially while conducting the FO process near a coastal area. There are two evident advantages: 1) seawater does not need to be regenerated while acting as the draw solute and the diluted seawater can be directly discharged to the sea; and 2) using the seawater as the draw solute could reduce the costs in terms of raw materials and transport.

Table 14.1. Summary of FO-based systems for the nutrients recovery

FO-based system	Wastewater resource	Draw solution	Recovered product	Performance	Reference
FO-MD	Digested sludge centrate	$MgCl_2$	Struvite	> 97% of PO_4^{3-}-P removed; > 90% of NH_4^+-N removed	Xie et al. [39]
OMBR-MF	Municipal wastewater	Seawater brine	Calcium phosphate	> 90% of PO_4^{3-}-P recovered; 99% of NH_4^+-N removed	Qiu et al. [37]
FO	Digested sludge centrate	Seawater	Calcium phosphate	90% of PO_4^{3-}-P recovered	Ansari et al. [40]
OMBR	Municipal wastewater	$MgCl_2$ & NaCl	Calcium phosphate	> 95% of PO_4^{3-}-P recovered; 98% of NH_4^+-N removed	Qiu and Ting [36]
OMBR-biofilm	Municipal wastewater	NaCl	Calcium phosphate and magnesium phosphate	63.8–79.6% of PO_4^{3-}-P recovered; 99% of NH_4^+-N removed	Qiu et al. [55]
OMBR-MF-RO	Raw sewage	NaCl	Calcium phosphate	recovered precipitate containing 15–20% (wt/wt) of phosphorus	Luo et al. [35]
OMBR-UF	Domestic wastewater	NaCl	N/A	> 99% of PO_4^{3-}-P removed; 82% of NH_4^+-N removed	Holloway et al. [56]
Anaerobic FO	Digested swine wastewater	$MgCl_2$	struvite	> 99% of PO_4^{3-}-P recovered; > 93% of NH_4^+-N removed	Wu et al. [57]
AnOMBR-MRC	Synthetic wastewater	NaCl	Calcium phosphate and/or struvite	65% of PO_4^{3-}-P removed; 45% of NH_4^+-N recovered	Hou et al. [58]
FO-MEC	Synthetic sidestream centrate	NH_4HCO_3	Struvite & NH_4HCO_3	> 79% of PO_4^{3-}-P removed; > 99% of NH_4^+-N recovered	Zou et al. [59]
FO-MFC	Municipal wastewater	Distilled water	struvite	> 80% of PO_4^{3-}-P removed; > 98% of NH_4^+-N removed	Ye et al. [60]

Besides of causing the loss of the draw solute, the reverse draw solute flux may also negatively influence the quality of feed solution [66, 67]. However, it will be beneficial for recovering nutrients if the Mg- and/or Ca-based salts are utilized as the draw solution, which could supplement Mg^{2+}/Ca^{2+} ions for the nutrient recovery via chemical precipitation [39]. Through the analysis of scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS), however, Wu et al. [57] found that the struvite formed in the FO system was coated by $MgCl_2$ while using the $MgCl_2$ as the draw solute. As a result of this, the purity of struvite was deteriorated. In addition, some contaminants may migrate across the FO membrane to the draw solution, and then accumulate in the side in the closed-loop FO-based system [68]. As an example, Coday et al. [69] found that in an FO-RO system, organic foulants were enriched in the draw solution. Similar findings have also been reported in other FO-based systems [39, 62]. The increase in the cumulative permeate volume may result in higher micropollutant concentrations, which exerts detrimental effect on the FO-based system. Thus, it is of great significance to control the accumulation of contaminants to ensure system performance and reliability.

Ansari et al. [54] compared the current wastewater treatment process with FO-based wastewater treatment technologies. They believed that the current conventional wastewater treatment process requires substantial energy supply with reference to the aeration and pressurized membrane systems (Fig. 14.3a). Besides, water recovery is the main objective of such a process, but the recovery of chemical energy and nutrients is ignored and wasted [70]. Therefore, Ansari et al. [54] proposed an FO-based system for the recovery of nutrients, clean water and chemical energy stored in the organics (Fig. 14.3b). Specifically, the primarily treated effluent is treated by the FO-MD hybrid system to produce clean and freshwater while applying organic ionic solution as the draw solutes. As a result of this, the effects of reverse draw solute flux could be reduced as well as the probability of the methane inhibition during anaerobic digestion. Subsequently, the FO pre-concentrate is processed with anaerobic digestion for the biogas production, in which the biogas is used for heating the MD process and converting into electricity for the treatment operations. Apart from this, anaerobic effluent containing rich nutrients are treated by an FO-MO hybrid system. As discussed above, the nutrient could be recovered by chemical precipitation in such a system [39] whilst clean water could be produced through the MD system.

 # Insert Fig. 14.3 #

In addition, the FO technology has always been integrated with bioreactor in the current wastewater treatment plant. Qiu and Ting [36] proposed an OMBR-based system in the sewage treatment (Fig. 14.4), in which > 90% of nutrients could be directly recovered by chemical precipitation with additional alkaline chemicals. As discussed above, ammonium and phosphate ions were enriched in the feed side of the bioreactor as well as mineral salts (i.e., magnesium and calcium ions). Consequently, it is evident that additional mineral salts may not be needed for recovering nutrients by chemical precipitation. Furthermore, the researchers utilized $MgCl_2$ as the draw solute as the transport of Mg^{2+} ions caused by reverse draw solute flux could offer more Mg^{2+} ions for the struvite formation. The selection

and enrichment of polyphosphate-accumulating organisms (PAOs) was not carried out in this study, which aims to prevent the phosphate being converted and adsorbed by the activate sludge. This could both reduce the production of surplus sludge and loss of phosphate consumed by the biological process. Due to the removal of nutrients and mineral salts in the OMBR-based system, it is possible to achieve a moderate level of the system's salinity, improving the membrane performance. Overall, most nutrients in the feed solution can be recovered by chemical precipitation despite partial nutrients consumed through biological uptake.

Insert Fig. 14.4

To increase the purity of recovered nutrients, microfiltration (MF) membrane was installed into the feed side in the OMBR-based system [37] (Fig. 14.5). In this case, the nutrients and mineral salts were retained by the FO membrane in the feed side, and then extracted by the MF membrane to the draw side. The MF extraction can not only recover phosphate, but also control the salt accumulation in the feed solution. Through the addition of sodium hydroxide, the calcium phosphate precipitates were formed, realizing the objective of nutrient recovery. In this study, Qiu et al. [37] found the phosphate concentration in the feed side could reach to 70 mg/L through the FO retention effect, which results in a 98% phosphate recovery. However, it may be a challenge while applying the OMBR-MF system to wastewater containing high concentrations of nutrients and mineral salts. The possible reason for this is that the risk of spontaneous precipitation in form of struvite, magnesium phosphate precipices, calcium phosphate precipitates or other precipitates still exists, which negatively affects the performance of both the FO membrane and MF membrane. Besides, this also exerts detrimental impacts on recovering nutrients due to the reduction in the concentration of nutrients and mineral salts. Qiu et al. [37] believed that a higher flow rate of the MF membrane may be an effective strategy. In contrast, Luo et al. [35] claimed that a low permeate flux or the periodic extraction mode of the MF membrane would be more effective for the system because this scenario could reduce the energy input and fouling potential of the MF membrane, and thus, increases this system's economic feasibility.

Insert Fig. 14.5

Based on the OMBR-MF system, Qiu et al. [55] developed a new OMBR system coupling with a fixed bed biofilm (OMBR-BF), in which the fixed bed biofilm replaced the MF membrane. Qiu et al. [55] indicated that there is no extraction of side-stream for the solid/liquid separation under this mode, which means the biomass from the FO membrane could be quarantined. Besides, the system could effectively and continuously remove the suspended growth in the absence of the MF membrane. Consequently, the membrane fouling of the FO membrane significantly decreased due to substantially reduced bacteria deposition and colonization. Similarly, Holloway et al. [56] utilized ultrafiltration (UF) membrane to replace the role of the MF membrane in the OMBR-MF system. Up to 50 mg/L of phosphate could be enriched in the UF permeate, which improves the economic feasibility of the phosphate recovery. Furthermore, an RO or MD membrane could be added in the OMBR-based system in order to reconcentrate draw solute of the FO process and recover clean water [35, 72].

For the anaerobic FO-based systems, some researchers utilized the FO process to recover nutrients from the anaerobically digested wastewater [33, 73]. For example, Wu et al. [57] employed the FO process to recover nutrients *in situ* in the form of struvite from digested swine wastewater. They utilized the reverse solute flux, in which 0.5 M $MgCl_2$ was used as a draw solution and the reverse-fluxed Mg^{2+} ions thereby improved the struvite precipitation [74]. As a result of this, > 99% of phosphate and > 93% of ammonium could be recovered/removed. Simultaneously, the 3.12 LMH of the water flux could be obtained in the FO system. A preliminary economic analysis was carried out in this study, demonstrating that the total value of recovered products (both struvite and water) was around 1.35$/m^3. It should be noted here that the calculations of manpower costs and energy consumption of pump and magnetic stirrer were not included in the assessment.

However, the relevant studies on the AnOMBR-based systems used for the nutrients recovery are rare. In 2017, Hou et al. [58] coupled AnOMBR with a microbial recovery cell (MRC) system, in which the current coming from the MRC drove nutrient and mineral salts from the AnOMBR and enriched them into a separate chamber to form nutrient-rich solution (Fig. 14.6). The recovery efficiencies of phosphate and ammonium were 65% and 45%, respectively, with simultaneous production of 0.19 L CH_4/g COD.

 # Insert Fig. 14.6 #

Although the aforementioned FO-MD/RO systems could effectively reconcentrate the draw solute, the doubts about the economic analysis exist [57]. These aforementioned systems may consume an amount of energy and costs, and it is not sure whether the production of recycled water could offset the economic consumption associated with the draw solute recovery [75]. Besides, it was found that ammonium bicarbonate could be used as a promising draw solute since the moderate heating could easily recover the chemical [76]. According to this finding, Qin and He [77] integrated a microbial fuel cell with the FO process (MEC-FO) and used the system for ammonium recovery and wastewater treatment (Fig. 14.7). The wastewater was fed into the anode chamber of the MEC, after which the wastewater circulated between the anode chamber of the MEC and the feed side of the FO process. The current applied in the MEC could drive the ammonium transfer from the anode chamber to the cathode chamber, and then the ammonium would be stripped out of the cathode chamber due to the air supply as well as the hydroxyl ions resulted from the cathode reaction of the MEC. Besides, the stripped ammonia reacted with carbon dioxide derived from a power plant to form ammonium bicarbonate, which was used as the draw solute in the FO process. The waste power from the plant was utilized to heat the draw solution to recover ammonium bicarbonate and extract the freshwater, which makes the draw solute sustainable. Similarly, Qin et al. [78] also examined the MEC-FO system for the ammonium and water recovery from landfill leachate. In this scenario, around 66% of ammonium in the feed solution could be recovered while 51% of water can be recycled from the MEC anode effluent. Zou et al. [59] developed an MEC-FO system for simultaneous recovery of ammonium and phosphate from high-strength sidestream centrate. In this scenario, the phosphate was concentrated by the FO process while ammonium was enriched via the MEC. Subsequently, the additional

magnesium ions reacted with the nutrients for the struvite precipitation, in which approximately 99.7% of ammonium nitrogen and 79.5% of phosphorus could be recovered. However, they also indicated the current challenges involved in this system, such as optimizing MEC-FO coordination towards being energy-efficient and reducing nutrient loss.

 # Insert Fig. 14.7 #

14.4   Recommended systems and key challenges

14.4.1 Recommended systems

Currently, the nutrient recovery at a large scale is often conducted from specific types of wastewater sources containing high concentration of ammonium and phosphate [38]. Due to being effective in separation and concentration nutrients from wastewater, membrane technologies have been widely utilized to integrate with current nutrient recovery approaches to improve the efficiency of such recovery system [79]. Obviously, the FO membrane process has advantages over other pressure-driven membrane technologies, including: (1) low overall costs due to absence of hydraulic pressure conditions; (2) high water flux caused by great rejection rate for a wide range of contaminants and low scaling effect; and (3) low membrane fouling potential compared to pressure-driven membrane processes [80, 81]. As a result of this, the FO-based systems have been widely explored for the nutrient recovery. The nutrient recovery via FO-based systems, in fact, could be divided into two processes: nutrient enrichment and nutrient recovery. In nutrient enrichment, the concentration of ammonium and phosphate is mainly affected by several factors including solution pH and membrane property.

The rejection of ammonium and phosphate ions in the FO systems is highly influenced by the pH values of feed solution because the surface of FO membrane could be negatively charged in the alkaline environment (pH > 7) [82]. Therefore, at high pH, the electrostatic attractions between the ammonium ions and FO membrane surface could result in the enhanced adsorption of ammonium ions to the membrane surface, which contributes to the further transfer of ammonium ions to the draw side. This scenario, however, may decrease the number of ammonium ions being recovered in the feed side. On the other hand, the phosphate enrichment in the feed side is enhanced due to the electrostatic repulsion. Apart from this, the existing forms of ammonium and phosphate in the aquatic environment are also influenced by the solution pH as shown below [83]:

$$NH_4^+ \rightleftharpoons H^+ + NH_3 \text{ (aq)} \ (pK_a = 9.3) \tag{14.2}$$

$$H_2PO_4^- \rightleftharpoons H^+ + HPO_4^{2-} \ (pK_a = 7.2) \tag{14.3}$$

Eq. 14.2 shows that high pH (> 9.3) could cause the formation of a large proportion of NH_3(aq), which has higher transport rate than NH_4^+ ions in the FO process according to the Donnan exclusion [38]. It should be noted here that high pH of the feed solution may also convert the ammonium ions into volatile ammonia. Overall, alkaline conditions in the feed solution in FO systems lead to a reduction

in the amount of ammonium retained in the feed side. On the other hand, the phosphate retention in the feed side of the FO system could be enhanced at higher pH due to the increased amount of negative charges that the phosphate ions have at the alkaline condition. For this reason, high pH is beneficial for the phosphate enrichment. However, the neutral pH may be more favorable for both phosphate and ammonium enrichment in the FO process.

Xue et al. [38] explored the impacts of membrane properties on the rejection rate of nutrients in the FO-based system, in which commercial flat-sheet cellulose triacetate (CTA) membrane and thin-film composite (TFC) membrane were compared. Specifically, the TFC membrane is prepared using polyamide [84, 85] while CTA utilized cellulose triacetate as the active layers with asymmetric structure. In the study, they found that CTA membranes could obtain higher rejection rate for ammonia compared to the TFC membranes. There are two explanations for this: (1) the TFC membrane has higher ammonium permeability than the CTA membrane; and (2) the TFC membrane has similarly high negative zeta potential to cation-exchange membranes [84, 85], so the transport of ammonium to the draw side across the membrane could be improved, which is attributed to the cation exchange-like mechanism. Compared to the ammonium ions, the phosphate ions are more easily retained in the feed side in FO systems [63, 86]. The possible reason for this is that the negatively charged surface of the FO membrane could retain a large amount of negatively charged phosphate ions due to electrostatic repulsion. Furthermore, phosphate ion has a large hydrated radius (0.339 nm) than that of the ammonium ion (0.104 nm) [87], and the associated sieving effect of the FO membrane could deter the transport of phosphate ions [38, 87].

In addition, the chemical recovery of nutrients enriched by the FO process is mainly influenced by pH values and chemical dose. Due to the pH effects on the species of nutrients and solubility of precipitates [88], the importance of pH value is highlighted, which could affect the quantity and quality of recovered products. High pH (more than 8 at least) is a prerequisite for the chemical precipitation. In contrast, increasing solution pH over 10 may negatively influence the chemical nutrient recovery. This is because ammonium can be converted into volatile ammonia at high pH, which means a decreased amount of ammonium being recovered by struvite precipitation. Apart from this, Mg- and Ca-based salts are always used to react with nutrients for realizing the chemical nutrient recovery, but the metals ions could be transformed into their hydroxides at alkaline conditions. As a result of this, nutrient recovery through chemical precipitation is detrimentally affected.

For the struvite precipitation, the theoretical molar ratio of Mg: N: P is 1:1:1. However, the amount of magnesium used in practical application is larger than the theoretical value since partial Mg^{2+} ions may react with other ions existing in wastewater [89]. On the other hand, hydroxyapatite precipitation follows a theoretical molar ratio of Ca: P at 1.67:1 [90]. More importantly, if such molar ratio is less than 1.67, a decrease in the phosphate recovery efficiency will be detected.

As discussed above, Table 14.2 summarized the recommendation of such factors.

Table 14.2 Recommendations for optimizing the FO-based systems for nutrient recovery

Nutrient enrichment	Factors	Recommendations
	pH	Neutral pH kept in the feed side of the FO-based system could facilitate the enrichment of both phosphate and ammonium.
	Membrane property	The FO membrane with small membrane thickness and high density active layer can improve the nutrient retainment.
Nutrient recovery	**Factors**	**Recommendations**
	pH	The solution pH in the range of 8-10 could be beneficial for the nutrient recovery via chemical precipitation.
	Chemical dose	The molar ratio of Mg: N: P for struvite precipitation and Ca: P for calcium phosphate precipitation should be more than 1:1:1 and 1.67:1, respectively.

14.4.2 Key challenges

Although the versatility and robustness of the FO-based systems for the nutrients enrichment and further recovery have highly accepted [91, 92], this technology is not without limitations.

Firstly, one significant hindrance is the membrane fouling and scaling. In the FO-based systems, the nutrient recovery is obtained through chemical precipitation. In this scenario, an excessive amount of nutrients which are close to the surface of the FO membrane may result in the spontaneous precipitation with mineral salts, and the precipitates could be thereby formed on the membranes surface. Consequently, the membrane performance is negatively influenced as well as the membrane life-span. This is despite the fact that chemical cleaning, physical cleaning and other methods could be employed for the membrane scaling. Apart from this, few studies reported such membrane scaling. The possible reason for this is that the current FO-based systems for the nutrients recovery were operated in a relatively short period. For this reason, the nutrient recovery via FO-based systems should be carried out at pilot-scale or even plant-scale so the risk of membrane scaling in the system could be evaluated, according to which we can settle on relevant strategies for addressing the membrane scaling. In addition, the cake layer of the FO membrane could be formed due to the existence of calcium and phosphate in the feed solution [93], but membrane flushing could be used to remove cake formation [39, 40].

Secondly, the membrane materials in the FO-based system should be given more attention [31] because the water permeability – solute selectivity tradeoff is a key factor affecting the performance of the FO membrane [94, 95], especially the rejection rate of nutrients. Specifically, phosphate and ammonium ions could be

highly concentrated in the feed solution for the production of nutrient-rich stream, while applying an FO membrane with high solute selectivity effectively. However, the reverse salt flux is negatively affected by this kind of membrane, which may reduce the concentrations of Mg^{2+} and Ca^{2+} coming from the draw solution. As a result of this, the nutrient recovery via chemical precipitation is impaired. In contrast, the FO membrane with high water permeability could result in higher reverse salt flux which supplements Mg^{2+} and Ca^{2+} ions for the chemical precipitation. Nevertheless, phosphate and ammonium ions more easily transfer across the membrane to the draw side at the same time, which leads to a significant loss of nutrients. Hence, how to find a balance between the water permeability and solute selectivity is critical for recovering nutrients in the FO-based systems.

14.5 Future roadmap

Since phosphate and ammonium could be biologically consumed for the bacterial growth and phosphate could be adsorbed to the sludge through the PAOs, the anaerobic FO-based system for nutrient recovery seems more effective.

As discussed above, the FO-based systems offer a unique opportunity to obtain the nutrients recovery from wastewater. Although the aerobic FO-based systems have presented excellent potential for nutrient enrichment and further recovery [36, 56], the nutrients could be consumed or converted by activated sludge in the aerobic environment; simultaneously, ammonium may be converted into volatile ammonia under aeration supply. As a result of this, the amount of nutrients involved in the nutrient recovery via chemical precipitation could be decreased whilst the economic feasibility would be detrimentally affected. In contrast, anaerobic FO-based systems can maximize the content of phosphate and ammonium through biologically releasing nutrients and then converting them into soluble forms for further recovery by chemical precipitation. Furthermore, the amount of nutrients consumed by biological uptake is fewer under anaerobic conditions when compared to aerobic conditions. More importantly, the anaerobic FO-based process converts organic substances into methane-rich biogas, which could offset the energy consumption in the recovery system [96]. The easy integration of the anaerobic FO process with current wastewater treatment infrastructure also makes the system more accessible [39, 40]. In conclusion, anaerobic FO-based systems are advantageous than the aerobic FO-based processes in wastewater treatment, including low energy consumption due to the absence of air supply, significant potential to achieve energy-neutral balance because of biogas production and the high content of nutrients due to anaerobically biological nature [97, 98]. Therefore, more studies are needed to focus on the anaerobic FO-based systems for recovering nutrients in wastewater treatment.

As discussed above, the MEC-FO system presented excellent performance for simultaneously recovering ammonium and phosphate from wastewater. Microbial fuel cells (MFCs) which have similar functions to MECs should be highly encouraged to combine with the FO process for the nutrients recovery. The possible reason for this is that: firstly, the MFC could generate electricity because of a series of bioelectrochemical activity of anaerobic bacteria, which shows less energy input than MECs; and secondly, the solution pH can be increased by the

cathode reaction in MFCs without adding alkaline chemicals. Recently, Ye et al. [60] combined the FO membrane with the MFC to form an MFC-FO system, in which 98.81% of NH_4^+-N and 83.18% of PO_4^{3-}-P were removed/recovered from municipal wastewater. It was also reported that the current generated in MFCs could reduce the membrane fouling potential [99]. Thus, more investigations on MFC-FO systems would be beneficial for the nutrients recovery in wastewater treatment.

In addition, the environmental value of nutrients has not been fully recognized by current economic analysis. More importantly, people still prefer to achieve the nutrients used for the fertilizer production through industrial activities such as mining and the Haber-Bosch process because of mineable phosphate-based rocks, low costs of natural gas and electricity. In this scenario, the incentives to focus on the nutrient recovery in wastewater treatment are not adequate. Besides, the recovered nutrients (e.g., struvite) are not widely accepted worldwide despite some applications in a certain areas of the world; thus, their market values still need more researching. It was reported that the purity and quality of recovered products determine the market value [54], but research conducted to date has not been concentrated on the purity of recovered products achieved in the aforementioned FO-based nutrient recovery systems. Overall, the nutrient recovery from wastewater is essential for the food security, especially given the rapid rise of global population, decrease of the phosphate-bearing rocks and high costs for producing industrial ammonium. Therefore, publicity and governmental incentives may be acted as an effective strategy to enhance the research on nutrient recovery in the FO-based systems.

14.6 Conclusion

FO technology could highly retain nutrients within reactors with low energy input and membrane fouling potential from a wide range of wastewater sources. The strategic integration of the FO process with other technology (e.g., chemical precipitation) can harvest the valuable nutrients. Compared to aerobic FO-based technologies, anaerobic FO-based systems could better improve nutrient recovery efficiency and produce reusable biogas, which enhances the economic feasibility of the recovery systems. Nonetheless, the challenges such as anaerobic system integration, membrane fouling and scaling, and membrane materials still exist. Innovative approaches highlighted in this chapter may be effective to resolve these challenges. Besides, the performance of the FO-based systems for the nutrients recovery needs more investigations into the economic feasibility and applications at the pilot- and plant-scale to make these systems more viable.

14.7 References

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Figure captions

Figure 14.1. Schematic of FO-based system configurations in wastewater treatment: (a) aerobic FO system; and (b) anaerobic FO system.

Figure 14.2. Schematic of FO-based system for the nutrients recovery.

Figure 14.3. Schematic of (a) current wastewater treatment technologies (adapted from Verstraete and Vlaeminck [73]); and (b) FO-based wastewater treatment technologies (adapted from Ansari et al. [2])

Figure 14.4. Schematic of OMBR-based system for the nutrients recovery via chemical precipitation (modified from Qiu and Ting [56]).

Figure 14.5. Schematic of OMBR-MF hybrid system for the nutrient recovery (modified from Qiu et al. [55]).

Figure 14.6. Schematic of MEC/MRC-AnOMBR system (adapted from Hou et al. [30]).

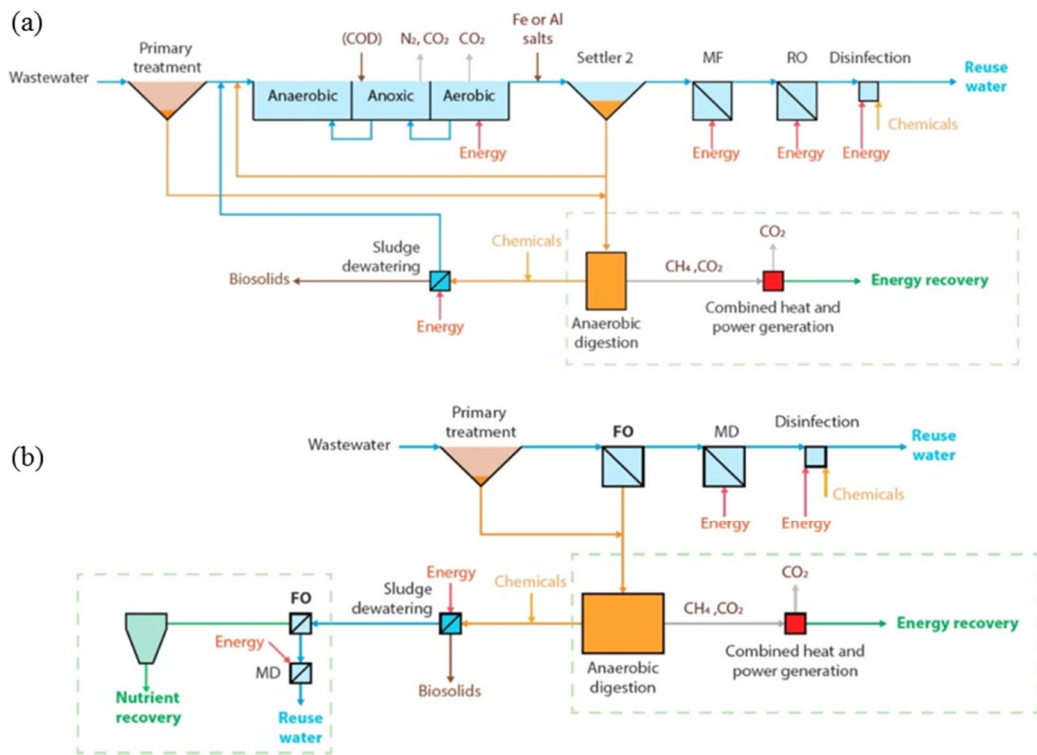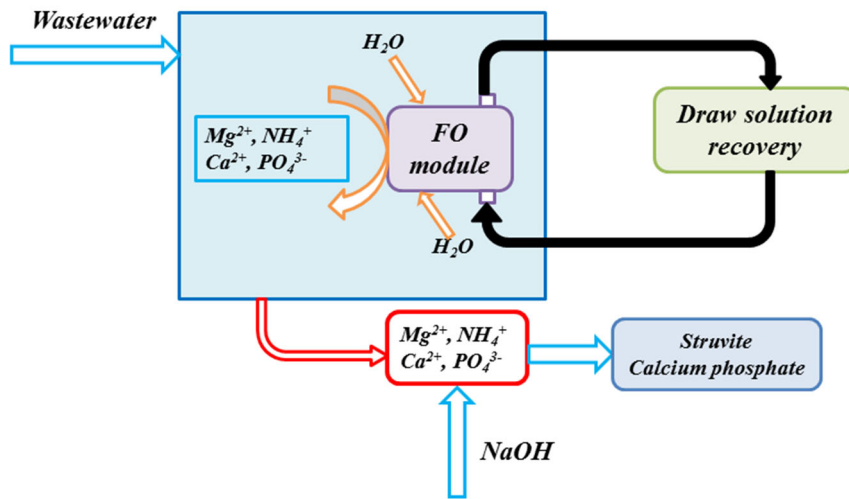**Figure 14.7.** MEC-FO system for nutrients recovery (modified from Zou et al. [98]).
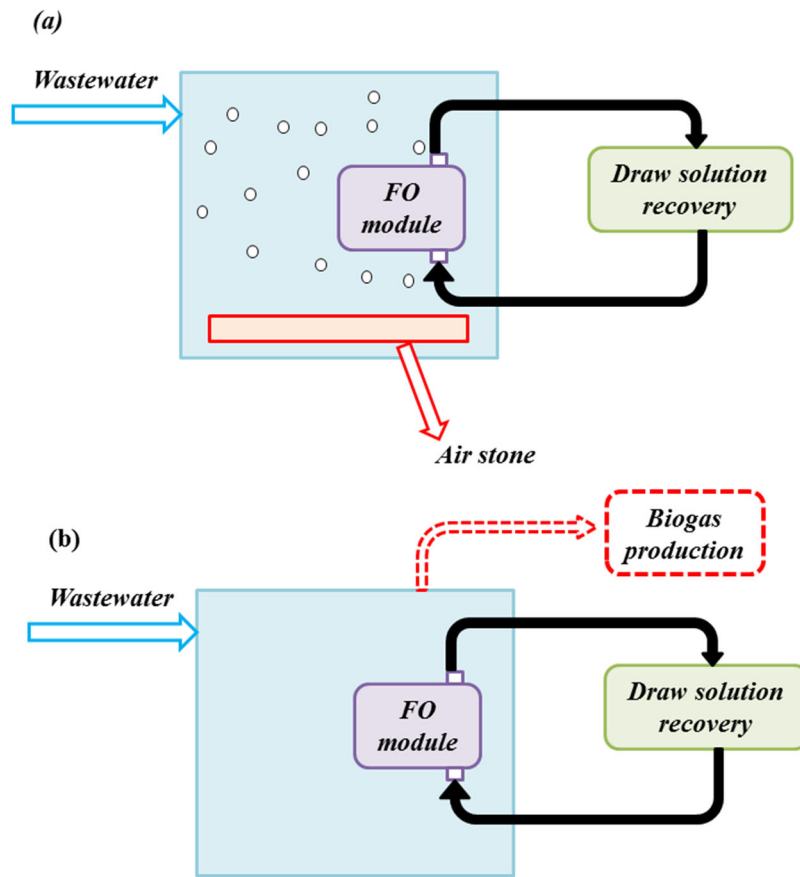
Fig. 1
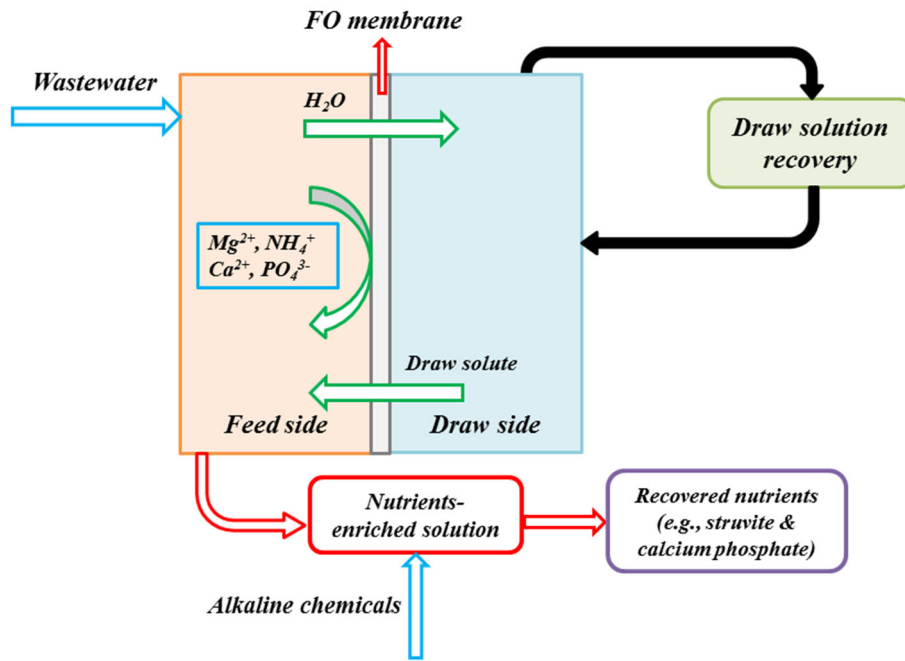
Fig. 2

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