Chapter

Integration of Forward Osmosis in Municipal Wastewater Treatment Applications

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Abstract

In recent years, the research community has made constant efforts to develop new technologies for the recovery and valorization of water, nutrient and energy content of municipal wastewater. However, the recovery process is significantly limited due to the low-strength of sewage. Over the last 10 years, the Forward Osmosis (FO) process, has gained interest as a low-cost process with low membrane fouling propensity, which can convert municipal wastewater into a concentrated low-volume effluent, characterized by high organic and nutrient concentration. This chapter presents the main configurations that have been implemented for the concentration of municipal wastewater using FO, including their performance in terms of contaminant removal and water/reverse salt flux (J_w/J_s). Furthermore, the draw solutions and respective concentrations that have been used in FO for the treatment of sewage are reported, while at the same time the positive and negative characteristics of each application are evaluated. Finally, in the last section of this chapter, the spontaneous FO followed by anaerobic process is integrated in a municipal wastewater treatment plant (WWTP) and compared with a conventional one. The comparison is done, in terms of the mass balance of the chemical oxygen demand (COD) and in terms of the energy efficiency.

Keywords: forward osmosis, municipal wastewater, configurations, draw solution, COD mass balance

1. Introduction

Water scarcity is one of the most serious threats which our planet faces [1]. Globally, water demand is predicted to increase by 35% more than sustainable supply by 2040/50, if the linear water management model continues to be implemented [2]. The European Union (EU) encourages the implementation of a circular economy model, through its strategy called "Closing the loop—a EU action plan for the Circular Economy" in 2015 and European citizens must seize the opportunity to close the loop of water, resource and energy management [3]. Among various types of water, seawater and wastewater are two alternative sources, which are readily available, especially in coastal, arid areas [4]. Both need to be treated before they can be rendered suitable for use. Membrane processes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO) are particularly effective in the purification of non-conventional water sources and count many applications in both the wastewater and the desalination sector [5]. In particular, the RO, holds a prominent position in water desalination, compared to traditional thermal desalination processes [6]. As high energy consumption is required to overcome the osmotic potential, reverse osmosis is not applied in many water-stressed areas [5].

Forward osmosis is one of the most attractive membrane-based processes that requires two solutions of different osmotic concentrations (high and low), separated by a semi-permeable membrane to be realized [7]. Water molecules are spontaneously diffused from the low osmotic potential solution (feed side) to the high osmotic solution (draw solution or DS), to equalize the concentration difference, while the semi-permeable membrane acts as a barrier that rejects the salts and contaminants [8]. The natural osmotic pressure of FO makes it stand out from conventional RO, by offering high water recovery, reduced membrane fouling potential, greater effectiveness, low cost, and reduced energy demand [8, 9]. All these positive aspects have led to a notably high trend of publications on FO applications in various water sources, such as seawater and wastewater, with more than 97.5% of publications since 2009 [10]. Among them, several researchers investigate the feasibility of integrating the FO process in a novel sewage treatment system based on the circular economy concept, as the main goal is to valorize the chemical energy, water, and nutrients of sewage. This innovative application of FO and its combination with appropriate downstream technologies is really promising. As the results show, the wastewater is converted into a small volume liquid, characterized by a high concentration of organic matter, as it can be concentrated up to 8–10 times, while the recovery of phosphorus can reach up to 90%, replacing the need for chemical fertilizers [11, 12]. However, there are many challenges that need to be overcome for this application, the most important of which is the selection of the most appropriate DS, which despite the significant efforts has not been found to date [13–15].

This chapter presents the main configurations that have been implemented to concentrate municipal wastewater using FO, including their performance in terms of contaminant removal and J_w/J_s. The draw solutions and their concentrations that have been used in the FO process for the treatment of sewage are reviewed, while at the same time the positive and negative characteristics of each application are evaluated. Finally, in the last section of this chapter, the spontaneous FO followed by an anaerobic process is integrated into a municipal wastewater treatment plant and compared with a conventional activated sludge process (CAS), in terms of COD and corresponding energy efficiency, emphasizing the key impact of the FO in the latter process.

2. Forward osmosis configurations and performance in municipal wastewater management

The main benefit of the FO process in municipal wastewater treatment is that it converts sewage from low-strength liquid to a concentrated bulk, which consists of high a concentration of organic matter and nutrients [16, 17]. According to Korenak et al. [18], the FO process is characterized by high membrane fouling reversibility, while it can significantly minimize space requirements in a municipal WWTP. Considering all the above, three basic configurations have emerged for the integration of the FO process in the municipal WWTPs, which are illustrated in **Figure 1**.

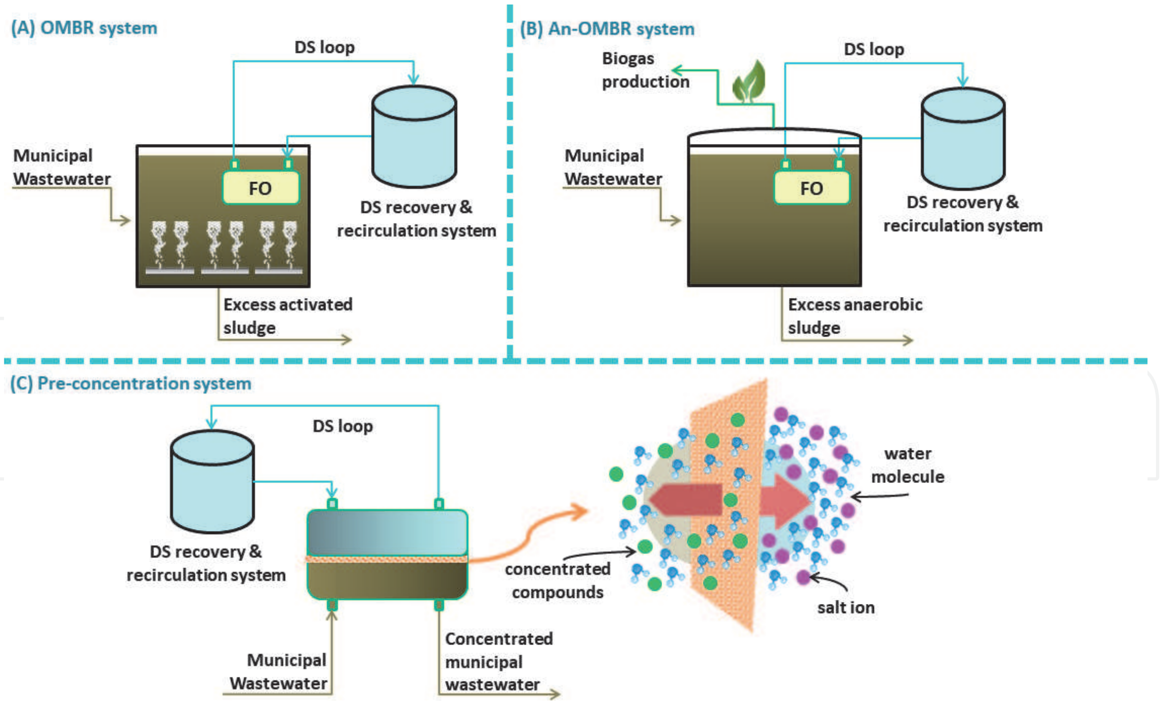

Figure 1.
Configurations for the integration of the FO process in the municipal WWTPs.

2.1 Osmotic membrane bioreactor (OMBR)

In 2008, an innovative system was introduced, in which FO membranes were submerged into a typical membrane bioreactor (MBR) module; this system was called OMBR (**Figure 1(A)**) [19]. The replacement of UF or MF membranes in the conventional system by FO membranes resulted in better performance in terms of contaminants' rejection (79.7–100% of COD, **Table 1**). In addition, the absence of hydraulic pressure contributed to lower fouling tendency and probably lower energy requirements. Despite the benefits of OMBR over traditional systems, two major challenges are still under investigation; low J_w rate and salinity accumulation [19, 31]. The findings confirm that the decline in J_w was greatly affected by the salt accumulation, even with the implementation of improved membrane materials, such as thin-film composite (TFC), achieving an average rate equal to 3.9 ± 0.5 L m^{-2} h^{-1} [32]. In addition, the microbial community of the reactor can either be partly or fully inhibited, due to the gradual building-up of salts, which occurs due to the J_s [31, 32].

2.2 Anaerobic OMBR (An-OMBR)

The combination of MBR technology with the anaerobic process has been extensively investigated in the last 10 years, due to the environmental benefits of both [33]. However, the low-strength nature of sewage is a major obstacle to the effective application of the anaerobic process in municipal WWTPs; containing a high amount of water with low organic and nutrients concentration. Due to the methane's solubility in water (22.7 mg L^{-1}, at room temperature), a large part of the produced gas escapes with the treated effluent of the anaerobic process (ranges between 20 and 60%) [34]. Due to the aforementioned barriers, it is difficult to implement anaerobic processes for municipal wastewater treatment particularly in areas, where the sewage temperature drops below 15°C, during the winter period. The incorporation of FO, either as a pre-treatment step or submerged into the MBR system, significantly enhances the resource recovery potential in the anaerobic

Membrane Type	Feed	Draw	Removal Efficiency (%)					Jw (L m^{-2} h^{-1})/Js (g m^{-2} h^{-1})	Ref.
			TOC	COD	TP	NH_4-N	TN		
CTA (HTI, USA)	Municipal wastewater	Seawater brine	—	90	97.9	99	—	N.A.	[12]
CTA (HTI, USA)	Synthetic municipal wastewater	1 M NaCl	98 ± 1	—	—	99 ± 1	—	8–5/NA.	[20]
TFC (HTI, USA)	Synthetic municipal wastewater	1 M NaCl	96 ± 1	—	—	99 ± 1	—	3/N/A	[20]
CTA (HTI, USA)	Municipal wastewater	42 g L^{-1} NaCl	—	99	99	—	82	4–4.5/ N.A.	[21]
CTA-ES (HTI, USA)	Sunthetic municipal wastewater	48.4 g L^{-1} $MgCl_2$	98	—	—	98	—	3.7–3.3/N.A	[22]
CTA (HTI, USA)	Raw municipal wastewater	Synthetic seawater	79.7 ± 9	—	92 ± 3.3	88.1 ± 5.5	—	4.86–3.24/ N.A	[23]
CTA-ES (HTI, USA)	Sunthetic municipal wastewater	48.4 g L^{-1} $MgCl_2$ & 49 g L^{-1} NaCl	98	—	98 (PO_4^{3-}-P)	98	—	6.64–8.95 (NaCl); 6.46 ($MgCl_2$)/ NA.	[24]
TFC (HTI, USA)	Sunthetic municipal wastewater	1 M NaCl	100	—	95.6 (PO_4^{3-}-P)	43		14/ N.A.	[25]
CTA (HTI, USA)	Raw sewage	0.5 M NaCl	95	—	95	95	90	1.7/ NA.	[26]
CTA & TFC (HTI, USA), Biomimetic (Aquaporin, Denmark)	Synthetic wastewater	0.5 M NaCl.	>95	—	—	90–100	60–80	15.6–15 (TFC); 5.5 (CTA)/0.085 (Aquaporin); 82.7 (TFC); 5.5 (CTA) mmol m^{-2} h^{-1}	[27]
CTA-ES (HTI, USA)	Synthetic wastewater	1 M $MgCl_2$	—	—	99	99	—	5.45/ N.A.	[28]
TFC (Aquaporin A/S, Asia)	Synthetic wastewater	1.2 M NaCl with SDBS	100	—	>90	85	50–80	10/ N.A	[29]
TFC (Aquaporin A/S, Asia)	Synthetic wastewater	1.2 M NaCl without SDBS	98	—	>90	>90	60–100	<10/ N.A.	[29]
CTA-ES (HTI, USA)	Synthetic municipal wastewater	70 g L^{-1} NaCl	—	99	99 (PO_4^{3-}-P)	92	—	10.42–6.4/9.9–34.99	[30]

Table 1.
Osmotic Membrane Bioreactor applications to treat municipal wastewater.

process. Compared to OMBR, An-OMBR (**Figure 1(B)**) is characterized by significantly lower energy requirements, due to the replacement of energy-demanding aeration, while biogas production contributes to the coverage of specific energy needs. According to Zhang et al. [35], due to the 2–3 times smaller pore size of the FO membranes over conventional UF or MF membranes, the dissolved methane content in the An-OMBR treated effluent was eliminated, even as a function of different operating parameters. Regarding the yield of methane, Zhang et al. [35] and Gu et al. [36] observed particularly satisfactory production that reached 0.256 L CH_4 g^{-1} COD and 0.25–0.3 L CH_4 g^{-1} COD at mesophilic conditions, respectively. In addition, anaerobic biomass showed high resistance to increasing salt concentrations and was not affected even when the concentration was equivalent to 200 mM sodium chloride (NaCl) [36]. As shown in **Table 2** the FO membranes achieve high rejection of contaminants; specifically the Total Organic Carbon (TOC)/COD and PO_4-P removal was 95% and 73%, respectively. However, due to the lack of ammonia removal, its accumulation has been observed in the reactor, but not in concentrations that can lead to the interruption of the anaerobic process [36]. In recent years, an alternative configuration has been proposed, which includes the addition of MF membranes both to OMBR and An-OMBR systems, the so-called Microfiltration- Osmotic Membrane Bioreactor (MF-OMBR). The main goal of this hybrid system is 1) to balance the salts concentration in the reactor so as to prevent an inhibition event and 2) to apply resource recovery methods to its nutrient-rich

Membrane Type	Feed	Draw	Removal Efficiency (%)					Jw ($L\ m^{-2}\ h^{-1}$)/Js ($g\ m^{-2}\ h^{-1}$)	Ref.
			TOC	COD	TP	NH_4-N	TN		
CTA (HTI, USA)	Synthetic municipal wastewater	0.5 M NaCl	—	95	99	70–80	—	10–3/ N.A.	[36]
CTA (HTI, USA)	Synthetic municipal wastewater	0.5 M NaCl	—	96	100	62	—	9.5–3.5/ N.A.	[37]
TFC (Aquaporin Denmark)	Synthetic municipal wastewater	116.6 g L^{-1} $MgSO_4$	—	>95	95 (PO_4^{3-}-P)	>95	—	0.78–0.26/ NA.	[38]
CTA (HTI, USA)	Synthetic domestic wastewater	0.5 M NaCl	>96	—	—	—	—	6–3.4/ N.A.	[39]
CTA-ES (HTI, USA)	Municipal sewage	0.5 M, 1 M and 1.5 M NaCl	—	96	100	88	89	6 (0.5 M); 10 (1 M); 13 (1.5 M)/ 4.26 (0.5 M); 7.65 (1 M); 11.84 (1.5 M)	[40]
CTA-NW (HTI, USA)	Synthetic municipal wastewater	2 M $C_4H_6MgO_4$	—	96	73 (PO_4^{3-}P)	51.4	—	3.5–1.09/ 2.5–1.6	[35]
TFC (HTI, USA)	Synthetic municipal wastewater	0.5 NaCl	—	>95	99 (PO_4^{3-}P)	0	—	12 ± 0.7–2 ± 0.2/ N.A.	[41]
CTA (HTI, USA)	Synthetic sewage	0.5 NaCl	—	>93	99	28–45	—	8.7 ± 0.3–4.0 ± 0.2/ N.A.	[42]

Table 2.
Anaerobic Osmotic Membrane Bioreactor applications to treat municipal wastewater.

treated effluent. Nonetheless, according to Wang et al. [39], the FO membranes achieved much lower ammonia rejection rates (39–50%) compared to an An-OMBR system (62.7–81.2%), while the addition of another membrane significantly raises both the maintenance and the investment cost of the entire system [39, 43].

2.3 Pre-concentration with FO

Alternatively, the FO unit can be applied as a pre-condensation step in municipal WWTPs (**Figure 1(C)**), achieving a similar goal to the previously analyzed configuration, as it can be combined by suitable downstream processes for resources and energy utilization. As reported by Ansari et al. [34], the submerged FO configuration is significantly disadvantaged compared to the separate one, as the former gets in contact with the dense activated sludge, while the latter with the diluted primary treated effluent. In contrast, a recent study that examined both approaches in parallel, direct osmosis showed a significant decline in J_w performance compared to OMBR system [7]. On the other hand, a prolonged biodegradation study (approximately 7 months) of both cellulose triacetate (CTA) and TFC membranes demonstrated that the long-term exposure to activated sludge significantly affects their performance, in terms of water permeability and J_s [44]. Sun et al. [7] found that the direct FO module is characterized by reversible membrane fouling over the submerged OMBR membrane, mainly due to the lower abundance in the microbial load of the feed solution. In terms of performance, as shown in **Table 3**, this FO configuration achieves the retention of organic load by a percentage ranging from 71.9 to 100%. At this point, it should be noted that based on the current literature most studies refer to FO as either a separate or integrated system of an Anaerobic Membrane Bioreactor (An-MBR), while alternative anaerobic treatment systems are not frequently investigated.

3. Draw solutions

In contrast to other osmotic, membrane-based technologies, the application of high osmotic potential is the driving force in the spontaneous FO process [52]. Therefore, the selection process of the most effective solution acts as a cornerstone of the FO and plays a crucial role on its performance as well as on downstream processes [15]. In an ideal physicochemical context, the parameters listed in **Table 4** must be met to classify a solution as appropriate [52–54].

In recent years, significant efforts have been made by researchers to combine the above parameters and develop an ideal DS, which will be compatible with the application of FO in the municipal wastewater treatment sector [15, 55, 56]. Alternative systems have been developed; different configurations have been applied to integrate the FO in several stages of a municipal WWTP; as pre-treatment, secondary and post-treatment steps for nutrient recovery. Obviously, the treatment level and the quality-target of the recovered product must be considered in the DS selection process [57]. First on the list and most commonly used as DS is NaCl, even in high concentrations up to 4 M, due to its high aqueous solubility, small molecular size, high availability, and relatively low cost [58]. As shown in **Table 5**, the 0.5 M concentration is most frequently applied, as it simulates the osmotic pressure of seawater [53]. The ultimate goal is to adopt a circular solution, by applying an abundant water source without any economic burden or a process' by-product, such as the RO brine as DS (**Table 6**) [58, 65]. High rejection rates of TOC/COD and PO_4-P have been reported using NaCl as DS in OMBR systems, equal to 100% and 95.6%, respectively, although the same is not achieved for ammonium nitrogen

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Membrane Type	**Feed**	**Draw**	**Removal Efficiency (%)**					**Jw ($L\ m^{-2}\ h^{-1}$)/Js ($g\ m^{-2}\ h^{-1}$)**	**Ref.**
			TOC	**COD**	**TP**	**NH_4-N**	**TN**		
CTA (HTI, USA)	Raw Sewage	3.5% NaCl	—	71.9 (AL-FS); 69.3 (AL-DS)	—	—	—	5.2 (AL-DS) and 5.4 (AL-FS)/ N.A.	[45]
CTA (HTI, USA)	Municipal wastewater	0.5 M NaCl	—	99.8 ± 0,6	99.7 ± 5	48.1 ± 10.5	67.8 ± 7.3	7.7–6.5/ 5	[17]
TFC, (Aquaporin, Denmark)	Domestic sewage	1, 1.5 & 2 M $MgCl_2 \cdot 6H_2O$	—	76–80	75	66	—	5.11 (1 M); 6.66 (1.5 M); 6.92 (2 M)/ N.A.	[16]
CTA (HTI, USA)	Sewage	0.2–4 M NaCl	—	96.5	95.4	93.3	96.5	10 (0.5 M) - 25 (4 M)/ N. A.	[46]
CTA (HTI, USA)	Municipal wastewater	Synthetic seawater	79.9 ± 6.7	—	93.3 ± 3.3	85.4 ± 5.6	—	5.37–2.5/ N.A.	[23]
TFC (Toray, Japan)	Municipal wastewater	11.7 g L^{-1} NaCl	—	85.5	90	no rejection (AL-FS); 50 (AL-DS)	—	5.1 ± 1 / 4.8 ± 8.6	[47]
Aquaporin TFC (Sterlitech Corporation, WA)	Synthetic wastewater	Synthetic seawater with *C. vulgaris*	—	100 ± 0.6	99.5 ± 0.5	46.1 ± 3.4	—	11.59 ± 0.49	[48]
Aquaporin TFC (Sterlitech Corporation, WA)	Synthetic wastewater	Synthetic seawater without C. vulgaris	—	99.5 ± 0.7	99.1 ± 0.1	46.2 ± 2.6	—	12.02 ± 0.35	[48]
TFC (HTI, USA)	Municipal wastewater &prefiltered	Synthetic seawater brine	—	2.38–2.67 CF	3.3–3.5 CF	1.31–1.75 CF	1.58–1.94 CF	19.90 (raw sewage); 18.15 (filtered)/ N.A.	[49]
CTA (HTI, USA)	Synthetic municipal wastewater	4% NaCl	—	99	—	67–68	56–59	N.A.	[50]
CTA-ES (HTI, USA)	Synthetic municipal wastewater	0.25 M $(NH4)_2SO_4$ (SOA), KH_2PO_4 (MKP),$(NH_4)H_2PO_4$ (MAP)	—	98	99	89,56 ± 1.52 (SOA); 98.96 ± 0.33 (MPK); 89.67 ± 1.27 (MAP)	—	2.58/ 0.57 (SOA); 2.11/ 1.17 (MPK); 1.97/ 0.11 (MAP)	[51]

Table 3.
Pre-concentration of municipal wastewater using FO.

Parameter	Impact
Osmotic Pressure	DS with higher osmotic pressure than the feed generates higher J_w
Water Solubility	Soluble compound produces higher osmotic concentration and therefore retains J_w and water recovery at higher percentages
Concentration	Higher concentrations contribute to higher J_w rates, but particularly high concentrations inhibit flux's increase, as mass transfer phenomena occur, such as concentration polarization
Diffusion, viscosity and molecular weight	Small molecules are distinguished by high aqueous solubility and high osmotic pressures, but in comparison with the large ones they are characterized by a higher diffusion coefficient and J_s
Toxicity and degradation	Low reverse salt diffusion to minimize the risk of toxicity and contamination of downstream systems and the recovered product. Low rate of degradability, unless it is beneficial
Cost-effective	Easily re-concentration at competitive cost
Availability	Available in large quantities with low price and easy handling

Table 4.
Main parameters that characterized the ideal DS.

(NH_4-N), which in most studies ranges between 43 and 90% [25, 59]. Nevertheless, the biggest challenge in OMBR systems using NaCl as DS is the accumulation of salts in the concentrated stream of mixed liquor and the subsequent negative effect on bacterial growth, due to reverse sodium leakage [12]. Relevant mitigation measures of the above obstacles have been proposed, such as the reduction of sludge retention time (SRT), but also the application of hybrid solutions, such as MF and UF membranes downstream for the parallel recovery of phosphorus [32].

Similar results are demonstrated in bench and pilot scale FO systems for the pre-concentration of municipal wastewater using NaCl. The bidirectional diffusion of monovalent ammonium ions from the feed to the sodium cations of DS remains a major drawback [17]. In a recent study, Yang et al. [49] demonstrated the effect of the pH parameter on low NH_4-N rejection rates and suggested a functional range of less than 8 for optimized performance. More specifically, at elevated pH as the main form of ammonium nitrogen is ammonia, diffusion becomes independent of the reverse sodium leakage [49]. Alternatively, the application of divalent molecular compounds as DS (**Tables** 7 & 8), such as magnesium chloride ($MgCl_2$) and magnesium sulfate ($MgSO_4$), which are characterized by lower reverse salt transport than NaCl, is suggested in many investigations [16]. Another superiority of inorganic solutions containing Mg ions is their combination with MF-OMBR hybrid systems and the utilization of the reverse Mg flux in the mixed liquor to nutrients' recovery, after proper pH adjustment. Although, a comparative study demonstrated that Mg transport leads to the formation of both organic and inorganic fouling in the active and support layer of the TFC membrane, correspondingly, causing a dramatic reduction in membrane flux [56]. As shown in **Table 8**, a highly charged compound, ethylenediamine tetraacetic acid disodium salt (EDTA 2Na) was applied as DS to remove the water from the activated sludge in a hybrid Forward Osmosis – Nanofiltration (FO-NF) system; the NF module was used for the recovery of DS. Water flux dropped rapidly after 8 operating hours (8.45 to 4.22 L m^{-2} h^{-1}), mainly due to the reduction of the osmotic driving force and the formation of a cake layer on the membrane surface. It is worth noting that the reverse salt flux was equal to 0.2 g m^{-2} h^{-1}, while suspended solids were concentrated from 8 g L^{-1} to 32 g L^{-1} [75].

Draw	Feed	Configuration/ Membrane Type	Findings	Ref.
0.5, 1 & 1.5 M NaCl	Municipal Wastewater	An-OMBR/CTA-ES (HTI,USA)	• Salinity accumulation, 1 M NaCl was advantageous	[40]
0.5 M NaCl	Municipal Wastewater	Direct FO/ Spiral Wound CTA (HTI, USA)	• High contaminant rejection rates, except for ammonia	[17]
0.5 M NaCl	Synthetic Wastewater	OMBR/ CTA (HTI, USA)	• Accumulation of nutrients and salts in the OMBR	[59]
3.5 g L^{-1} NaCl	Domestic wastewater	Direct FO (coupled with MD)/ CTA (HTI, USA)	• Fouling of organic substances such as proteins and polysaccharides, resulting in reduced J_w	[60]
0.5 M NaCl	Synthetic municipal wastewater	OMBR/ Biomimetic (AQUAPORIN Asia, Singapore) TFC & CTA (HTI,USA)	• Aquaporin FO membrane showed better performance than CTA & TFC, in terms of salinity accumulation	[27]
3 M NaCl	Synthetic wastewater	Post FO/ TFC-ES (HTI, USA)	• J_w decline due to the dense adsorption layer and the gel layer formed by the deposition of carbohydrates and proteins	[61]
30 g L^{-1} NaCl	Algae effluent	Algae - hybrid FO-RO system/ TFC Porifera Inc. (California, USA)	• The FO rejected organic, multivalent cations and anions, providing an effective pretreatment for the RO system	[62]
0.2, 0.5, 1, 1.5, 2, 3 & 4 M NaCl	Municipal wastewater	Direct FO/ CTA-ES (HTI,USA)	• Disproportionate concentration between contaminants and water	[46]
			• Low fouling propensity, with layers formed by humic acid, protein, and polysaccharide	[46]
1 M NaCl	Synthetic municipal wastewater	OMBR/ TFC (HTI, USA)	• NH_4-N rejection rate was low	[25]
			• The water flux decreased with increasing salinity in the mixed liquor	[25]
70 g L^{-1} NaCl	Raw anaerobic centrate	Direct FO-RO/CTA (HTI,USA)	• FO/RO system affected by centrate replenishment and concentration; it decreased with increased replenishment and concentrations	[63]
0.7 g L^{-1} NaCl & 0.7 g L^{-1} Na_2SO_4	Synthetic wastewater	AnFOMBR/ CTA (HTI, USA)	• NaCl recorded better methane ratio in the biogas produced	[64]
53 g L^{-1} NaCl and Industrial effluent (consisted of SO_4^{2-} and NH_4-N)	Synthetic wastewater	OMBR/ CTA-NW	• Industrial wastewater had higher J_w and less membrane fouling compared to NaCl, but, a higher J_s was observed in the former	[65]
0.5 M NaCl & NaCl with 0.01 M $((NH_4)_2HPO_4)$	MBR permeate	FO-RO/TFC, FTSH2O (Sterlitech Company, USA)	• The enhanced DS with DAP recorded higher J_w than NaCl Higher rejection of TP than NH_4-N	[66]

Table 5.
Sodium chloride as DS in FO treating municipal wastewater.

Draw	Feed	Configuration/ Membrane Type	Findings	Ref.
Brine	Primary municipal effluent & raw municipal wastewater	Direct FO/ homemade TFC	• J_w depends on temperature, high viscosity at low temperatures increase ICP effect • Reversible fouling after physical cleansing	[49]
Synthetic seawater	Synthetic feed and MBR permeate	Direct FO/ TFC, CTA-ES and CTA-NW, (HTI,USA)	• Concentration of nutrients successfully performed • pH plays a key role in ammonia rejection by the FO membranes (close to 7)	[67]
Brine and industrial effluent (mainly consisted of SO_4^{2-} and NH_4-N)	Anaerobically digested sludge centrate	Direct FO/ CTA-NW (HTI,USA) and Aquaporin (AQUAPORIN A/S, Denmark)	• 2 industrial effluents successfully implemented as DS in the FO process • Increased NH_4-N concentration in concentrated sludge due to application of DS rich in ammonium sulfate	[68]
Seawater	Anaerobically digested sludge centrate	Direct FO / TFC, Porifera, Inc. (California, USA)	• Extensive membrane fouling due to nutrient precipitation in both feed solution and membrane • Filtration time plays an essential role in process performance	[69]
Synthetic seawater & brine	Wastewater after a hydrolytic anaerobic reactor	Direct FO / TFC Porifera Inc. (California, USA)	• With seawater & brine the condensation factor can reach over 10 • Inevitable biodegradation of VFAs in this environment	[70]
Synthetic seawater with algae strain	Synthetic municipal wastewater	Direct FO / Aquaporin TFC (Sterlitech Corporation, WA)	• Low ammonium rejection, low removal of ammonia after the application of algae at about 35%.	[48]
Seawater (0.599 M, 0.428 M & 0.770 M NaCl solutions)	Anaerobically digested sludge centrate	Direct FO / Aquaporin (Aquaporin A/S, Denmark)	• Better performance at pH <9 and application of DS with low reverse salt	[63]
Brine	Raw municipal wastewater	MF-FOMBR/ CTA (HTI, USA)	• 90% recovery of phosphorus using the MF system • Accumulation of salts in the bioreactor is still a challenge	[12]

Table 6.
Seawater, Brine, and industrial effluents as DS in FO treating municipal wastewater.

To enhance the valorization of the resources contained in municipal wastewater, through the application of the anaerobic process several organic and ionic organic draw solutions have been investigated [13, 14, 74]. Bowden et al. [14] compared 10 different ionic organic compounds as DS and slightly altered the selection methodology proposed by Achilli et al. [15], introducing the parameter of biodegradability

Draw	Feed	Configuration/ Membrane Type	Findings	Ref.
0.5, 1, 1.5 & 2 M $MgCl_2$	Synthetic Secondary effluent	Post FO / CTA-NW and CTA-ES (HTI,USA)	• Cl diffusion was higher compared to Mg ions. 95% rejection of nutrients using 2 M $MgCl_2$ • Higher diffusion of Cl (about 3 times) by applying CTA-ES membrane	[71]
3000:1; 1500:1: 1000:1 $MgCl_2$ &Triton X-144	Synthetic domestic wastewater	SMB-OSMBR/ CTA-ES (HTI, USA)	• 1.5 mM $MgCl_2$ and 1.5 mM Triton X-114 was the best solution ratio in terms of performance • Biomass growth media favored the achievement of stable J_w and low membrane fouling	[72]
1 M $MgCl_2$	Synthetic municipal wastewater	OMBR-MD/ CTA - ES (HTI, USA)	• Successful rejection of PO_4-P and NH_4-N & recovery in the form of struvite • Recovery of the DS with the MD system with a small drop in the J_w	[28]
1 M $MgCl_2$	Raw anaerobic centrate	Direct FO-MD/ CTA (HTI,USA)	• High rejection of nutrients, reversible membrane fouling, potential recovery of struvite with the application of FO-MD system	[73]
48.4 g L^{-1} $MgCl_2$ & 49 g L^{-1} NaCl	Synthetic municipal wastewater	OMBR, CTA-ES (HTI,USA)	• Membrane fouling was not severe as shown by the decline in J_w with both NaCl and $MgCl_2$ • Accumulation of salts was observed with the use of both solutions	[24]
0.5 M NaCl & 0.35 M $MgCl_2$	Synthetic municipal wastewater	AnOMBR-MF system/ TFC (HTI,USA)	• Zero rejection of NH_4 –N using NaCl and 57.5–87.5% using $MgCl_2$ • $MgCl_2$ caused severe membrane inorganic fouling	[56]
1, 1.5 & 2 M $MgCl_2$	Municipal wastewater	Direct FO, TFC (Aquaporin A/S, Denmark)	• Both the increase in the $MgCl_2$ concentration and increase in the cross-flow rate contributed to the higher J_w, but COD concentration remained stable	[16]

Table 7.
Magnesium chloride as DS in FO treating municipal wastewater.

of the DS in the protocol. A bench-scale FO unit was used, while CTA membranes (Hydration Technology Innovations, HTI, USA) were applied to all experiments; the main purpose of this study was to evaluate the applicability of ionic organic solutions to OMBR systems.

Magnesium acetate ($C_4H_6MgO_4$) and sodium propionate ($C_3H_5NaO_2$) recorded the best performance as DS in terms of J_s, potential recovery, and biodegradability. Siddique et al. [76] showed similar results with the application of synthetic wastewater, highlighting $C_4H_6MgO_4$ as suitable DS for OMBR applications, while sodium acetate ($C_2H_3NaO_2$) led to the development of dense membrane biofilm. Despite the many benefits of ionic organic solutions, it should be noted that their potential application is limited, as the re-concentration cost is high compared to inorganic solutions.

A recent study aimed to integrate all the parameters of **Table 4** with the compatibility of FO as a pre-treatment step preceding the anaerobic process [83].

Draw	Feed	Configuration/ Membrane Type	Findings	Ref.
0.6 M $C_2H_3NaO_2$, 0.3 M EDTA-2Na & 0.5 M NaCl	Synthetic municipal wastewater	OMBR-RO/ CTA (HTI,USA)	• Minimized membrane fouling, lower J_s for the tested solutions over NaCl, but also lower J_w and reduced salt accumulation	[74]
0.1, 0.3, 0.5, 0.7 & 1 M EDTA-2Na	Activated sludge	FO-NF/ CTA membranes (HTI, USA)	• 0.7 M EDTA-2Na was the preferred concentration	[75]
			• At pH equal to 8, the FO membranes achieved the best performance	[75]
			• Reduced J_s compared to inorganic salts	
0.25 M $CaCl_2$, 0.25 M $MgCl_2$, 0.25 M $C_4H_6MgO_4$ & 0.25 M $C_2H_3NaO_2$	Synthetic municipal wastewater	OMBR-MD/CTA membrane (HTI, USA)	• $C_4H_6MgO_4$ optimized sludge flocculation, $C_2H_3NaO_2$ and $MgCl_2$ achieve steady flows in repeated tests	[76]
			• Inhibition of biological activity due to Cl^- presence	
			• $C_2H_3NaO_2$ generates significant fouling	
NaCl, $MgSO_4$, $C_2H_3NaO_2$, $C_4H_6MgO_4$, $CHNaO_2$, EDTA-2Na, $C_6H_{12}O_6$, $C_2H_5NO_2$, $C_3H_8O_3$, CH_4N_2O	Municipal wastewater	Direct FO/ CTA membrane (HTI)	• Organic solutions, such as $C_2H_3NaO_2$, are appropriate for this configuration. Reverse leakage of NaCl does not interrupt the anaerobic process	[13]
$CHKO_2$, $K_4P_2O_7$, $(C_3H_3NaO_2)_n$, $C_{2n}H_{4n}$ + 2O_{n+1}, $MgSO_4$ & NaCl	Synthetic secondary effluent	Post FO/ spiral wound (SW) CTA and TFC & flat sheet TFC and CTA	• $MgSO_4$, $(C_3H_3NaO_2)_n$ and $K_4P_2O_7$ identified as suitable solutions in terms of cost-effectiveness, toxicity, recovery and pH range	[77]
$(C_3H_3NaO_2)_n$, $MgSO_4$ & $MgCl_2$	MBR permeate	FO-NF/ TFC (Porifera, CA, USA)	• $(C_3H_3NaO_2)_n$ was unsuitable for irrigation, $MgSO_4$ caused prolonged membrane fouling, while $MgCl_2$ had the best performance	[78]
Commercial fertiliser diamond blue	Raw wastewater, MBR supernatant & permeate	Direct FO/ TFC membrane (Toray Industry Inc.)	• Liquid fertilizer has a good performance comparable to the application with common inorganic salts for green wall irrigation	[79]
KNO_3, KH_2PO_4 &KNO_3 (fertilizers)	Synthetic secondary effluent	FDFO/ CTA membrane (HTI, USA)	• Occurrence of severe biofouling using KNO_3 as DS compared to KCl and KH_2PO_4; membrane flux decline by 63%, 45% and 30%, respectively	[80]

Draw	Feed	Configuration/ Membrane Type	Findings	Ref.
NH_4NO_3, $(NH_4)_2SO_4$, NH_4Cl, $Ca(NO_3)_2$, KCl, $NH_4H_2PO_4$, $(NH_4)_2HPO_4$, KNO_3 & KH_2PO_4	Synthetic municipal wastewater	FDFO/ TFC membrane, (Toray Industry Inc.)	• KCl and NH_4Cl showed the highest water recovery and MAP, KH_2PO_4 and SOA showed the lowest J_s • High dilution is required about 100 tunes	[55]
$(NH_4)_2SO_4$, KH_2PO_4 & $(NH_4)H_2PO_4$	Synthetic municipal wastewater	FO-MBR (Direct FO)/CTA-ES membrane (HTI)	• MAP had the best performance with the lowest J_s • SOA altered the sludge characteristics	[51]
Commercial liquid fertilizer	Raw wastewater	Direct FO, CTA membrane (HTI, USA)	• Effective application of liquid fertilizer as DS for green wall irrigation • Reverse nutrient leakage worsened as the temperature rose	[81]
KH_2PO_4, KCl & KNO_3	Synthetic municipal wastewater	FDFO-AnMBR	• KH_2PO_4 < KCl < KNO_3 in terms of reverse leakages; alterations observed in anaerobic biomass, especially using KNO_3	[82]

Table 8.
Fertilizers, organic, inorganic, and ionic organic compounds as DS in FO treating municipal wastewater.

Among the 5 different zwitterions solutions tested, glycine ($C_2H_5NO_2$), L-proline ($C_5H_9NO_2$), and glycine betaine ($C_5H_{11}NO_2$) exhibited comparable J_w to NaCl (4.3–4.9 L m^{-2} h^{-1}), with lower J_s. From a physicochemical perspective, the process efficiency depends significantly on the pH value, affecting both the charge and the molecular size. Despite the rapid biodegradation (Adenosine triphosphate (ATP) levels range from 7 to 14 μg L^{-1} after degradation tests) of all zwitterions compounds, the replacement cost, which is 3–4 times more than the cost of commercially available solutions, is a potential barrier to their implementation in municipal wastewater streams. It is worth noting that the above experiments were performed with deionized water as feed, which favors the overall performance over the application of a more complex ionic matrix, such as sewage [83].

Commercial fertilizers are another largely inorganic solution medium that has been tested in various effluents resulting from a WWTP, such as typical secondary and MBR permeate and raw municipal wastewater. As illustrated in **Table 8**. Li et al. [82] compared the effect of 3 different commercial fertilizers on the downstream anaerobic process when applied as draw agents directly in raw wastewater. The following order of compatibility with the anaerobic treatment revealed Potassium Nitrate (KNO_3) > Potassium Chloride (KCl) > Potassium dihydrogen Phosphate (KH_2PO_4), with their reverse solute flux showing a similar sequence when the concentration of all DS was equal to 1 M. Water flux can be dramatically reduced by applying KNO_3 as DS, as extensive biofouling has been observed, while increasing nitrate concentrations can inhibit the subsequent anaerobic process, rendering them as unsuitable [80]. The implementation of different fertilizers in a hybrid FO-RO system to concentrate MBR permeate proved that the amplification of enhanced NaCl with Diammonium Phosphate (DAP) ($(NH_4)_2HPO_4$) can reduce reverse solute leakage by 35%, achieving NH_4-N rejection rates more than 95% at different flow rates (1.2 and 2 L m^{-2} h^{-1}) [66]. In addition, a long-term study examining the pilot application of a hybrid FO-NF system that treated MBR permeate found that Sodium Polyacrylate ($(C_3H_3NaO_2)_n$) was inappropriate for irrigation practices. On the contrary, the combination of $MgCl_2$ with NF membranes significantly improved the process efficiency and operating costs, as the application of chemical cleaning was not required. However, a notably high loss of the osmotic agent was observed [78]. A particularly interesting investigation was carried out by Adnan et al. [51] in which the possibility of applying 9 different fertilizers to the direct FO for the wastewater valorization and its parallel application in agricultural practices was examined. Water recovery was high by applying KCl (J_w = 21.1 L m^{-2} h^{-1}; J_s = 11.2 g m^{-2} h^{-1}; Osmotic Pressure (OP) = 44.6 bar) and Ammonium Chloride (NH_4Cl) (J_w = 21.1 L m^{-2} h^{-1}; J_s = 7.5 g m^{-2} h^{-1}; OP = 43.5 bar), while other fertilizers recorded particularly low reverse flux, such as Ammonium Sulfate (SOA) ($(NH_4)_2SO_4$) (J_w = 15.5 L m^{-2} h^{-1}; J_s = 1.7 g m^{-2} h^{-1}; OP = 46.7 bar), KH_2PO_4 (J_w = 13.2 L m^{-2} h^{-1}; J_s = 2.3 g m^{-2} h^{-1}; OP = 36.5 bar), and $NH_4H_2PO_4$ (Monoammonium Phosphate, MAP) (J_w = 13.8 L m^{-2} h^{-1}; J_s = 1 g m^{-2} h^{-1}; OP = 44.4 bar). However, this process becomes inapplicable, as a large amount of water is required to dilute the concentrated fertilizer (at least 1/100), to reach the irrigation limits [51].

The analysis of the existing literature makes it clear that the FO process is still under investigation and the determination of the ideal DS plays a vital role in upgrading the process of this technology. Despite the properties of the DS, the selection of the suitable configuration, the techno-economic factors, and the re-condensation method should be combined during the selection process; the optimization of the FO membrane's properties is a major challenge that can solve many issues. The development and fabrication of higher rejection membranes can be the answer to the implementation of both monovalent and divalent ions, which have been widely used as DS and their performance is already known to the research community.

4. Integration of FO followed by anaerobic treatment in a WWTP

4.1 COD valorization in municipal WWTPs

For more than a century, the CAS process has been applied as the main urban wastewater treatment system worldwide, making a significant contribution to environmental protection and public health. However, the low energy efficiency of the CAS process ranks WWTPs among the largest energy consumers in a country; on an annual basis, in developed counties, about 1–3% of electricity consumption is spent on their operation [84]. In addition, WWTPs are characterized by a high energy and carbon footprint, as during biological processes, large amounts of greenhouse gases are produced, mainly carbon dioxide generated due to the oxidation of organic matter and indirectly by electricity consumption [85]. Therefore, about 0.3–0.5 kWh m^{-3} of energy is required for sewage treatment by applying the CAS process, while the contained chemical energy and nutrients are not utilized [86].

According to Wan et al. [87] the traditional CAS process needs an average of 0.45 kWh to treat one m^3 of sewage, which equals to 1620 kJ m^{-3}. Assuming a concentration of 600 mg L^{-1} COD, energy consumption becomes 2.7 kJ g^{-1} COD.

As shown in **Figure 2(A)**, the energy recovery in convectional CAS systems occurs through the anaerobic digestion of the primary and secondary sludge, which corresponds to 32–39% of the organic material in COD terms. The latter percentage is equal to 2.9–3.5 kJ g^{-1} COD, since 1 g of methane-COD is equal to 13.9 kJ (65% methane percent in produced biogas). Considering that only 35% of the produced methane can be utilized for the production of electricity [86], about 1–1.2 kJ g^{-1} COD can be recovered from municipal wastewater, by applying anaerobic digestion to the sludge treatment line. Comparing the aforementioned energy requirement, 2.7 kJ g^{-1} COD, it is estimated that about 40% of it can be recovered using anaerobic digestion (1–1.2 kJ g^{-1} COD). The anaerobic digestion process also generates approximately 50–55% heat, part of which is used to heat the digesters. The excess heat can only be valorized locally [88].

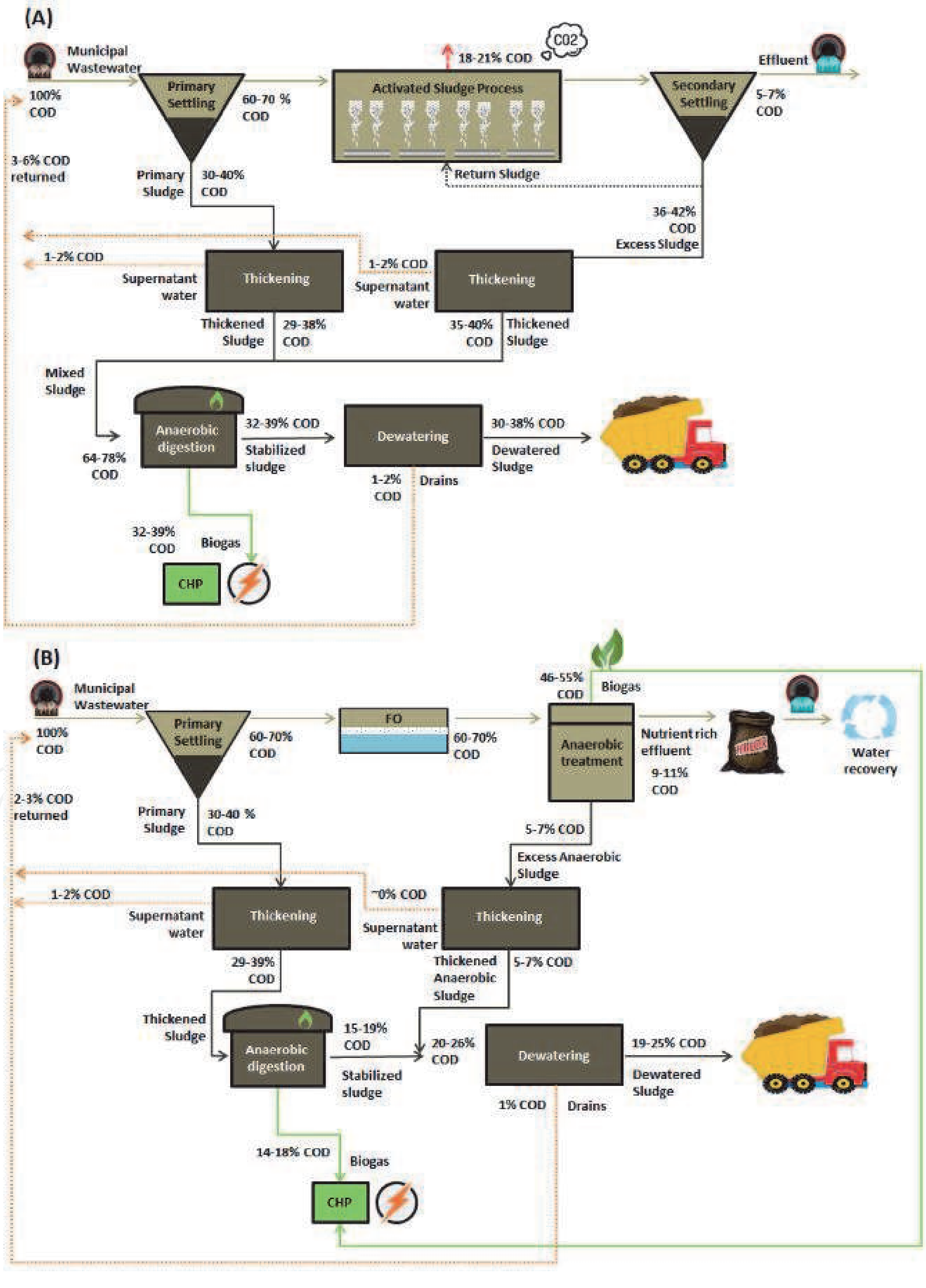

Figure 2.
(A) COD mass flow in a convectional WWTP, (B) COD mass flow, when FO followed by anaerobic treatment is integrated into a WWTP.

Obviously, COD capture, and subsequently valorization of the chemical energy contained in municipal wastewater can lead WWTPs to sustainable development, transforming WWTPs from energy consumers to producers, while significantly reducing the environmental footprint and operating costs.

The integration of FO in municipal wastewater treatment and the benefits of its application have been investigated in various studies [17, 49]. This chapter presents the combination of FO and anaerobic treatment in a typical WWTP for the utilization of the chemical energy, which is inherently present in sewage. As shown in **Figure 2(B)**, by placing the FO in the main treatment line of a WWTP and taking into account the efficiency of a typical anaerobic system, such as An-MBR, which is equal to 80% in ambient conditions [89], 46–55% of COD is converted to biogas (65% of the aforementioned percent corresponds to methane). Following the same procedure as before, the energy recovery in the main treatment line through the implementation of anaerobic treatment is between 4.2–5 kJ g^{-1} COD. Another 1.3–1.6 kJ g^{-1} COD of energy is recovered from the anaerobic digestion of the sewage sludge (13.9 kJ g^{-1} methane-COD). Since only 35% of the produced methane can be converted into electricity [86], the power production from the wastewater treatment line ranges between 1.3–1.7 kJ g^{-1} COD, while from the sludge treatment line it is equal to 0.4–0.6 kJ g^{-1} COD. On aggregate, 1.9–2.3 kJ g^{-1} COD of electricity can be utilized from this innovative treatment scheme, which can counterbalance 80% of the existing energy consumption of a typical municipal WWTP. The treated effluent of the anaerobic system is rich in nutrients, which can be valorized by applying recovery technologies for the production of slow-release fertilizers, while the reclaimed water content can also be reused.

4.2 Salinity, the greatest impact of FO on anaerobic treatment

Despite the benefits of the wastewater management system presented in the above section, there are two factors that can be particularly limiting to the subsequent operation of the anaerobic process. The solute flux that characterized the FO system results in the accumulation of salts in the feed stream, potentially resulting in partial or complete inhibition of the downstream anaerobic and aerobic biological treatment processes [14, 17, 32]. Salinity has been identified as an inhibitory agent of the anaerobic process, as the increased osmotic pressure across the cell membrane can cause plasmolysis, leading to cell death and total inhibition of the anaerobic process. More specifically, Lefebvre et al. [90] stressed that the activity of methanogenic bacteria is inhibited at concentrations of NaCl equal to 5 g L^{-1}, while acidogenic microorganisms are affected at much higher concentrations, i.e. 20 g L^{-1}. Ansari et al. [91] studied the effects of NaCl on anaerobic treatment of concentrated wastewater effluents in batch mode experiments and observed that by increasing water recovery rates of FO (from 50 to 90%), the anaerobic process achieves higher methane production (approximately 5 times higher), while the presence of salinity has a negligible negative effect.

Based on the existing literature, the limiting parameter of salinity has been investigated and observed only in aerobic/anaerobic systems, where the FO unit is plugged into MBR systems for a relatively short time, while in pre-concentration systems few studies have examined the effect of salinity on the downstream anaerobic process and suggest mitigation measures. Chen et al. [37] and Wang et al. [39] did not observe significant effects of salinity on anaerobic reactors by recording an average methane yield of 0.2 and 0.3 L CH_4 g^{-1} COD, respectively, in studies that cannot be characterized as long-term. As mentioned above, the application of minimization strategies such as the corresponding regulation of the hydraulic residence time (HRT) seems to regulate the salinity conditions to which the biomass is exposed. Accordingly, the addition of MF membranes is a particular interesting approach for the minimization of salinity and the parallel application of nutrient recovery methods. Another interesting perspective is the acclimatization of the anaerobic biomass to high salinity conditions. This mitigation technique is not recent as the presence of specific microorganisms, such as halotolerant bacteria has shown particularly high efficiency in the anaerobic treatment of saline industrial wastewater [92]. In a recent study, where no acclimatized biomass was used, Gao et al. [93] separately investigated the effect of high salinity and ammonia nitrogen concentration and the combination of the two inhibitors in the anaerobic treatment of pre-concentrated municipal wastewater. The results showed that the presence of NH_4-N and NaCl concentrations separately, up to 200 mg L^{-1} and between 5 and 8 g L^{-1}, respectively, did not significantly affect the activity of anaerobic microorganisms. The combination of the two parameters in non-acclimatized and acclimatized biomass showed that the latter had significantly better performance and can respond without the risk of inhibition. Further research into anaerobic biomass acclimatization should be conducted in the future, as higher condensation rates could be applied from the upstream FO unit.

All the acquired knowledge of the above studies would be particularly interesting to be used in the long-term investigation of a FO system combined with a downstream anaerobic process, in which all the limiting parameters and the proposed mitigation measures can be examined in-depth, for the rational assessment of its performance.

5. Conclusion

There is no doubt that FO is a promising technology that has been investigated for a range of applications at various stages of a municipal WWTP. Among them, its combination with the anaerobic process has significant advantages, as much of the chemical energy inherently contained in sewage can be recovered as biogas, while resource recovery technologies can be applied downstream, utilizing the nutrient-rich effluent. However, the transition of the FO from laboratory scale to full-scale applications requires further research to address important issues, such as the salinity accumulation in the downstream technologies and the reduced rejection of NH_4-N by existing FO membranes. The application of NaCl indicates a possible suitability for the concentration of municipal wastewater. The background knowledge available on the basic criteria of FO has to be utilized for the development of membranes with higher selectivity. Future investigations should carry out extensive long-term monitoring and targeted combination/interaction of different parameters for the concentration of real wastewater, to assess from a technical, environmental and economic perspective the feasibility of applying FO technology to municipal wastewater management.

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Nomenclature

ATP	adenosine triphosphate
NH_4-N	ammonium nitrogen
SOA	ammonium sulfate
An-OMBR	anaerobic osmotic membrane bioreactor
CTA	cellulose triacetate
COD	chemical oxygen demand
CAS	conventional activated sludge
DAP	diammonium phosphate
DS	draw solution
EDTA 2Na	ethylenediamine tetraacetic acid disodium salt
EU	European Union
FO	forward osmosis
FO-NF	forward osmosis-nanofiltration
$C_2H_5NO_2$	glycine
$C_5H_{11}NO_2$	glycine betaine
HTI	hydration technology innovations
HRT	hydraulic residence time
$C_5H_9NO_2$	L-proline
$C_4H_6MgO_4$	magnesium acetate
$MgCl_2$	magnesium chloride
$MgSO_4$	magnesium sulfate
MBR	membrane bioreactor
MF	microfiltration
MF-OMBR	microfiltration-osmotic membrane bioreactor
MAP	monoammonium phosphate
NF	nanofiltration
PO_4-P	orthophosphate as phosphorus
OMBR	osmotic membrane bioreactor
KCl	potassium chloride
KH_2PO_4	potassium dihydrogen phosphate
KNO_3	potassium nitrate
RO	reverse osmosis
J_s	reverse salt flux
SRT	sludge retention time
$C_2H_3NaO_2$	sodium acetate
NaCl	sodium chloride
$(C_3H_3NaO_2)_n$	sodium polyacrylate
$C_3H_5NaO_2$	sodium propionate
TFC	thin film composite
TOC	total organic cabon
UF	ultrafiltration
WWTP(s)	wastewater treatment plant(s)
J_w	water flux

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