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Forward osmosis as a platform for resource recovery from municipal wastewater - a critical assessment of the literature

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Abstract

Forward osmosis (FO) is an emerging membrane separation technology that has the potential to serve as a game changer in wastewater treatment. FO-based processes can simultaneously produce high quality effluent and pre-concentrated wastewater for anaerobic treatment to facilitate the recovery of energy and nutrients. Complex wastewaters can be directly pre-treated by FO and fresh water can be produced when coupled with a draw solute recovery process (i.e. reverse osmosis or membrane distillation). By enriching organic carbon and nutrients for subsequent biogas production, FO extends the resource recovery potential of current wastewater treatment processes. Here, we critically review recent applications of FO for simultaneous treatment and resource recovery from municipal wastewater. Research conducted to date highlights the importance of successfully integrating FO with anaerobic treatment. Emphasis is also placed on the development of novel FO-based hybrid systems utilising alternative energy sources for draw solute recovery. There remain several technical challenges to the practical realisation of FO for resource recovery from wastewater including salinity build-up, membrane fouling, and system scale-up. Strategies to overcome these challenges are critically assessed to establish a research roadmap for further development of FO as a platform for resource recovery from wastewater.

Keywords

platform, resource, recovery, municipal, wastewater, -, critical, assessment, literature, osmosis, forward

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Abstract

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Keywords: forward osmosis (FO); wastewater treatment; resource recovery; anaerobic treatment; biogas; phosphorus recovery.

1. Introduction

The recovery of water, energy, and nutrient resources from municipal wastewater presents a promising solution to a number of prevalent economic, environmental, and social issues. Wastewater reclamation can address both water scarcity and environmental pollution [1, 2]. Utilisation of the biogas produced from the organic content of wastewater can offset the energy requirement for treatment [3]. Nutrient recovery from wastewater also deserves special attention due to the increasing stringency of effluent discharge regulations and uncertainties associated with minable phosphorus supply for food security [4-6]. Increasing awareness of the potential resource value of municipal wastewater has prompted significant research efforts to synergise emerging wastewater treatment processes and resource recovery techniques [3, 7, 8].

Activated sludge treatment is an established biological process that focusses primarily on purifying wastewater of organic matter, pathogens, and nutrients, but does not effectively facilitate energy and nutrient recovery. Activated sludge treatment is energy intensive due to the high electricity demand for aeration and also produces excessive amounts of sludge residuals [9]. During activated sludge treatment, the carbon (i.e. chemical energy) and nitrogen (i.e. nutrient) contents of wastewater are converted to biomass, carbon dioxide, and nitrogen gas. In other words, much of the energy and nutrient contents of wastewater are dissipated at the expense of significant energy input. As an alternative, anaerobic treatment converts organic substances into methane rich biogas in the absence of oxygen and transforms phosphorus to a more chemically available state for subsequent recovery [10]. Transitioning from aerobic towards anaerobic based treatment processes has significant potential to lower the energy consumption of wastewater operations (i.e. by avoiding aeration), as well as achieve energy-neutral wastewater treatment (i.e. through biogas production) [11-17].

The opportunity for wastewater treatment plants to provide a renewable source of useful heat and electricity through biogas conversion is immense [18, 19]. In fact, the chemical energy content in municipal wastewater exceeds the electricity requirement of operating an activated sludge plant by at least nine times [20]. Despite this significant embedded energy content, there are a number of major challenges that currently restrict the feasibility of directly anaerobically digesting raw wastewater for energy recovery. The concentration of organic matter in wastewater is typically low. Therefore, a sufficient organic loading rate cannot be maintained in the anaerobic digester, resulting in a low biogas yield and inadequate removal of organic pollutants from wastewater. In addition, since methane is slightly soluble in water (22.7 mg/L), at a low biogas yield, much of the generated methane can be lost via effluent discharge [10]. Several membrane filtration technologies have been integrated with anaerobic treatment to overcome these challenges, aiming to improve the retention of biomass in the reactor and to increase effluent quality. Anaerobic membrane bioreactors (An-MBRs) utilising low pressure membranes such as microfiltration (MF) or ultrafiltration (UF) is a notable approach. Nevertheless, the MF/UF membranes used in conventional An-MBRs cannot retain dissolved organic carbon. Thus, they are not effective for energy recovery and cannot produce a high effluent quality [10].

Further development in An-MBR technology has resulted in the novel hybridisation of anaerobic treatment with high retention membrane processes including nanofiltration (NF), membrane distillation (MD), and forward osmosis (FO) [21]. Among these high retention membrane processes, FO stands out as the most promising candidate for integration with anaerobic treatment due to a combination of high separation efficiency and high fouling reversibility [22-25]. The integration of FO with anaerobic treatment has been widely reported in the literature [26-30]. FO is a unique membrane process that utilises the physical phenomenon of osmosis to transport water across a semipermeable membrane. As a major advantage, the FO process itself can operate with minimal external energy input [31]. However, further treatment of the draw solution is required to extract fresh water and can be achieved using pressure driven or thermally driven membrane processes [32]. Lutchmiah, et al. [33] provided a critical assessment of FO applications for water reclamation. They also highlighted the need to develop new membrane materials and optimise draw solute selection as well as key operating conditions to facilitate full-scale implementation of FO for water reclamation applications [33]. In another excellent review, Holloway, et al. [34] systematically summarised and reviewed all relevant works related to osmotic membrane bioreactors for the production of high quality potable water from impaired sources including wastewater. In particular, Xie, et al. [7] identified the untapped potential of FO amongst several other membrane separation processes for recovering nutrients from municipal wastewater. Indeed, there is a consensus that FO has the potential to be an important technology in the future of wastewater treatment [31, 33, 35, 36].

Integrating FO with anaerobic treatment is essential for energy and nutrient recovery. The viability of the anaerobic osmotic membrane bioreactor (An-OMBR) has been demonstrated where the FO membrane is submerged inside the anaerobic bioreactor [26, 28, 29]. An alternative approach uses FO to firstly pre-concentrate raw wastewater to a high strength for subsequent anaerobic treatment. The concept of wastewater pre-concentration is yet to be fully explored, but it holds significant opportunities for resource recovery applications. Preliminary investigations into FO draw solution selection [27, 37] and process efficiency [38-40] have been conducted. However, issues of salinity accumulation, membrane fouling, and anaerobic treatment integration have not been adequately addressed.

Here, we critically review recent applications of FO for recovering energy and nutrients from municipal wastewater by integrating with existing resource recovery techniques (i.e., anaerobic digestion and phosphorus precipitation) and other complementary processes (e.g., membrane distillation (MD) and reverse osmosis (RO)) for clean water extraction. The challenges and potential opportunities associated with FO-based treatment processes are evaluated in terms of treatment efficiency and resource recovery potential. The outlook of an integrated FO membrane-based system for simultaneous wastewater treatment and resource recovery is discussed. A research roadmap for further development of FO for resource recovery from wastewater is also provided and discussed.

2. FO for wastewater treatment

Interest in applying FO for wastewater treatment has grown significantly in recent years [32, 33, 35, 41-43]. These potential applications are motivated by several advantages of FO over current wastewater treatment technologies. Given its high fouling reversibility, FO can be directly applied to a complex solution without extensive pre-treatment [44]. A high rejection of dissolved contaminants is another important advantage of FO for wastewater treatment. When FO is combined with a draw solute recovery process, clean water can be produced from the draw solution, furthering water reuse opportunities. These unique features of FO have spurred the development of several system configurations for wastewater treatment and water reclamation.

2.1 FO system configurations for wastewater treatment

Three major system configurations have been developed for FO wastewater treatment applications and vary depending on the type of solution in contact with the FO membrane (Figure 1). Firstly, the most widely recognised approach is the aerobic osmotic membrane bioreactor (Ae-OMBR) [45-51] (Figure 1A) whereby wastewater is fed into an activated sludge reactor. Secondly, several research groups have explored the potential of An-OMBRs [26, 28, 29] (Figure 1B) for wastewater treatment and the production of biogas. Both OMBR configurations typically utilise a submerged FO module, as the high solids content of the mixed liquor and digested sludge can cause blockages in other arrangements. The third configuration (Figure 1C) adopts a similar concept to the An-OMBR (Figure 1B). However, in this configuration, wastewater is firstly pre-concentrated by the FO membrane prior to anaerobic digestion [27, 39, 52]. A key benefit of this configuration is that the FO membrane is in contact with concentrated wastewater, which has lower fouling propensity compared with the mixed liquor inside an An-OMBR. Similar to conventional MBRs, the submerged configuration appears most suited for wastewater pre-concentration, to reduce the costs associated with circulating the feed solution through an external membrane module [53].

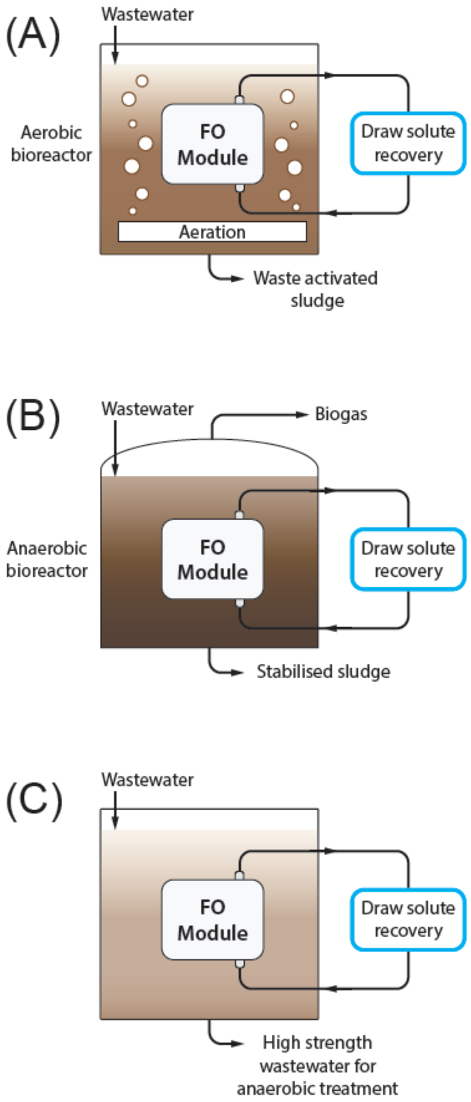

Figure 1: Schematic representation of three major FO system configurations for wastewater treatment: (A) Ae-OMBR, (B) An-OMBR, and (C) wastewater pre-concentration intended for subsequent anaerobic digestion.

2.2 Treatment performance of FO systems

The level of treatment provided by each FO system can differ considerably, and can be attributed to the type of applied biological treatment, process conditions, and membrane properties (Table 1). The treatment performance of an FO system is generally indicated by the efficiency to remove organic matter, nitrogen, phosphorus, and trace organic contaminants (TrOCs).

Table 1: Summary of FO wastewater treatment performance in terms of the removal efficiency of organic matter (i.e. total organic carbon (TOC) and chemical oxygen demand (COD)), phosphorus (i.e. total phosphorus (TP)), and nitrogen (i.e. NH_4^+-N and total nitrogen (TN)).

FO system configuration	**Membrane (arrangement)**	**Removal efficiency (%)**					**Ref.**
		Organic matter		**Phosphorus**	**Nitrogen**		
		TOC	**COD**	**TP**	**NH_4^+-N**	**TN**	
Ae-OMBR	CTA (cross-flow)	98%	-	-	99%	-	[54]
	TFC (cross-flow)	96%	-	-	99%	-	[54]
	CTA (submerged plate-and-frame)	-	>99%	>99%	-	>82%	[47]
	CTA (submerged plate-and-frame)	98%	-	>99% PO_4^{3-}	80-90%	-	[49]
	CTA (submerged plate-and-frame)	98%	-	-	98%	-	[55]
	CTA (submerged plate-and-frame)	>98%	-	-	>98%	-	[24]
An-OMBR	CTA (submerged plate-and-frame)	-	>95%	>99%	FO only Ammonia = 70-80%	-	[28]
	CTA (submerged plate-and-frame)	-	96.7%	99%	60%	-	[26]
	CTA (submerged plate-and-frame)	92.9%		-	-	-	[29]
Wastewater pre-concentration	CTA (submerged plate-and-frame)	-	99%	99% PO_4^{3-}	Ammonia = 67-68%	56-59%	[56]
	CTA (pilot-scale spiral wound)	-	99.8%	99.7%	48.1%	67.8%	[40]

In all FO system configurations discussed above, a high removal efficiency of a broad range of contaminants can be achieved, since FO membranes are highly effective at retaining organic compounds, colloidal particles, and microbes in the feed solution (Table 1). Similarly, FO membranes have consistently demonstrated near complete rejection of phosphorus for two reasons. Electrostatic repulsion occurs between negatively charged phosphate ions and the negative surface charge of the FO membrane, deterring phosphate transport through the membrane. Another important rejection mechanism for phosphorus is size exclusion, as phosphate has a large hydrated radius, it is rejected by a sieving effect [57]. The superior rejection capability of FO membranes for organic matter and phosphorus has far reaching implications for wastewater treatment and resource recovery. To highlight this point, conventional An-MBRs (i.e. which utilise MF or UF membranes) cannot achieve sufficient phosphorus removal and have a significantly lower organic matter removal efficiency compared to An-OMBRs [28]. Thus, the integration of FO with anaerobic treatment in the form of An-OMBR can significantly improve the overall system treatment capacity and viability for wastewater treatment.

The removal of nitrogen by FO-based systems is highly variable and depends on the actual structure of nitrogen bearing compounds in the solution and the biological treatment process [58]. FO membranes alone have an incomplete rejection of neutral ammonia (i.e. <80%) [28, 56] compared with positively charged ammonium ions [57]. At neutral pH, Ae-OMBRs can provide some nitrogen removal capacity as a result of both biological degradation (i.e. via nitrification/denitrification) and FO membrane rejection (Table 1). On the other hand, An-OMBR and FO-anaerobic systems do not provide any biological nitrogen removal capacity. However, it is noteworthy that nitrogen removal could be achieved via struvite (i.e. magnesium ammonium phosphate) recovery, by deploying a dedicated ammonia recovery process, or by converting ammonia microbiologically into nitrous oxide for enhanced biogas utilisation [59].

The high TrOC removal capability of FO membranes is another notable advantage [60, 61]. Safe implementation of potable water reuse schemes relies on the ability of treatment processes to remove a wide range of TrOCs including, pharmaceutical residues, steroid hormones, phytoestrogens, UV-blockers, and pesticides [62-64]. In terms of FO configurations for wastewater treatment, the Ae-OMBR is likely to offer the most effective removal of TrOCs due to the combined effect of biodegradation and membrane rejection

[60]. It is noteworthy that the removal of TrOCs by An-OMBRs has scarcely been reported in the literature [65].

2.3 *FO membrane-based hybrid systems for water recovery*

Additional separation processes must be integrated with FO to recover fresh water and re-concentrate the draw solution. Key considerations for the draw solute recovery process include the ability to reject the draw solutes, draw solution compatibility with the subsequent biological treatment process, and energy requirements of the overall hybrid system. Hybrid systems that couple FO with pressure driven (e.g. NF and reverse osmosis (RO)) [48, 66], thermally driven (e.g. MD) [67-69], or electrically driven (e.g. electrodialysis (ED)) [70] membrane processes have been reported in the literature (Figure 2). In these hybrid systems, FO pre-treats wastewater and provides a foulant-free solution for draw solute recovery. As a result, FO membrane-based hybrid systems have the potential to produce a higher quality effluent and improved process efficiency compared with treating raw wastewater directly with the above mentioned high retention membrane processes [35]. FO membrane-based hybrid systems are often termed as a double-barrier defence for a wide range of contaminants. However, as discussed in the next section, some contaminants can accumulate in the draw solution, presenting a limitation for the practical application of these hybrid systems.

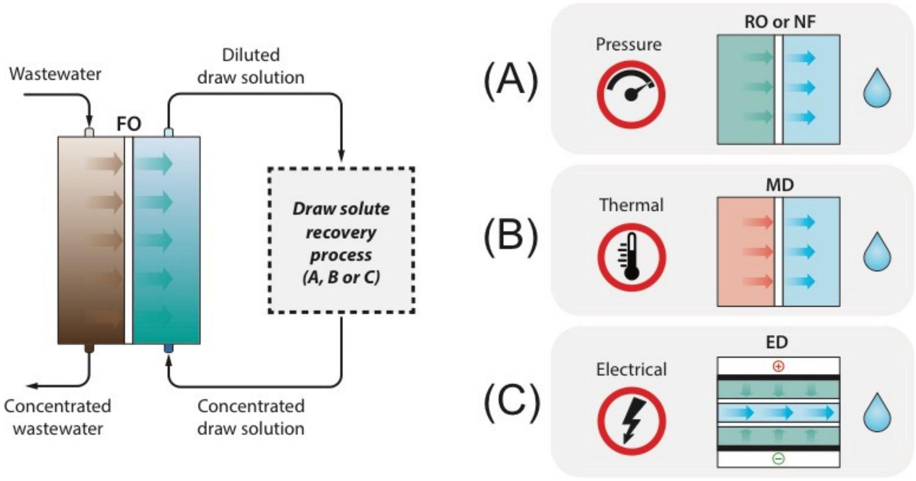

Figure 2: Schematic of FO membrane-based hybrid systems utilising: (A) pressure driven RO or NF, (B) thermally driven MD, and (C) electrically driven ED.

2.3.1 Contaminant accumulation in the draw solution

A major limitation for the practical application of FO membrane-based hybrid systems is the potential accumulation of contaminants in the draw solution. FO membranes are not completely impermeable to all dissolved solutes. Thus, contaminants that pass through the FO membrane but are retained by the draw solute recovery process inevitably accumulate in the draw solution of the closed-loop system. Previous studies have observed the accumulation of small organic compounds, ammonium, and phosphate using FO-RO [48] and FO-MD [71] hybrid systems. Accumulation of TrOCs has also been observed, with the type of TrOC depending on the rejection capability difference between the FO and draw solute recovery processes [71, 72].

Contaminant accumulation is an issue for the practical application of FO hybrid systems as the product water quality can be hampered and may even lead to membrane fouling in the draw solute recovery process [32, 73]. Luo et al. [48] presented evidence that the accumulation of contaminants in the draw solution of an Ae-OMBR-RO system caused an increased RO permeate concentration of organic matter and ammonium, hence, negatively affecting product water quality. Similar results were reported by D'Haese et al. [72] when they modelled TrOC accumulation in an FO-RO system. They observed TrOC build-up to a value in excess of the feed concentration and led to a contaminated product water [72]. The risk of membrane fouling in the draw solute recovery process caused by contaminant accumulation in the draw solution has also been demonstrated. The permeability of the RO membrane in an OMBR-RO system was shown to gradually decline, suggesting that some small organic molecules can accumulate and act as foulants on the RO membrane [48]. The risk of fouling is also applicable to other draw solute recovery processes after long-term operation, unless mitigation strategies are adopted.

To safegard the production of high quality product water and to reduce the risk of membrane fouling in FO membrane-based hybrid systems, additional treatment processes can be integrated to mitigate contaminant accumulation in the draw solution. The type of treatment process generally depends on the contaminant of concern. In wastewater applications, granular activated carbon (GAC) adsorption and ultraviolet (UV) oxidation have both proved to be effective processes, targeting the mitigation of organic matter and TrOCs [71]. On the other hand, ion exchange has been applied for the removal of accumulated boron in the draw solution of a seawater desalination process [74]. For wastewater specific applications, further research is required to address a number of practical considerations when mitigating contaminant accumulation in the draw solution. It is noted that draw solution selection can greatly impact the applicability of the applied mitigation strategy. For example, GAC and UV are not compatible when organic-based draw solutions are adopted as the draw solute can interfere with the adsorption process or be degraded by UV radiation, respectively [38, 75].

Further research is necessary to assess the extent and impact of contaminant accumulation over long-term operation in wastewater applications using FO. Ongoing research progress in the fabrication of FO membranes can improve the rejection of target contaminants and supress their accumulation in the draw solution [76]. Promising results have been achieved through the application of novel side-stream processes to remove contaminants from the draw solution in systems that utilise RO, NF, or MD for draw solute recovery. When ED is used for draw solute recovery, post-treatment methods may be necessary since ED has a relatively low removal capacity for organic compounds [70]. In addition, FO operating parameters can also be optimised to minimise the forward diffusion of contaminants into the draw solution.

2.3.2 Energy consideration for FO membrane-based hybrid systems

Energy considerations for membrane-based hybrid systems are of paramount importance as the draw solute recovery process dictates the energy consumption of the entire hybrid system [77]. In fact, the FO process itself only requires minimal energy for water transport through the membrane as the draw solution provides the osmotic driving force [31]. FO based hybrid systems can utilise mechanical (i.e. pressure), thermal, or electrical energy to power the draw solute recovery process (Figure 2). Although the energy demand for draw solution regeneration either by RO or MD is high [35], it is noteworthy that membrane fouling associated with FO wastewater treatment is highly reversible compared with direct RO [23] or MD filtration [71]. By comparison, during conventional wastewater treatment, intensive pre-treatment is required (i.e. activated sludge treatment and MF) prior to RO for potable water production. In other words, the costs associated with these conventional wastewater treatment processes could be replaced by the FO process.

The most promising avenue for FO membrane-based hybrid treatment systems to provide low energy treatment of wastewater arguably involves applications whereby low-cost heat can be utilised for draw solute recovery. MD is a thermally driven membrane process that has significant potential, since alternative low-cost or waste thermal energy can be applied to power the draw solute recovery process. It is noteworthy that in all thermally driven processes, the energy efficiency is inversely proportional to temperature (thermal quality) [78]. Thus, the abundance of cheap or free low-grade heat is an important factor. In areas of high solar radiation, solar thermal can be used as the primary energy source. Alternatively, low-grade waste heat could be captured from nearby industrial processes. Lastly, the heat co-generated from the production of biogas from wastewater organic matter presents a practical approach to supply such thermally driven separation processes.

In terms of energy consumption, very few comprehensive comparisons of draw solute recovery processes have been reported in the literature. Life cycle analyses of FO-RO hybrid system primarily focus only on seawater desalination applications. The results were inconclusive and show that at the current stage of FO development, FO-RO processes may have comparable costs [79] or a higher energy use and environmental impact [80] compared with current technology for seawater desalination and water reuse. It is also noted that there has not been any life cycle analysis of FO-based hybrid system specifically for wastewater treatment applications. Further studies are crucial to practically evaluate the energy outlook of FO processes related to wastewater treatment and resource recovery applications.

Another potential opportunity to improve the energy favourability of FO systems involves the case where the diluted draw solution has a direct use, therefore no draw solute recovery process is required. For example, the use of fertilizers as a draw solution to extract clean water for irrigation from compromised sources has been recently demonstrated. The product is a diluted fertiliser solution that can potentially be directly applied for fertigation purposes [30, 81, 82]. In other words, water is recovered in a directly usable form. There is a similar argument for the use of seawater RO brine as the draw solution. Researchers have proposed that diluting the brine by treating wastewater with FO, and subsequently extracting water by seawater RO desalination can provide a sustainable approach to dual issues (i.e. wastewater management and fresh water availability) [83]. In some cases, it has been reported that the required energy for the combined osmotic dilution and water recovery by RO is more than a single RO process [35]. The suitability of osmotic dilution is highly dependent on local factors, however the low energy consumption of osmotic dilution is a major advantage.

2.3.3 Other limitations of FO-based hybrid systems

Further to contaminant accumulation in and energy considerations, there are a number of inherent limitations of FO-based hybrid systems. During the process the loss of draw solute (i.e. reverse solute flux) negatively impacts process efficiency by lowering the osmotic driving force [84], increasing operating costs as solute must be periodically supplemented [85], and elevates salinity accumulation in the feed solution [86]. Another limitation is the low water flux of the FO process [87]. Unless significant improvements in membrane materials and draw solution efficiency are made, the capital costs associated with the required FO membrane area to compensate the low flux are extensive.

3. Resource recovery using FO

Extending the established efforts of wastewater treatment, FO has been recognised as a highly suitable technological building block to facilitate nutrient and energy recovery from wastewater. Numerous recent studies have demonstrated the capability of FO-based processes to improve the recovery of energy and nutrients from various wastewaters (Table 2). Some of these FO-based processes are able to recover resources whilst simultaneously providing wastewater treatment when coupled with a draw solute recovery process. Despite these promising demonstrations of simultaneous wastewater treatment and resource recovery by FO-based processes, a number of key technical challenges require further development. Further research is needed to optimise the integration of FO with anaerobic processes for biogas production, to overcome issues of salinity accumulation and membrane fouling. Also, it is necessary to focus efforts to develop nutrient recovery using FO to address the key issues of product purity and membrane fouling/scaling during long-term operation.

Table 2: Summary of FO-based resource recovery processes.

Feed solution	FO-based process	Recovered resource	Draw solution	Draw solute recovery process	Performance	Ref.
Synthetic wastewater	An-OMBR	Biogas	NaCl	Manual re-concentration	Methane yield = 0.21 L CH_4/g COD	[26]
	An-OMBR	Biogas	NaCl	Manual re-concentration	Methane yield = 0.3 L CH_4/g COD	[28]
	An-OMBR	Biogas	NaCl and Na_2SO_4	Manual re-concentration	NaCl An-OMBR had a higher biogas methane composition than Na_2SO_4 An-OMBR	[29]
Activated sludge	Ae-OMBR	Calcium phosphate	$MgCl_2$ and NaCl	Manual re-concentration	Phosphorus content >11%	[49]
	MF-Ae-OMBR	Calcium phosphate	Seawater brine	Osmotic dilution	MF extracted dissolved nutrients. Phosphorus content = 11–13%	[88]
	MF-Ae- OMBR-RO	Calcium or magnesium phosphate Fresh water	NaCl	RO	Precipitate = 15-20% phosphorus	[48]
Secondary treated effluent	FO pre-treatment	Nutrient concentrate (i.e. ammonia and phosphate)	Synthetic seawater	Osmotic dilution	Ammonia removal = 66.7% Phosphate removal = 92.1	[57]
Digested sludge centrate	FO-RO	Nutrient concentrate (i.e. ammonia and phosphate) Fresh water	NaCl	RO	Ammonia removal =82.9–92.1 % Phosphate removal=99.6–99.9% Optimum water recovery=70%	[89]
	FO-MD	Struvite ($MgNH_4PO_4.6H_2O$) Fresh water	$MgCl_2$	MD	Ammonium removal >90% Phosphate removal >97% Bidirectional diffusion of Mg^{2+} and protons improved struvite recovery.	[90]
	FO pre-treatment	Calcium phosphate	Seawater	Osmotic dilution	Phosphate removal > 98% Bidirectional diffusion of protons improved calcium phosphate recovery.	[91]
Urine	FO pre-treatment	Nutrient concentrate (i.e. ammonium, phosphate, and potassium)	Synthetic seawater and brine	Osmotic dilution	Ammonium removal = 50–80% Phosphate removal > 90% Potassium removal >90%	[58]

3.1 *Integrating FO with anaerobic treatment for biogas production*

Integrating the FO process with anaerobic treatment is a promising avenue to produce biogas and recover nutrients from wastewater. Demonstrations of FO-based biogas producing systems have focused almost exclusively on An-OMBRs, where the FO membrane is submerged within the anaerobic bioreactor (Table 2). Recent research has reported the potential of An-OMBRs as methane yields between 0.2-0.3 L CH_4/g COD were achieved in lab-scale studies [26, 28]. Compared to conventional anaerobic digesters the inclusion of the FO membrane can provide a number of important advantages. The treatment performance of AnOMBRs surpasses conventional anaerobic systems in terms of organic matter and nutrient removal [10, 30]. FO membrane separation also allows the system to operate at a high organic loading rate by decoupling the hydraulic retention time and the solid retention time, hence, lowering the process footprint [92]. Lastly, potable water production is enabled by adopting an appropriate draw solute recovery process for the draw solution.

An alternative approach that could essentially achieve the same objective of An-OMBRs involves directly processing primarily treated wastewater by FO and then feeding the concentrate to an anaerobic treatment system. As a key advantage of this configuration (Figure 1C), the FO membrane is in contact only with wastewater, which is more dilute than sludge. Sun, et al. [93] reported that fouling reversibility was higher in a direct FO system compared to an OMBR, attributed to differences in the solutions microbiological behaviour [44]. Similarly, membrane degradation may be less severe in direct FO configurations, as prolonged exposure to activated sludge in OMBRs has shown significant performance degradation to both cellulose triacetate (CTA) and thin film composite (TFC) FO membranes [94]. Furthermore, the volumetric loading of the anaerobic treatment system could be drastically reduced, owing to the pre-concentration of wastewater by the FO membrane. Preliminary studies have demonstrated that FO can pre-concentrate COD in dilute wastewater up to approximately eightfold, corresponding to a tenfold volume reduction [38]. Enriching the COD concentration of wastewater has the potential to increase the energy recovery per unit volume of digestate and to minimise heating energy requirement [52].

The primary purpose of considering anaerobic treatment for wastewater treatment is to recover the chemical energy contained in wastewater through biogas conversion. In the proposed FO-based process (Figure 3), biogas produced from the anaerobic treatment process has significant potential to supply the energy requirements of the system. In this case, MD presents a favourable opportunity for draw solute regeneration, as the driving force of MD is temperature. A combined heat and power engine can convert biogas into heat for the MD system. Furthermore, electricity can be utilised onsite or fed back into the grid. According to an energy audit of the Prague wastewater treatment plant, under an optimal condition, 70-80% energy self-sufficiency could be achieved by fully utilising the embedded chemical energy in wastewater for biogas production [95]. Thus, energy self-sufficiency is possible with further improvement in engineering efficiency. Lastly, anaerobic treatment partially mineralises organic nitrogen and phosphorus to their soluble forms (i.e. ammonium and phosphate). This action increases the chemical availability of nutrients for subsequent recovery. Despite these benefits, the major technical challenges that limit the feasibility of integrated forward osmosis and anaerobic treatment systems are salinity accumulation and membrane fouling.

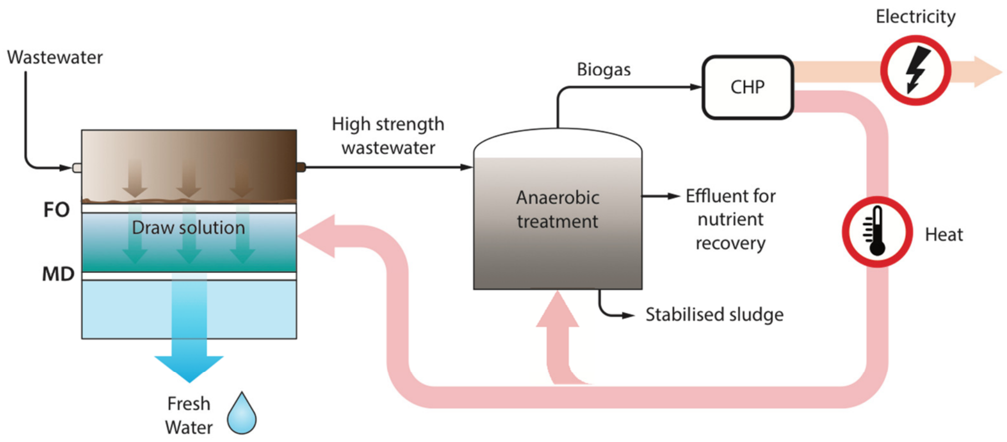

Figure 3: Schematic illustration of an FO pre-concentration process for energy recovery via anaerobic treatment.

3.1.1 Salinity accumulation

Salinity accumulation is a prevalent issue for the integration of high retention membrane processes with biological treatment [21]. For FO, this issue is further exacerbated by the reverse diffusion of solutes from the draw to the feed solution (i.e. reverse draw solute flux). The accumulation of salt in the feed solution inevitably increases its osmotic pressure and can negatively impact water flux. More importantly, salinity accumulation is a major hindrance when integrating FO with anaerobic treatment since methanogenic activity can be inhibited at high inorganic salt concentrations, leading to severely reduced biogas production rates [96]. It is noteworthy to mention that methane solubility decreases as salinity increases [97]. This is beneficial in terms of reducing methane loss via permeate. The extent of salinity accumulation and the impact on water flux and anaerobic treatment is strongly affected by the selected draw solution and the FO operating conditions (i.e. concentration factor). The relative contribution of each salinity accumulation mechanism can be predicted based on the operating conditions and draw solute properties [86, 98]. For this application whereby organic loading rates should be increased, the FO concentration factor must be maximised. Yet, the concentration factor is proportional to the rate of salinity build-up and therefore a trade-off exists between the effects of salinity accumulation and process efficiency. Thus, a variety of strategies have been proposed to alleviate salinity accumulation in FO-based systems.

The draw solution significantly influences both the rate of reverse draw solute flux and the type of solutes that accumulate in the feed solution [84, 99]. Feasibility studies have shown that the use of sodium chloride as the draw solution in An-OMBRs inevitably leads to severe salinity accumulation that detrimentally affects water flux and system efficiency [26, 28]. Furthermore, the accumulation of both sodium chloride and sodium sulphate draw solutes significantly impacted growth of methanogens in An-OMBRs [29]. One approach to mitigate this problem is to utilise alternative draw solutes (Figure 4A). A draw solution selection criterion has been developed specifically for FO processes that integrate anaerobic treatment, to assess the risk of methanogenic inhibition as a result of reverse draw solute flux [27, 30]. Overall, ionic organic draw solutes such as sodium acetate (NaOAc) and ethylenediaminetetraacetic acid (EDTA) based salts hold the biggest promise. The reverse solute flux of NaOAc and EDTA-2Na are 70% and 86% lower than sodium chloride, which reduces the rate of salinity accumulation and draw solute replenishment [27, 100]. In addition, the biodegradation of these solutes can enhance biogas production [27]. To date, organic ionic draw solutes have been demonstrated in a lab-scale Ae-OMBR and have shown excellent mitigation of salinity build-up in the reactors [67, 75]. However, further research is required to assess the application of organic ionic draw solutes within anaerobic FO systems.

The high cost of ionic organic draw solutes remains an important barrier for the practical implementation of these FO draw solutions. For this reason, a number of recent demonstrations of FO integrated anaerobic systems have generally adopted cost effective sodium chloride or seawater as the draw solution and relied on non-optimal operating conditions, such as excessive sludge wastage or periodic supernatant discharge in order to avoid the effects of salinity build-up on the process [26, 28]. Although these studies present the feasibility of biogas production (i.e. 0.2-0.3 L CH_4/g COD) via the An-OMBR process, conditions are unrealistic and are not a feasible long-term solution to salinity accumulation. A proof of concept which can potentially lead to a full-scale sustainable option for salinity mitigation involves the integration of an MF membrane within an Ae-OMBR [88, 101]. The MF membrane acts as a bleeding stream since dissolved solutes can easily pass through the MF membrane (Figure 4B). This integrated system manages to sustain the FO process, whilst at the same time producing MF quality effluent for reuse applications requiring lower water qualities. Similar benefits may also be realised if MF is integrated with An-OMBR, however this approach would result in the partial loss of organic substances.

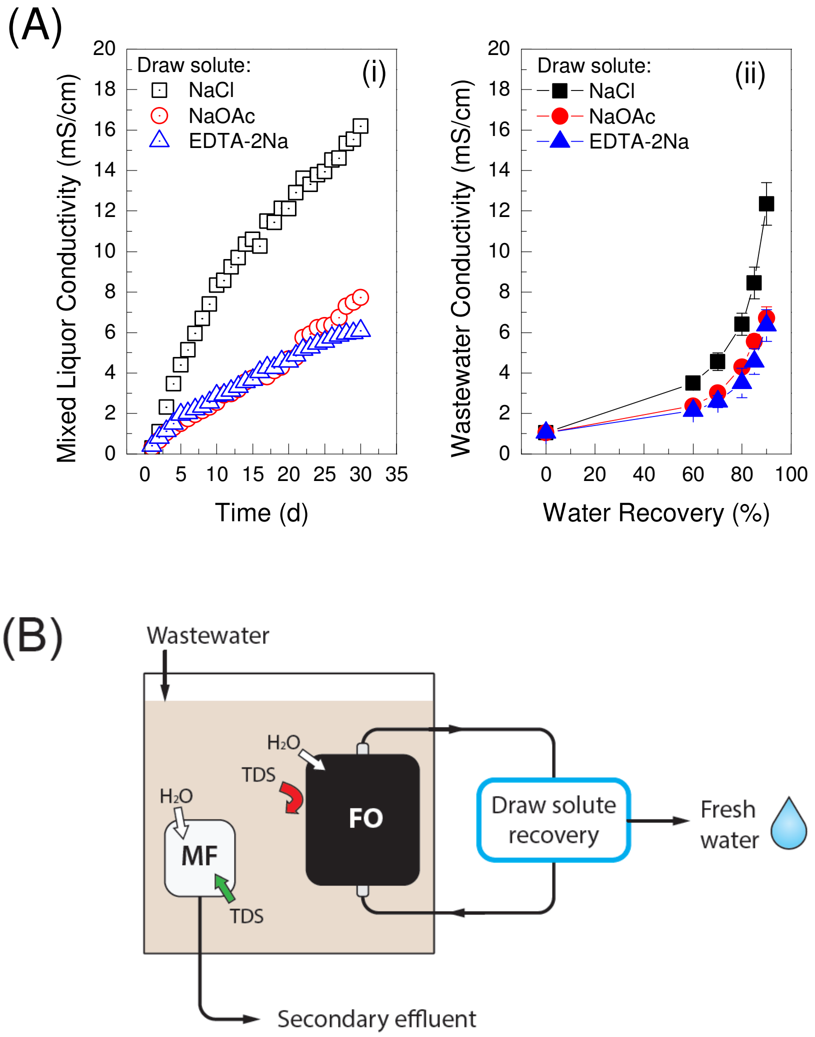

Figure 4: Mitigation of salinity accumulation by (A) alternative draw solutions for (i) Ae-OMBR [75] and (ii) wastewater pre-concentration [38], and (B) MF withdrawal of total dissolved solids (TDS) in an OMBR (adapted from Qiu et al. [88]).

Another promising approach involves acclimatising the anaerobic microbial community to saline environments. In anaerobic systems, microorganisms are able to tolerate high salt conditions if acclimated to the conditions [102, 103]. Indeed, the anaerobic treatment of high saline industrial wastewater is feasible with adequate biomass adaption or by using halotolerant organisms [104]. Further research on identifying and implementing certain halotolerant bacteria in an anaerobic system would be significantly beneficial to developing FO-based anaerobic systems. The presence of halotolerant organisms would allow the FO system to operate at a higher concentration factor. Furthermore, when draw solutions with a low reverse solute flux are applied, the negative impacts associated with salinity accumulation on biogas production would be circumvented. Overall, a greater focus is required to assess and advance the practicality of FO-based systems that integrate anaerobic treatment for biogas production. A combination of the previously mentioned strategies in a pilot-scale system would significantly contribute to assessing their long-term effectiveness, and is imperative to improving our understanding of FO-based anaerobic systems.

3.1.2 Membrane fouling

Although FO membrane fouling is readily reversible, fouling remains a pertinent issue for FO-based processes applied to complex solutions such as wastewater and mixed liquor [44, 73, 105]. During the filtration process, the accumulation of foulants on the membrane surface forms a cake layer and hinders the efficiency of the process by two prominent mechanisms. The cake layer builds hydraulic resistance and also creates the cake-enhanced concentration polarisation effect that lowers the osmotic driving force. Both these mechanisms adversely impact membrane performance, by decreasing water flux and membrane life-span [35, 44]. Various approaches have been demonstrated to manage membrane fouling. These include physical and chemical cleaning methods, as well as modification of membranes to be fouling resistant.

A key benefit of the FO process when applied for wastewater pre-concentration is the highly reversible nature of membrane fouling compared to other pressure driven membrane processes. Therefore, membrane fouling control can often be accomplished by hydraulic means, whereby hydrodynamic shear forces are introduced to prevent the accumulation of foulants near the membrane surface [106, 107]. This method is not possible when using pressure driven membrane processes for direct wastewater treatment since fouling cannot be removed without chemical cleaning. Hydrodynamic strategies including periodic rinsing at high cross flow velocities, inclusion of spacers, and air sparging via biogas recycling, which have proved effective in wastewater treatment applications [26, 106, 108]. Despite these results, the intensity of the fouling control strategy inevitably leads to heightened energy consumption. Therefore, a significant focus should be placed on evaluating and optimising the energy consumption of proposed fouling mitigation strategies. It is also necessary to develop a membrane cleaning protocol specific for intense wastewater pre-concentration applications by FO membranes.

3.1.3 Issues arising from the anaerobic treatment of FO pre-concentrated wastewater

In addition to the key challenges of salinity accumulation and membrane fouling, a range of other issues may arise as a result of the anaerobic treatment of FO pre-concentrated wastewater. Inorganic salt inhibition and ammonia toxicity may plague the efficiency of the anaerobic treatment process, regardless of mitigation strategies. In this case, the co-digestion of readily available organic substrates (i.e. food waste or industrial by-products) could significantly improve the digester efficiency [109, 110]. Furthermore, phosphorus may precipitate in the anaerobic reactor due to the enriched content of phosphorus, calcium, and magnesium in the pre-concentrated wastewater [26]. This may lead to complications for phosphorus recovery, as the availability of phosphorus in the liquid phase would be limited. However, this scenario could be easily avoided by acidifying the pre-concentrate. The conventional MF An-MBR is an ideal candidate for biogas production from the pre-concentrated wastewater. In addition, the ammonia and phosphorus rich supernatant (i.e. anaerobic digestion effluent) can be withdrawn via the MF membrane for subsequent recovery.

Studies to date have focused almost exclusively on the integration of FO and anaerobic treatment to form An-OMBRs [26, 28, 29] or to filter anaerobic effluent [111-113]. Therefore, there is a significant gap in current knowledge regarding the anaerobic treatment of FO pre-concentrated wastewater.

3.2 Nutrient recovery

The rejection of nutrients by FO membranes results in high quality product water, and can also facilitate the removal and recovery of nutrients from wastewater. Phosphorus in particular has significant environmental value and consistently presents a high rejection by FO membranes from a range of different feed solutions and operation conditions (Table 1). In recent years, there has been a significant growth in nutrient recovery research using FO-based processes [7]. Phosphorus recovery from a number of diverse source waters, including waste activated sludge [48, 49, 88], secondary treated effluent [57], digested sludge centrate [89-91], and urine [58] has been demonstrated in the literature. Several FO-based configurations have been applied including Ae-OMBR and direct FO filtration. Overall, FO is utilised to firstly concentrate nutrients, and then conventional nutrient recovery techniques are applied to chemically precipitate either struvite, or calcium phosphates (Table 2).

FO has several features that are ideal for nutrient recovery from wastewater. Firstly, FO membranes can effectively retain phosphorus, thus enriching its concentration and providing favourable conditions for phosphorus recovery. As an example, struvite recovery requires the addition of magnesium salt and ammonium to exceed the stoichiometric ratio for struvite precipitation. Thus, the phosphorus rich solution provided by the FO process improves precipitation kinetics and lowers the chemical demand (i.e. magnesium salts and caustic). Secondly, the reverse solute flux (which is usually seen as problematic in FO) can be utilised for nutrient recovery applications. Xie et al. [90] strategically utilised $MgCl_2$ as a draw solution to enrich the magnesium content of the feed solution via the reverse magnesium flux mechanism. Lastly, the bidirectional diffusion of solutes in the FO process enables the feed solution pH to naturally increase. Several researchers have observed this bidirectional transport phenomenon. In particular, Xie et al. [90] and Ansari et al. [91] have demonstrated the direct benefit of the bidirectional transport of Mg^{2+}/Ca^{2+} and proton (H^+) for struvite and calcium phosphate precipitation, respectively.

There are a number of configuration options for FO-based systems for nutrient recovery. Ae-OMBRs treating dilute wastewater have demonstrated excellent potential for nutrient enrichment within the mixed liquor or by supernatant withdrawal [49, 88, 114] (Figure 5A). Also, direct pre-concentration processes applied to anaerobic digestion effluent has presented promising results as this system could be easily integrated with current wastewater treatment infrastructure [90, 91]. In terms of nutrient recovery efficiency, the direct pre-concentration of anaerobic effluent (i.e. digested sludge centrate) is possibly the most viable approach as there is minimal loss of nutrients caused by biomass uptake, as is the case in Ae-OMBRs. In aerobic processes, nutrients are consumed or converted by activated sludge, therefore, a lower theoretical amount of phosphorus is available for recovery. Conversely, anaerobic treatment biologically releases nutrients, transforming them into more chemically available forms for precipitation (Figure 5B).

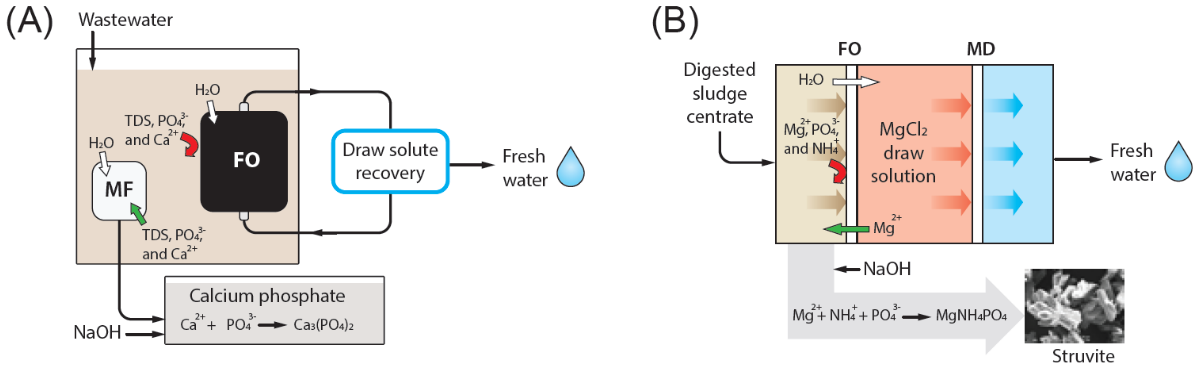

Figure 5: Phosphorus recovery using (A) MF withdrawal from Ae-OMBR mixed liquor (adapted from Qiu et al. [88]) and (B) FO-MD of anaerobically digested sludge centrate (adapted from Xie et al. [90]).

Investigations into FO performance when treating nutrient rich solutions are increasing [111-113], however there are still several key aspects to be addressed. These include membrane fouling and scaling, precipitate purification, and issues related to the market development for bio-fertilizers produced from wastewater.

Membrane scaling could be a prominent barrier for FO application to nutrient recovery; however, this issue has not been investigated. It is important to consider the possibility of membrane scaling during resource recovery as it dramatically affects process performance and chemical cleaning is often required, resulting in a decreased membrane life-span. The super saturation of phosphate minerals close to the membranes surface may lead to the precipitation of salts onto the membrane surface. Research to date has not identified any significant problems associated with membrane scaling during nutrient recovery applications. This is likely due to the short term nature of the proof of concept studies in the current literature. Pilot-scale evaluation and modelling are required to assess the risk of membrane scaling for nutrient recovering FO processes and formulation of chemical cleaning protocols.

In addition to membrane scaling, the presence of calcium and phosphate in the FO feed solution can lead to cake layer formation [115]. Nevertheless, membrane flushing has been reported to be an effective strategy to remove cake formation [90, 91].

One key advantage for nutrient recovery is the potential profit obtained from the sale of the bio-fertilizers produced. Nevertheless, a market for fertilisers sourced from wastewater is currently not well-defined. The product value largely depends on the purity of the obtained product. At this stage, product purity has not been a significant area of research for the previously mentioned FO-based nutrient recovery systems. For example, for calcium phosphate recovery, the competition of calcium and magnesium for phosphate and the presence of organic matters can drastically degrade product quality [88, 91]. There is significant potential for FO-based processes to be further integrated with established resource recovery techniques. These may include the introduction of seed crystallisation [116] or by further purification of FO pre-concentrated nutrient solutions by technologies such as ED [7].

4. Integrated FO-based wastewater treatment and resource recovery process

Based on current FO research and development, an integrated FO-based wastewater treatment and resource recovery process is proposed and compared with current wastewater treatment practices (Figure 6). Current wastewater treatment (Figure 6A) is highly energy intensive, with aeration and pressurised membrane systems being significant energy consumers. The process also focusses strictly on water reclamation and does not effectively integrate energy and nutrient recovery practices. Although sludge is often anaerobically treated, a large portion of the chemical energy in wastewater is dissipated by the initial aerobic biological process [18].

Unlike current wastewater treatment practice, the proposed FO-based process (Figure 6B) focuses on the separation of water and non-water components to enable more efficient resource recovery. In this process, primarily treated effluent is firstly filtered by the FO process coupled with MD to produce high quality effluent for reuse. Organic ionic draw solutes are employed to minimise reverse draw solute flux, and to lower the risk of methane inhibition during anaerobic digestion. The FO pre-concentrate is fed to an anaerobic digester to produce biogas. A combined heat and power system converts biogas to useful heat for operating MD, and electricity for treatment operations. Furthermore, nutrient rich anaerobic effluent is processed by an FO-MD system to further harvest valuable nutrients for subsequent recovery. Struvite recovery can be achieved using MgCl as the draw solution [90], whilst calcium phosphate can be recovered using seawater [90]. This MD system would also produce high quality effluent for reuse, which is a significant benefit, as anaerobic effluent is commonly returned to the headworks in conventional treatment plants. For these reasons, FO can potentially serve as a game changer in municipal wastewater treatment.

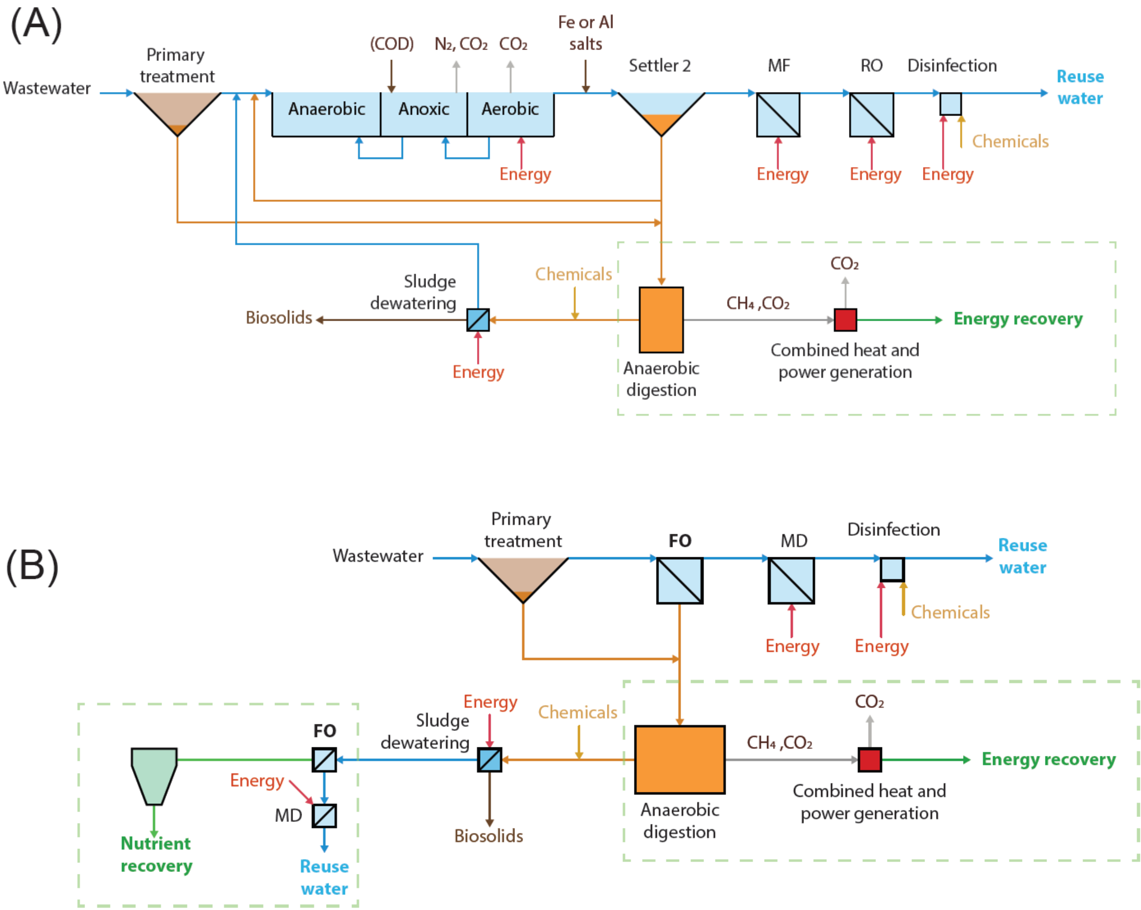

Figure 6: Comparison of current and FO-based wastewater treatment technologies. (A) Current processes consume significant energy, dissipate wastewater organic matter, and do not effectively manage nutrients (adapted from Verstraete et al. [16]). (B) The proposed FO-based treatment process achieves simultaneous wastewater treatment and resource recovery, utilising produced energy within its operations and recovers nutrients.

5. Outlook

FO-based processes have a proven capability and offer a unique opportunity to achieve simultaneous wastewater treatment and resource recovery. Yet, FO technology is still in the early stage of development and therefore the realisation of full-scale implementation will continue to evolve as the field becomes more mature. Two important considerations for this concept include the applicability of FO-based systems to a decentralised or centralised level and economic barriers that strongly affect the acceptance of the technology.

Issues regarding the scale-up of FO based processes involve the inherently low water flux of the FO process. Low water flux corresponds to a large footprint which substantially increases capital and operational costs. Considering the direct filtration of raw wastewater by FO, with the current state of FO membranes, environmental and economic benefits may only be realised for decentralised applications. This is due to the significantly large volumetric loading of centralised wastewater treatment systems in urban areas. Furthermore, there is an increasing drive to house treatment facilities onsite or nearby to the water reuse locations (i.e. farming areas or industrial areas) [117]. This concept of sewer mining strategically avoids the energy needed to convey reuse water from a centralised wastewater treatment plant, however quality control would be an added issue to be addressed. Further investigations to assess the feasibility of FO scale-up must be conducted in terms of both technical and economic viability. In the future, improvements of FO membrane materials, module design, draw solutions, and draw solute recovery processes may provide practical opportunities for the scale-up of FO systems at a centralised level.

Regarding nutrient recovery using FO-based technology, important advantages are likely to be realised sooner as the process can be integrated with current wastewater treatment infrastructure (i.e. treating anaerobically digested sludge centrate). Furthermore, nutrient recovery presents a practical business case for struvite blockage prevention, phosphorus effluent discharge compliance, and fertilizer production potential [118]. In fact, struvite recovery has been demonstrated at several full-scale wastewater treatment plants in North America [119]. We envisage that FO can greatly improve the process efficiency and therefore break-down some of the economic barriers that prevent nutrient recovery being an established practice [120].

Resource recovery from municipal wastewater presents a promising outlook for a number of contemporary environmental challenges. However, several economic barriers exist and restrict the acceptance and implementation of such practices. The environmental value of water, energy, and nutrient resources cannot be readily captured by current economic analysis. This is illustrated by the availability of low cost electricity, natural gas, and mineable phosphorus that strongly resist investment appeal. Furthermore, the lack of a well-defined market for saleable bio-fertilizers remains may influence the acceptance of nutrient recovery technologies. Nonetheless, resource recovery from wastewater represents a renewable source of water, energy, and nutrients. Particularly when considering how population growth and urbanisation will continue to stress non-renewable resource reserves in the future. The introduction of government incentives may provide a profound milestone in implementing resource recovery practices. Further investigations into the economic feasibility of technologies that enable resource recovery from wastewater should be a high priority.

6. Conclusion

The FO process is a favourable avenue to advance a membrane-based platform to achieve simultaneous wastewater treatment and resource recovery. FO membranes can be applied to a complex and high fouling solution and retain a wide range of contaminants. FO membrane-based hybrid systems that combine FO with a draw solute recovery process (i.e. MD) effectively enable fresh water recovery from wastewater. Extending this effort, energy and nutrient recovery from wastewater can be initiated through the strategic integration of FO with anaerobic biological treatment. FO membranes can successfully pre-concentrate wastewater and improve the organic loading rate of anaerobic treatment systems for biogas production. Similarly, the FO process can harvest the valuable nutrients within anaerobic effluent, and significantly benefit the efficiency of established phosphorus recovery techniques.

Despite the potential of FO to emerge as an important membrane technology in the future, several major technical challenges still remain. These include contaminant accumulation in the draw solution, salinity accumulation, membrane fouling, and anaerobic system integration. A number of innovative approaches can be utilised to resolve these challenges as highlighted in this review. Further development of the practical aspects of this concept via pilot-scale demonstrations is recommended. One major milestone in the development of FO technology for this application involves the successful demonstration of integrated FO and anaerobic treatment systems. Furthermore, energy considerations for the proposed process must also be clearly dictated through techno-economic assessments that address the likely advantages of the process compared with current technologies. Issues associated with the scale-up of FO-based processes at a decentralised or centralised level must also be addressed. Development of FO membrane materials and anaerobic microbial selection techniques are expected to strongly benefit research progress towards FO-based technology for simultaneous wastewater treatment and resource recovery.

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