Forward Osmosis for Industrial Effluents Treatment

- Sustainability Considerations

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Nomenclature

BFW	Boiler feed water	PA	Polyamide
BOD	Biological oxygen demand	PAN	Polyacrylonitrile
CA	Cellulose Acetate	PAM	Polyacrylamide
CC	Choline chloride	PBR	Photo-bioreactor
COD	Chemical oxygen demand	PES	Polyethersulfone
CP	Concentration polarization	PRO	Pressure retarded osmosis
DO	Direct osmosis	RO	Reverse osmosis
DOC	Dissolved organic carbon	SAGD	Steam assisted gravity drainage
DS	Draw solution	SDBS	Sodium dodecylbenzenesulphonate
DHSP	Di-sodium hydrogen phosphate	SDS	Sodium dodecyl sulfate
EDTA	Ethylene diamine tetra acetic	SEM	Scanning Electron Microscope
ED	Electrodialysis	SGDF	Shale gas drilling flow-back fluid

FO	Forward Osmosis	SHMP	Sodium hexametaphosphate
FOMBRs	FO assisted membrane bioreactors	SLS	Sodium lingo sulphonate
IDA-GWI	International Desalination Association- Global Water Intelligence	SPPSU	Sulfonated poly phenylenesulfone
ILs	Ionic liquids	SWRO	Seawater Reverse Osmosis
ICP	Internal Concentration Polarization	TDS	Total dissolved solids
MBC	Membrane brine concentrator	TFC-FO	Thin Film Composite-Forward Osmosis
MBR	Membrane bioreactor	TFT-LCD	Thin-film transistor liquid crystal displays
MD	Membrane distillation	TN	Total nitrogen
MDC	Microbial desalination cell	TOC	Total organic carbon
MF	Microfiltration	TOrCs	Trace organic contaminants
MFC	Microbial fuel cell	SWFO	Seawater Forward Osmosis
MLVSS	Mixed liquor volatile suspended solids	VFA	Volatile fatty acids
MLSS	Mixed liquor suspended solids	UCST	Upper critical solution temperature
NF	Nanofiltration	UF	Ultrafiltration
OMBR	Osmosis Membrane Bioreactor		

Abstract

Access to the clean water resources has become a global challenge in recent times, especially in developing countries, where huge amounts of highly polluted industrial and municipal effluents are produced and discharged into the receiving environments. Applications of membrane technologies to deal with the effluents from various origins have recently received a great deal of attention due to their inherent advantages, compared to other physico-chemical methods developed so far to this end. Forward osmosis (FO) is among the efficient membrane-based processes adopted by wastewater treatment facilities, with various configurations currently being transferred from laboratory and pilot-scales to the large-scale applications. However, FO technologies are still plagued with drawbacks such as fouling, internal concentration polarization (ICP), reverse solute flux and draw solution recovery, which invariably increases the cost of operation and restricts the feasibility for large-scale and long-term use because economic considerations are the most important sustainability criteria when selecting a wastewater treatment technique among various alternative technologies. Several modifications have been introduced in recent years in order to overcome the existing limitations, such as the incorporation of engineered nanomaterials onto the membrane surface to mitigate the membrane fouling and to enhance their life-time, thereby minimizing the cleaning and (when necessary) replacement costs. Several FO based

pre-treatment technologies have also been introduced for complex effluents treatment in order to minimize the operational costs arising from cleaning and replacement activities. Therefore, assessing the performance of such technologies according to sustainability indicators is the key to ensure long-term benefits from the application of FO technologies for the treatment of highly polluted effluents. In this review, sustainability criteria for the treatment of industrial effluents using FO technologies are covered and categorized in technical, economic, environmental and social branches and the current state of the technology regarding the mentioned sustainability criteria are discussed. Scaling-up opportunities for the existing lab-scale modules have been evaluated and the probable consequences, viz. environmental footprint of the under-developed technologies are discussed. This review will therefore aid in selecting the most suitable configurations of FO technologies (standalone/hybrid) to deal with the highly polluted effluents for real world applications and to direct future studies in this emerging area. Perspectives and recommendations for the future studies are also included in this review.

Keywords: Forward Osmosis; Sustainability, Wastewater Treatment; Draw solution; Recycling; Recovery.

1. Introduction

The amount of wastewater produced from industrial sectors and human activities are the major causes of environmental and health concerns. In this regard, rapid industrialization has led to the generation of huge quantities of highly polluted effluents including bio-wastes, causing severe environmental threats. In many cases, discharged toxic untreated liquid waste of various origin and type along with the persistent emerging pollutants entering into the clean water bodies is a future cause of concern. Large quantities of fresh water is consumed in industrial processes such as tanning (leather industry), dying (textile), sugar industry, mining, automobile, electroplating etc. with nearly >90% of used water being discharged into the environment as effluents having unused raw materials and by-products in them [1, 2]. However, the absence of proper wastewater treatment technologies has led to severe problems pertaining to the discharge of highly polluted effluents and production of reusable water.

According to a recent United Nations World Water Development report (2017), approximately 80% of the global industrial and municipal wastewater is discharged without appropriate treatment. Although the ratio varies within different income group countries, but

it is estimated that more than 90% goes untreated/partially treated[1]. To overcome such a huge problem is a daunting task, nevertheless various physico-chemical and biological treatment technologies have been developed and are being used to treat highly polluted effluents before being discharged into the environment [3-5]. In this respect, membrane-based technologies are considered as the most sustainable among the other water treatment technologies [6-8]. Of all these methods, forward osmosis (FO) has received much attention in recent years due to its remarkable potential in water and wastewater reclamation and reuse. FO technologies are now considered as emerging alternatives to other pressure-driven membrane processes such as reverse osmosis (RO) and nanofiltration (NF) for complex effluents treatment purposes [9]. Currently, pieces of evidence are pointing towards the successful FO membrane manufacturing, process development and operational optimization to demonstrate the large scale potential of these technologies considering the sustainability criteria [10-14].

Attempts are actively being pursued to achieve a better understanding on the dynamics of separation process at the membrane interface with the emphasis on draw solution recovery and reducing energy consumption both for sustainable water and draw solute/solution recovery [15, 16]. Unlike unidirectional drift that happens in pressure driven membrane processes [17], mass transport is complex in FO membranes, with significantly different active layer parameters [18]. There are several parameter, which can influence the efficiency of the FO membranes such as the nature of the membrane, its morphology and pore characteristics, thickness, and wettability as well as the solubility factors[19]. In addition, operating parameters such as high flux, low fouling propensity and good rejection rates with low concentration polarization need to be considered for promoting the FO membranes.

In a recent study, Guo et al. reviewed the studies on designing suitable membranes which can address the issues such as severe internal concentration polarization (ICP), which is highly expected in an FO process, while maintaining its long term integrity under wastewater treatment conditions [20]. Researchers concluded that the concentration polarization effect depend on parameters related to membrane properties and the process design aspects. Therefore, designing an efficient treatment configuration is of utmost necessity to satisfy the sustainability criteria, which considers all the technical, environmental, economic and social aspects [21-23].

FO has been widely used in applications such as shale wastewater reclamation [9], hyper saline wastewater desalination [24], hybridization with other processes for multiple

resource recovery and energy production [25], value addition [26] and in specialized applications in biological and other fields [27]. However, similar to any other novel technologies, FO is now in competition with the existing membrane processes as well as other traditional technologies. At this stage, studies on various aspects of FO applications in a systematic framework are highly beneficial. Although there are several publications on the development and utilization of FO processes in waste treatment aspects, critical reviews considering the sustainability aspects are rare in the literature [28-30]. Hence, an effort has been made to discuss FO and hybrid FO processes for wastewater treatment, its sustainability aspects along with the challenges and future opportunities.

2. Forward Osmosis and Sustainability

Despite numerous methods developed to recover useful constituents from the wastewater sources [31, 32], stringent environmental regulations continue to put pressure on the industries to meet the set standards. There is thus an urgent need to develop the sustainable alternatives for the treatment of highly polluted effluents. Due to the pressure from regulatory authorities, various sustainability criteria are being considered for selecting the most appropriate technology to treat specific wastewater [33]. Among all, membrane-based technologies are in the forefront [34]. FO is an engineered osmotic process that uses a semi-permeable membrane to promote the separation of water from the dissolved solutes, and is now widely explored for the treatment of wastes. The driving force for the separation in FO is an osmotic pressure gradient between the solution of high concentration (often called “draw”) and a solution of lower concentration (called “feed”), as illustrated in Fig. 1. FO is distinguished from reverse osmosis (RO), which uses hydraulic pressure as the driving force to separate the pollutants, and which counteracts the osmotic pressure gradient. Otherwise, water flux will be from permeate to the feed side due to chemical potential gradient [35].

Figure 1. General schematics of a lab-scale working FO module with components and operational specifications.

Hence, the energy required to separate water from the contaminants using RO is considerably higher than that of FO. Osmotic pressure gradient in FO can be used to induce a net flow of water from the feed through the membrane into the draw without an external pressure source. Having differentiated between the operating principles of FO and RO, the net energy consumption of a FO process has to be taken into account where the recovery of draw solute/solution is an energy intensive step. If the process design allows for the diluted draw to be used as it is, viz fertilizers as draw solutions [36], the operational cost is much lower compared to pressure-driven RO[37]. The draw solution replenishment over the course of operations needs to be taken into account. On the other hand, if a recovery step is essential so that the draw can be recycled, the energy input is considerable and can be equal to or even more than for conventional RO systems[38]. It should be emphasized here that these calculations were made with seawater as feed and not complex industrial wastewaters.

Feeds rich in organics viz. dyes, pharmaceutical compounds, trace organic contaminants (TrOCs) etc. biological wastes, heavy metals, inorganic scalants with high salinity will be difficult if not impossible to treat by stand-alone RO and requires pretreatment and post-polishing. This invariably adds up to the cost and external chemical inputs for membrane cleaning at regular intervals, addition of anti-scalants etc., thereby reducing the sustainability of the overall process. Where RO fails, FO shines. Treatment of these complex wastewaters with reduces fouling propensity due to no external pressure requirement makes FO process suitable as well as a sustainable alternative.

FO processes have shown superior performance over other membrane technologies for the treatment of wastewater. Coday et al. [39] studied the physiochemical and transport mechanisms involved in FO by evaluating the performance of cellulose acetate (CA) and polyamide(PA) TFC-FO membrane in terms of flux (3-9 L/m²/h and 5-13 L/m²/h for CA and PA respectively), for the oil and gas wastewater. Both the membranes experienced irreversible decline in flux over the course of operation while the authors attributed to osmotic de-swelling and charge neutralization. Polyamide and CA polymers have been extensively studies for their anti-fouling properties. However, the presence of high concentration in feed solutions, such membranes have shown higher susceptibility for solute accumulation on their surfaces. An important aspect explored here is the development of early types of surface coated TFC membranes specifically for FO process. Herein, a hydrogel coated TFC membrane was reported to have led to a reduction of reverse solute flux (RSF) by 50% with low fouling propensity under test conditions.

Cui et al. [40] studied the removal of micro-pollutants such as phenol, aniline and nitrobenzene using different polyamide-TFC FO membranes based on Matrimid, polyethersulfone (PES) and sulfonated poly(phenylenesulfone) (SPPSU) polymeric support. In all the cases, FO resulted in 70% to 90% rejection of organic pollutants using 1M NaCl draw solution, while the rejections were considerably lower when the same membranes were tested under RO mode using the same feeds. What is more interesting is that these membranes, being tested in FO mode, surpassed commercial RO membranes operating under pressure. Introduction of 2M NaCl draw solution increased the flux two-folds (compared to 0.5 M NaCl) following the increase in the driving force. It also increased the rejection of pollutants by at most 17 %. Although the data are supported by experimental evidence, the rationale behind the enhanced rejection is not thoroughly discussed. This is attributed to higher rejection efficiency in FO mode to the transport mechanism involved, i.e. solution-diffusion mechanism and a probable hindrance created due to RSF.

In RO mode, the transport mechanism is governed by both solution-diffusion and pore-flow models, which results in lower rejection rates. Although no relevant literature was cited to support this claim as RO membranes are also dense, with solution-diffusion being the preferred mode of transport through polyamide layer. Here, the presence of modified gel layer have proven water absorbing layer with capacity to swell and de-swelling under the influence of concentration barrier across the membrane. This has been the highlight of the study showing improved flux performance for FO compared to RO when similar polymer dope and fabrication conditions are used to form the membranes.

Various parameters affect the performance of a treatment method or technology are now being evaluated based on their overall efficiency and targeted waste mitigation. Considering many such criteria membrane technologies outperform all other existing technologies. For instance, treatment efficiency may attract the industries that need to deal with high amount of industrial effluents and minimizing concentrate for better disposal. Also, recovery of value added materials would also increase the sustainability economy of the process and prevent the release of such compounds to the receiving environment. In some cases, release of less harmful concentrates would help to reduce post-treatment costs, especially in the current competitive environment related industries[41]. In addition, FO technology must be socially acceptable for further developments. This review discusses about the performance of FO membranes, keeping in view of the sustainability criteria in order to emphasize the existing gaps.

3. FO as a single treatment step

FO has proven its ability to be an emerging low-energy technology for complex wastewater dewatering. In FO process, water is separated from the effluents which can be considered as the initial step involving the treatment of the effluents using such technologies. However, a complementary step would be required in order to extract the water and the recovery of the draw solutes. Some processes are currently being used for the draw solute recovery. For instance, for low molecular weight based draw solute recovery is mainly performed by using techniques such as NF and/or RO as energy intensive processes.

In this section, the application of FO as a single treatment step for the treatment of polluted water and wastewater streams are discussed. Emphasis is given on different feeds and recovery efficiency, draw solutes, recovery of by-products and fouling propensity, considering sustainability as the main criteria.

3.1 Feeds

FO performs under variety of feed streams to recover water and to diminish the adverse effects of polluted wastes and wastewater discharged into the environment. Various industries have already adopted or are in the process of adopting this technology to deal with effluents in many industrial sectors. [42-45]For instance, FO has gained much interest in oil and gas industries for reclamation and reuse of effluents discharged and the process offers a high level of selectivity for different contaminants, which make it suitable for water treatment and reuse. Coday et al. [24] used CA and PA-based TFC membranes in desalination steps of

shale formation process. During oil and gas (O&G) exploration, high concentrations of organic and inorganic contamination are released as effluents creating serious threats, thereby affecting the industrial process sustainability.

Figure 2. Simultaneous desalination of O&G pit water and waste stream concentration using (a) hybrid engineered osmosis system and (b) simple osmotic dilution. The hybrid engineered osmosis process employs seawater RO to re-concentrate osmotic draw solution and nanofiltration to remove divalent ions from the concentrated brine after FO, reprinted with permission from Coday et al., [46].

The authors examined hydrodynamic parameters on membranes performance such as flow dynamics, modulation of active layers, membrane packing, process parameters and module design. Further, economic and ecological impacts of O&G pit water were examined in terms of comparative life cycle impact and cost assessments. The FO water recovery as a stand-alone osmotic dilution process in the environmental impact category is displayed in Fig. 2. The state-of-the-art FO contributed immensely to diminish the negative environmental impacts with ~75% water recovery by reducing the pit wastewater management cost by an average of 60% compared to conventional treatment methods [46].

Gu et al. [47] used brown coal slurry as the draw agent for water recovery from the dye wastewater using FO to evaluate dewatering efficiency of brown coal slurry and its reusability in FO for repeated use. The authors identified osmotic dewatering to be the main mechanism involved with flux variation observed in terms of changes in moisture holding

capacity, particle size and porosity of the brown coal slurry. FO has the capability to solve the some target specific issues and unfamiliar applications where all other conventional technique fail. For instance, reclamation of water from drilling processes is a potential area where FO has been tested at the commercial levels. The commercial HTI membrane[48] is a sample of commercialized FO membranes, which was used to achieve both high organic and inorganic contaminant rejection[49-51].

Hickenbottom et al. [52] used FO for the treatment of drilling mud and fracturing wastewater from O&G operations, wherein simple osmotic backwashing could help to retain high osmotic flux (2.5-5 LMH) with >80% of water recovery. In this study, process sustainability factor in terms of pulsed washing and shutting of equipment's for the purpose of maintenance was well addressed. This study highlighted the major advantage of FO over other membrane process where the intermediate membrane washing surface is a time consuming task during operations. In another study, membrane brine concentrator (MBC) was employed[53] in which NH_3/CO_2 was used as the osmotic agent for desalinating waste produced during natural gas extraction operations. In this study, FO was effective to treat water with total dissolved solids (TDS) of $73,000 \pm 4200$ mg/L and an average hardness of $17,000 \pm 3000$ mg/L. Polyamide-based FO membrane produced 2.6 LMH to achieve nearly 65 % recovery of fresh water. Here, the only major energy consumption was during draw solute or product water recovery (275 ± 12 kWh/m³).

There is sufficient information available on cost and energy economic calculations for RO, however limited information is available on overall FO process in comparison to RO for similar applications. However, the overall process efficiency was higher with approximately 57% lesser energy consumption than conventional evaporation processes. To predict accurate cost of CAPEX and OPEX in comparison to other pressure driven membrane processes, there need to be specific studies conducted. As there is limited information on cost estimation in terms of energy consumption comparing FO with RO process with similar operational and performance parameters.

FO as a standalone single-step treatment process has been used to treat polluted waters and wastewaters. For instance, Roy et al., [54] used FO for recovering reusable water from soil saline contaminated water. The 1 M CaCl_2 as a draw solution could extract ~50% reusable water having an initial TDS of 16300 mg/L. The authors suggested that diluted draw solution (CaCl_2) in known concentration range of 0.41 and 0.44 M can be used as a cation exchange solution, reducing the treatment costs with better economic profitability. High salinity wastewater containing heavy metal loaded effluents have been treated using FO[55].

For instance, simulated water containing Ni^{2+} (100 mg/L) was treated with different types of FO membranes, wherein concentration polarization (CP) of Ni^{2+} on different membrane surfaces was monitored by energy dispersive spectral diffraction in SEM. This study showed the way to analyze first of its kind micro-visual technique to characteristics of membranes pre- and post-usage. Increasing the salinity of the feed, CP showed a decreasing trend, while the presence of sodium dodecyl benzene sulphonate (SDBS) surfactant increased the osmotic flux through CTA membrane.

TFC membranes find applications in FO for the treatment of boiler feed water (BFW) of steam assisted gravity drainage (SAGD) [56] and the results showed superior permeation properties of TFC polyamide membranes than commercially available TFC-FO membranes. Hu et al [57] tested the biogas slurry as a draw solution for the extraction of clean water from heavy metal ions containing micro-polluted feed solution. The results indicated rejections of 98.5% and 97% for Cd^{2+} and Pb^{2+} ions, respectively. Also, the diluted biogas slurry was effectively utilized as a hydroponic solution for cultivation of rice and leek.

In addition to water and wastewater treatment, sludge de-watering was effectively treated using FO as a single treatment step. A complete dewatering of the sludge was achieved [58] after 19 days of treatment resulting achieving simultaneous thickening, digestion, and dewatering of the feed. By this technique researchers accomplished mixed liquor suspended solids (MLSS) concentration of ~63% and liquor volatile suspended solids (MLVSS) concentration up to 80%.

In this study, the MLVSS/MLSS ratio continuously decreased from 81% to 67%, while MLSS concentration reached 39 g/L from an initial value of 7 g/L. Additionally, an initial sludge depth and draw solution (DS) concentration over 35% sludge dryness was achieved, indicating the potential of FO for commercializing the sludge dewatering process. These findings are the best examples of excellent dewatering ability of FO, where other technique fail to achieve desired end goal and otherwise would be difficult by any other processes.

3.2 Draw Solutes

Similar to the external hydraulic pressure in case of pressure-driven membrane processes, draw solution in an FO process induces the osmotic gradient. As the backbone of overall process, draw solutes and their recovery processes have amassed significant attention in the research community leading to a number of review articles being published on related topics [15, 59, 60]. Wherein the focus of the previous reviews was on novel draw solution

systems and their recovery methods, this review analyzes the advancements in draw solutions from the overall process perspective. As the draw solution regeneration and water recovery is an energy consuming step, those processes which does not require such steps can be considered much more efficient and economical. Application of by-products/wastes of industrial or agricultural sectors and recovery of by-products from feed streams also offsets the energy consumed. Finally, as the FO process has proven its niche in the field of treatment of complex wastewater feeds where RO fails, this review scrutinizes the available literature keeping in mind the sustainability considerations.

Various types of fertilizers have been widely explored as draw solutes to extract water from the waste streams. Some features such as high osmotic potential, recoverability, non-toxicity and reusability are among the most important characteristics of a good draw solute/solution. Some of the fertilizers well fit in the parameters set for being efficient draw solute as they may not necessarily require post recovery step. In some cases, diluted draw solutions can be directly used for farming purpose as nutrients. Phuntsho et al.[36] tested nine commonly used fertilizers to measure pure water flux and reverse draw solute flux. It was observed that KCl, NaNO₃ and KNO₃ solutions resulted in high water fluxes, while NH₄H₂PO₄, (NH₄)₂HPO₄, Ca(NO₃)₂ and (NH₄)₂SO₄ have shown the lowest reverse solute fluxes.

Other specific types of draw solutions were also employed in FO operations. For instance, Mazzuca Sobczuk et al., [61] utilized the submerged dialysis membrane bags during biodiesel production containing pure and crude glycerol as the draw solution. A microalgae concentration of 2500 mg/L was achieved with marine microalgae dewatering rate of 2.75 LMH via osmotic bag having glycerol as draw solution. Some specific types of draw solutes such as polyacrylamide (PAM) have been also used as draw solute for the recovery of water from the effluents. In a recent study [62], the efficiency of easily recoverable PAM was compared with conventional KCl draw solute to obtain high flux. Higher solute recovery by PAM was achieved compared to KCl. PAM was able to generate stable osmotic flux of >99.99% for dye rejection. PAM is a macromolecule and can be easily recovered using large pore size processes like MF with less energy input making it economical.

Zhao et al. [63] developed a new polymer-based draw solute to sustain long-term (50 h) FO process. In this study, fouling rate was considerably lower compared to pressure retarded osmosis (PRO) mode. The hydrolyzed PAM draw solute caused rigid scaling layer onto TFC-FO membrane in a PRO mode. According to Xue et al. [64], seawater was used as draw solution by employing CTA-FO semi-permeable membrane to concentrate nutrients

from the municipal wastewaters in a membrane bioreactor (MBR)-assisted FO process, which was run for up to 2 months continuously in a bench-top reactor. In addition to above-mentioned draw solutes, ionic liquids (ILs) were also used as draw solutes. For instance, Zhong et al. [65] employed thermo-responsive ILs of upper critical solution temperature (UCST) as draw solute. The protonated IL osmotically drew the water at an average flux rate of $10 \text{ Lm}^{-2}\text{h}^{-1}$. One of the advantages of using ILs is the potential of phase separation after treatment as displayed in Fig 3.

Figure 3. FzO process based on UCST-IL as draw solution, adopted from Zhong et al.[65].

In a target specific and greatly sustainable effort, Mondal et al. [66] employed sea bittren (a mother liquor left after salt production in solar salt ponds) as an efficient draw solution to concentrate sugarcane juice (see Fig. 4). This unique effort to achieve 4-fold concentration of sucrose in sugarcane juice with a LMH dewatering flux of 19 was achieved using vast left over natural resource after recovering common salt from solar ponds. Blandin et al. [67] also used seawater or brine to concentrate volatile fatty acids (VFA) higher than acetic acid to achieve $> 80\%$ rejection at pH7.5.

Organic draw solutes were also considered at 56°C for wastewater treatment containing aniline, nitrobenzene and phenol contaminants by Zhong et al [65] using protonated betaine-bis(trifluoromethylsulfonyl)imide ([Hbet][Tf₂N]) (3.2 M). Further, low-cost organic fertilizers were also employed as draw solutions to reduce the total cost of treatment. In a similar approach, Ansari et al. [68] also achieved $>90\%$ reusable water

recovery by concentrating organic matter from polluted effluents using sodium acetate and EDTA-Na as the draw solutions. Fig.5 illustrates organic matter concentration using this approach.

Figure 4. A schematic illustration of sucrose concentration in sugarcane juice through energy efficient FO using sea bittern as the draw solution, reprinted with permission from Mondal et al. [66].

Different salt draw solutions were employed by Bhinder et al. [69] in FO for treating oil-contaminated wastewater from bitumen extraction process. The efficiency of the method was studied as a function of concentration, temperature and pH. The FO could concentrate domestic wastewater containing high chemical oxygen demand (COD) and biological oxygen demand (BOD) from the produced sludge to an extent of ~10-folds using 2.2 M $MgCl_2$ as draw solution.

In efforts to optimize FO performance while dealing with domestic effluents, Xiang et al. [37] observed that higher concentration of draw would lead to higher water flux using submerged FO system, for which less energy was consumed. It is apparent that the higher concentration of draw solution, the higher osmotic pressure can exert across the membrane and higher concentration gradient which results in higher flux rates. However, due to higher draw solution concentration, it is expected to have high reverse solute flux. In this case higher reverse solute flux was also heightened due to membrane swelling.

The effectiveness of $MgCl_2$ as the draw solute was studied by Giagnorio et al. [70] to produce high-quality water from real brackish groundwater and secondary effluent wastewater with a salinity of 4 g/L. Feed solutions were first treated with FO to achieve water recovery of >60%, but the original osmotic pressure of diluted draw solution was regenerated using NF. When $MgCl_2$ and Na_2SO_4 were used as draw solutions, it was found that $MgCl_2$

showed larger specific reverse salt flux for groundwater to induce pronounced fouling-related flux decline. The average flux obtained was 5 to 11 LMH, while the combined system offered low flux providing good water.

Figure 5. A schematic of FO for the pre-concentration of organic matter in raw wastewater, reprinted with permission from Ansari et. al., (2016) [68].

Nguyen et al. [71] tested the potential of aqueous KOH solution as a draw solution for the recovery of iodide from thin-film transistor liquid crystal display (TFT-LCD) plant's waste stream. This process suffered from the formation of KI cake layer while recovering iodine from the waste stream, but flux was as high as 11.7 LMH at pH 11. Authors suggested that liquid crystal industrial wastes can also be efficiently utilized for iodine recovery, satisfying some aspects of sustainable development of membrane technologies including reuse of wastes, which is of high priority in terms of environmental considerations as well as to reduce the total cost of the treatment process.

It can be concluded that there is a growing need to design novel draw solutes which are compatible with FO membranes and exhibit low reverse solute flux. Thermo-responsive and pH-responsive draw solutes shows some potential for scale-up albeit with better recovery systems in place. In this regard, some notable examples have been so far studied in the literature. For instance, 1,4-bis (3-propane-sulphonate sodium)-piperazinediethanesulfonic acid disodium-sulfate, a pH responsive supramolecular draw solution, has been employed by Wu et al. to remove arsenic from water using FO process. The authors reported a high flux of 58.4 LMH for a diluted draw solution (0.24 M) using TFC FO membrane [72]. Another cage-like supra-molecular material, N-(hexamethylenetetramine) propane-1-sulfonate was also used by Liao et al. for wastewater remediation and protein enrichment. The most notable feature of the latter study is the

negligible loss in draw solution by reverse solute leakage. The authors attributed this to the formation of a supra-molecular network with water molecules via hydrogen bonding interactions [73]. Another group [74] also employed a multi-charge hydro acid chromium complex as a novel draw solution to treat electroplating wastewater. Upon ionization in aqueous medium, the draw solution produces multi-charge anionic $[\text{Cr}(\text{C}_2\text{O}_4)_3]^{3-}$ and three cationic $[\text{N}(\text{CH}_3)_4]$, thereby exerting high osmotic pressure with low reverse solute flux. Membrane Distillation (MD) was employed for regenerating the draw solution for subsequent FO cycles. Apart from these, thermo responsive polymer hydrogels have also proven to be efficient in dewatering various feed streams. A well-known thermo responsive polymer matrix of poly(M-isopropylacrylamide) (PNIPAM) were employed in such studies [75, 76]. Some other types of draw solutes have been studied and reported in the literature. Switchable polarity solvent, 1-cyclohexylpiperidine has been for instance tested as a viable candidate for FO process as reported by Omre and Wilson (2015)[77]. On exposure to carbon dioxide, they showed the generation of ammonium bicarbonate and amine functionalities which creates the osmotic gradient. Degassing at 70°C under ambient pressure to remove the CO_2 was also proved to be effective for draw solute regeneration. The authors concluded with a techno-economic process model for the overall process with favourable results.

3.3 Recovery of by-products

Recovery of materials is considered to be one of the most important aspects of any methods developed for the treatment of polluted water and wastewater streams, which would help to compensate the cost spent (or at least a part of it) and partially addresses the sustainability aspect in terms of carbon footprint. In this way, FO can be used for the recovery of useful compounds such as phosphate and calcium. In this pursuit, Ansari et al. [78] recovered 92% of phosphate from the digested sludge using seawater as the draw solute, and by this, overall treatment costs were reduced.

Gwak et al. [43] investigated the metal recovery from the printed circuit board wastewater using waste solution as the draw solution (containing Cu, Ni, etc.) and electroless nickel plating solution containing Pd, Au, and Ag as the feed solutions. In this study no permeation of heavy metals from feed solution noticed. Therefore, diluted draw solution was safe for disposal or reuse in different applications. In this study, re-concentrations of draw solution were not required, but modeling studies revealed that Pd can be theoretically concentrated to 17.2 times and this process would satisfy the sustainability considerations due

to zero cost for the supply and re-concentration of draw solution and enhanced energy efficiency in the precious metal recycling process.

Xie et al.[79] investigated in-situ formation struvite and recovery of nutrients from the digested swine wastewater by reverse-fluxed Mg^{2+} (which participates in struvite formation) in the FO system by achieving >50% water recovery and >93% ammonium nitrogen removal from the digested swine wastewater with 99% of phosphate recovery. This study also demonstrated the possibility of nutrient recovery from the livestock wastewater.

Sustainability considerations require the use of green materials for the treatment of polluted wastewaters and hence, there is a need for further studies on the application of natural origin and sustainable compounds for the FO processes. In this regard, Mondal et al. [27] utilized the naturally occurring green solvents as draw solution to enrich bio-macromolecules of industrial importance. Here, naturally extracted deep eutectic combination of choline chloride (CC), ethylene glycol and glycerol Gly were used in the ratio of 1:2 to concentration DNA and bovine protein in the FO process. The temperature and pressure sensitive biopolymers were efficiently concentrated to 3-7 folds via such a green sustainable approach. Owing to green and friendly nature of deep eutectic solvents for macromolecules, limited experiments have been conducted on the reverse solute flux parameters. It is worth mentioning that deep eutectic solvents can represent a benign nature towards macromolecules, as a result quality of feed was unaltered.

3.4 Membrane properties and fouling behavior

Membrane properties can also significantly influence the performance of the FO membranes. There are some studies available in the literature which can lead the selection of the most sustainable membrane among the possible alternatives. Thin film composites have gained popularity to be used in FO processes for the treatment. Singh et al. [80] concluded that the water flux in HTI-TCF FO membranes is much higher compared to HTI-CTA and aquaporin-TFC membranes.[81]Surface modifications have also demonstrated to affect the thin film membrane performance a lot. For instance, Aziz et al. indicated that incorporation the incorporation of carbon nitride can considerably enhance the performance of the FO process for desalination purposes [82].

Stability of the membrane in order to tolerate the harsh environmental conditions is another property of the membrane which should be considered carefully when selecting a suitable membrane to conduct FO processes. By reviewing the synthetic polymer materials for FO processes, Xu and Ge concluded that CA/CTA membranes can undergo chemical

reactions under non-neutral conditions and may not be good candidates for FO processes[83]. Duong and Chung employed FO using NaCl as a standard draw solute to separate water from oil dispersions [84]. In their study, the performance of poly(acrylonitrile)-based TFC membrane (PAN-TFC) was optimized to separate different concentrations (500 mg/L to 20000 mg/L) of stable emulsions. Over 99.88 % of oil rejection was achieved at a relatively high product water flux of 16.5 and 11.8 LMH using 1 M NaCl. Furthermore, PAN-TFC membrane showed moderate antifouling properties in the presence of emulsified oil particles making the process quite efficient. In a similar study [85], dialysis bags were used for treating highly contaminated oily wastewaters. Figure 6 displays the lab-scale process to fabricate double-skinned FO membrane wherein flat sheet was converted to dialysis bag to separate water from oil using 2 M NaCl draw solution. The double layered membrane resulted in good quantities of product water recovery at the flux rate of 12 LMH. The double layered FO membrane resulted in a high-water flux of 17.2 LMH with an insignificant reverse salt flux for NaCl draw solute, out-performing the single skinned CA membrane. The most significant outcome of this study considering the sustainability factors was that double-skinned membrane exhibited higher resistance to foulants. The presence of skin layer limited the excessive diffusion of draw solute in to the bulk of the membrane pore preventing clogging [85].

Figure 6. Lab-scale steps for double-skinned FO membrane fabrication for oil-water emulsion separation using NaCl as DS, reprinted with permission from [85]. This membrane consists of a TCF layer and a porous support in order to promote the rejection of the oil.

Membrane surface properties can also influence the fouling process. Also, fouling is still a major concern and a limiting factor for extensive generic use of various types of membrane technologies[33]. In standalone FO process, fouling phenomenon has been extensively studied and documented. Elevated concentrations of both feed and draw solutes/solutions are the responsible for the scaling at both sides of FO membrane[86]. . Some studies have already discussed the fouling properties of membrane as a result of feed high concentration (BOD and other biological contaminants [86, 87]. Here, thin-film composite (TFCs) have been generally used with one active layer to control the fouling of the membrane [88]. Draw solution facing open channels in membranes are exposed to highly concentrated draw solute resulting high reverse solute flux. However, very limited number of published reports can be found in the literature on the effect draw solution concentration on reverse solute flux exposed to non-active side of FO membrane.

The fouling process can be reversible or irreversible. There are some attempts in the literature in order to investigate the cleaning of the membrane in case of reversible fouling for instance, physical cleaning approaches such as surface wash and osmotic backwash. In case of irreversible fouling, which would result in process failure, substitution of the membrane is the only viable option leading to increase the overall cost of the treatment process. Im et al. [89] used synthetic seawater as the draw solution for real secondary wastewater effluent (feed solution) treatment. After the long-term use spiral wound forward osmosis process flux was drastically reduced from 6.0 to 3.2 LMH. Later membranes were studied for structural effects and fouling patterns. Highest organic content fouling of 2.67 mg-c/cm² in the inlet of SWFO was observed that was irreversible. The study concluded that liophilic organic foulants should be eliminated at pre-treatment step to reduce irreversible fouling. This is accounted for the formation of hydrophobic layer on the hydrophilic layer, which is developed initially on the membrane surface.

Zhao et al.[90] used cleaning agents such as sodium dodecyl sulfate (SDS), ethylene diamine tetra acetic acid (EDTA), and NaOH for efficient cleaning of FO membrane surface in 15 min by restoring its initial water flux. Further, Cartinella et al.[91]studied the concentration and removal of natural steroid hormones in wastewater in a FO process. The study indicated that two natural steroid hormones, estrone and 17-estradiol were retained with a rejection of >99.9% for urea and ammonia draw solution at the higher water recovery.

Air scaring or bubbling is considered to be another effective method to deal with membrane fouling. Sun et al. [92] employed an effective and simple air scaring method for municipal wastewater treatment to reduce membrane fouling in a FO system. The effect of air flow rate was optimized to $3.0 \text{ m}^3/\text{m}^2\cdot\text{h}$ to obtain significant improvements in water flux even after 52 days of operation. In a normal FO process, formation of a dense fouling layer onto membrane surface was possible due to the presence of bacteria, organic and inorganic matters, but in the presence of air bubbles, accumulation of foulants onto membrane surface may be reduced, but flux was increased.

Despite the potential benefits of FO for wastewater treatment, still some bottlenecks exist, which restrict its rapid commercialization. For instance, the majority of tested FO membranes are sensitive to bio-fouling despite many efforts in place to overcome the same exists. For instance, Nguyen et al. [93] utilized the ultrasonic irradiation to protect the membrane from microbial binding and bio-fouling due to sludge deposition. Under ultrasound irradiation of 40 kHz for 22 h were required to reach the sludge concentration of 20400 and 28400 from the initial sludge concentrations of 3000 and 8000 mg/L. While, in absence of ultrasound time required was 26 h to achieve the same. In order to reduce the sludge deposition, FO membrane maintained high removal efficiencies for $\text{NH}_4^+\text{-N}$ (96%), $\text{PO}_4^{3-}\text{-P}$ (98%) and dissolved organic carbon (DOC) (99%). In another study, dewatering of coal tailing slurry was achieved using the commercial CTA FO membrane [94]. The authors collected different slurry samples having various osmotic pressures, particle sizes and solid weight percentages to evaluate their dewatering efficiency. Over two-fold concentration of slurry was achieved in this study from an initial solid content of 30%. Addition of flocculants and gypsum also promoted FO dewatering process due to the disruption of cake structure.

574 Table 1. A Summary of parameters and conditions used in stand-alone FO used for treating polluted water/wastewater to extract the reusable
575 water.

Feed	Draw solution (DS)	Membrane	Flux (LMH)	DS Recovery	Value addition	Energy consumed (kWh _{th} /m ³)	Ref.
Activated sludge from MBR	Simulated RO brine	Asymmetric CTA (HTI) (CTA-HTI)	3.3 – 4.53-3-4.5	No	Real-scale applicability applied to RO reject stream	-	[58]
Drilling mud & + Fracturing Water	280 g/L NaClaq	CTA-HTI	~2.0-5	No	No	275±12 kWh _{th} /m ³ +8.5 kWh _e /m ³	[52]
Fracturing flowback wastewater	NH ₃ /CO ₂	polyamide membrane (OasysWater)	~2.6 (average over ~100h)	Yes	No	(Evaporative Desalination- 633 kWh _{th} /m ³ + 1-8 kWh _e /m ³)	[53]
Simulated oil-water emulsion	1-4M NaCl solution	PAN-TFC	Maximum 19.6±1.0 (PWF) to 11.8± 1.0 (200K ppm)	No	No	-	[84]
	0.5 M NaCl	Double skinned (PAN support coated on one side NEXLAR copolymer, other side TTFC)	10.9 (PRO) FO mode not tested	No	No	-	
Freshwater/ Marine microalgae broth	Simulated crude glycerol	Dialysis Bag	~4 (Freshwater)- 2.75 (Marine)	No	Applicability of Waste/Crude Glycerol from biodiesel plant	-	[61]
Simulated dye water (50 ppm)	Hydrolyzed PAM (HPAM)	Polyamide-TFC FO membrane	2.4-3.2	No	Diluted HPAM can be used for flooding oilfields.	-	[63]
Produced water (Shale production)	NaCl brine	CTA(HTI) ,TFC-HTI, TFC-HTI surface modified	Normalized flux data	No	-	-	[24]

oil & gas produced water	35 g/L NaCl	Polyamide-TFC-FO	9-7.5	No	Environmental/economic benefits; comparative life cycle; economic FO; cost assessment with	-	[46]
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4. Hybrid FO systems for wastewater treatment

Combinations of FO with other existing treatment technologies have been also used to satisfy the sustainability criteria and to improve treatment efficiency as well as to enhance the quality of the treated water for re-use. From the OPEX point of view, most of the FO processes have been implemented as pre-treatment step. This section gives an overview on the integrated FO-MBR methods reported in the literature for the treatment of polluted waters and wastewaters. With additional focus on to direct use of diluted draw solute/solution may lead to the real problem-solving capabilities of hybrid processes. However, FO-MBR has proven limited practicality. The overall system efficiency can be maximized using a combination of different processes (membrane-based with other technologies) while treating the complex feed streams. Further, the diluted draw solute has been recovered via pressure driven membrane process. Nevertheless, containing major foulants and to reduce operational stress, FO has shown limited potential.

4.1 FO-MBR

Integration of membrane bioreactors (MBRs) with FO is an attractive approach that has created much interest in industrial and academic sectors. MBRs utilizing other treatment technologies such as microfiltration (MF) or ultrafiltration (UF) systems are in much demand. Also, the FO assisted membrane bioreactors (FOMBRs) have indicated great potential to treat highly-concentrated feed mixture by optimizing the operational parameters.

Figure 7. Schematics of continuous-flow laboratory-scale OMBR apparatus [95].

Achilli et al. [95] utilized the commercial CTA-FO membrane along with TFC RO membrane for activated slug treatment using an OMBR as presented in Figure 7. For a draw

solution of 0.5 M NaCl, the treatment efficiency of 6.2 L/m².h at 20 ± 2°C. Laboratory scales OMBR processes have been also designed to explore the effects of various draw solutes on the overall treatment efficiency for the polluted effluents. As an example, the efficiency of monovalent (NaCl, NaNO₃) and bivalent ions (MgSO₄ and ZnSO₄) draw solutions was compared for water recovery from an activated sludge [96]. The monovalent ion salts performed better than the bivalent draw solution.

Alternatively, Qin et al. [97] designed the novel submerged FO process integrated with MBR system for water recovery from the meat extract simulated feed using a series of commercial CTA membranes for extracting potable water achieving maximum pure water flux (PWF) of ~18-12 LMH. However, average flux dropped down to 9 LMH over 30 days of operation with a relatively low reverse transport of solutes from DS into the mixed liquor. This study complemented well with MBR by rejecting 98% of organic carbon and 90% of ammonium-nitrogen.

The above-mentioned systems were also tested for their resistance to fouling that showed acceptable performance. For instance, a pilot scale was performed by Zhang et al. [98] using FOMBR setup for over 73 days of operation that gave reduced bio-fouling and a stable osmotic flux of 3 L/m².h even at the elevated mixed liquor feed salinity. These systems observed reduced fouling by using a continuous air plug at the feed side of the pilot FOMBR for treatment of a domestic sewage treatment, which is a source of bio-wastes. A steady state flux of 3 and 7.2 L/m².h was achieved using NaCl and MgSO₄ with different osmotic pressure draw solutions, respectively, over 150 h of operation. Another study by Lay et al. [99] proved the role of air injection for fouling mitigation, wherein it was observed that absence of air injection reduced the flux (30 %), resulting in lesser water recovery due to decrease in membrane stability. Air injection provided safe interfacial passage to transportation of water molecule with continuous membrane surface cleaning.

The configuration of an integrated system demonstrated significant impacts on the overall treatment efficiency of waste streams. For instance, Zhang et al. [98] used hollow fiber membrane module in submerged configuration to recover water and other value-added products in FOMBR process. Two configurations including: (a) active layer facing draw solution (AL-DS) and (b) active layer facing feed solution (AL-FS) were investigated. The feed solution containing mono and divalent ions, bacteria and extracellular polymeric substances passing through the membrane porous substrate created an interfacial contact with the selective layer. Using the former configuration, 45% flux decrease was observed. But when the configuration was reversed (AL-FS), flux decline was negligible due to fouling by

the blockage of membrane pores by the bacterial cluster and extracellular polymeric debris in addition to internal concentration polarization due to the migration of inorganic ions to the selective layer. The authors suggested that AL-FS dynamic configuration saves energy and is productive. In another study [100] on integrating membrane-based technologies such as anaerobic digestion and FO treatment was developed that was coupled with anaerobic MBR to enhance clean water recovery from the municipal wastewater. Here, FO was the pre-treatment step using a series of draw solutes/solutions and the process considerably improved methane production yield as well as treatment efficiency. On the other hand, Neoh et al. [101] used ionic draw solute such as sodium acetate that resulted in high and stable flux, thereby promoting anaerobic treatment step.

Huang et al. [102] used two different types of FOMBR including mesophilic FOMBR and ambient temperature FOMBR under similar operating conditions. Both these systems showed high efficiencies for the removal of $\text{PO}_4^{3-}\text{-P}$, $\text{NH}_4^+\text{-N}$, and total organic carbon (TOC) from the effluent. Mesophilic FOMBR demonstrated a lower flux compared to ambient-FOMBR. In a similar study, Wang et al. [103] achieved 96% and 100% removal of $\text{PO}_4^{3-}\text{-P}$, $\text{NH}_4^+\text{-N}$ and TOC, respectively for 1 M NaCl draw solution using FOMBR. The phosphorus was intermittently collected from concentrated portion (890–990 mg/L) of the feed by adjusting the pH using sodium hydrogen phosphite hydrate solution. Qiu et al. [104] employed MF-FOMBR for the treatment of real municipal wastewater using a bio-based FO membrane that showed enhanced removal of organic matter and nitrogen. The use of biofilm obviated the need for additional solid/liquid separation (MF) and assisted in efficient recovery of phosphorus and reduced the salt accumulation at the draw solution side. Fig 8 represents schematic of the process as well as actual images of the phosphorus recovery.

Removal of heavy metals and persistent pollutants has been intensely studied in the literature using FO technique. Aftab et al. [105] studied heavy metals removal using FOMBR consisting of CTA membrane in comparison to TFC membranes. The sludge supernatant containing 2, 5 and 10 mg/L of chromium and lead in the feed were studied. The CTA membranes have shown the resistance against biodegradation that are widely used in FO. In a comparative study by Luo et al. [106], both CTA and polyamide TFC membranes were subjected to biodegradation by exposing the membrane to the activated sludge. The prolonged exposure to activated sludge resulted in increase of water and reverse salt fluxes for CTA membrane.

Figure 8. Schematics of the MF-FO membrane bioreactors (MF-FOMBRs) and bio-film-forward osmosis MBR(BF-FOMBR) systems with subsequent phosphorus recovery from the side-streams, adopted from [104].

It was observed that both CTA and polyamide TFC did not satisfy sustainability considerations of FOMBRs in terms of efficiency, hence new approaches were suggested for designing new membrane configurations. The surface modification of CTA was done by Li et al.[107] with polydopamine (PD) coating and poly (ethylene glycol) (PEG) grafting (PD-g-PEG) to improve the stability of FO membrane for application in a submerged FOMBR configuration, which reduced the biopolymer adhesion and bio-cake formation on the membrane surface.

Sun et al. [108] through surface characterization studies concluded that fouling is more prevalent with FO than FOMBR. Further, Wang et al. [109] used β -D-glucopyranose polysaccharides as a biofouling agent to assess anti-fouling properties of both commercially used FO membranes (CTA and polyamide-TFC). The polysaccharides and microorganism accumulation were $\sim 77\%$ of the total biovolume on CTA FO membrane, while polysaccharides were the only dominant bio-foulants on TFC FO membrane.

4.2 FO-RO

FO-RO systems are widely studied combinations of hybrid processes wherein one process acts as pre-treatment and another as the post-treatment, depending on the nature of the feed and draw solutions used. Such combinations have recently gained much attention for targeting the specific applications. For instance, FO can be used as an efficient and economic pre-treatment in RO wastewater treatment systems, more efficient than other pre-treatment

techniques such as MF, which may bring some limitations in reducing COD, BOD and organic matters in the wastewater content which can create serious threats to the RO membranes. Also, such combinations are considered as good candidates to enhance the product water recovery for seawater desalination.

Qin et al. [110] designed FO-RO hybrid system in which FO was used as a pre-treatment step for RO. This combined system worked in synergy with RO, resulting in unstable water flux and retention of the foulants from the nutrient-rich digested biomass. The FO retained 70% of the final recovery in RO when applied in both continuous and bench-scale modes, which offered high water flux and good nutrient rejection. Holloway et al. [111] used modeling approaches on a pilot FO/RO process that has yielded 70% of water recovery. Other studies include the importance of FO as a cleaning step after the RO as for instance suggested by Qin et al. [86] for the reclamation of secondary municipal effluent in a RO process. Here, FO step reduced considerable amount of salinity and turbidity (~15 NTU to 3 NTU). Hancock et al. [112] designed a comprehensive FO system in both bench and pilot scales for treating the secondary wastewater coming from a full-scale treatment plant. The authors used Red sea water as the draw solution in combination with HTI-CTA membrane to recover the trace amount of TOCs with a flux of 5-1.5 L/m².h for over 7 days of experiments. These data suggested that FO and low-pressure RO may consume only 50% (~1.5 kWh/m³) of the energy compared to high pressure seawater RO desalination processes even after running up to 14 days of continuous operation. The hybrid FO/RO showed low degree of fouling retaining over 98% flux, which was removed with simple air purging. Also, recovered product water showed an acceptable quality for applications in irrigation, aquaculture, etc. Boo et al. [113] used FO to investigate the effect of organic foulants in seawater desalination and wastewater treatment using synthetic sea water as the draw solution (30 g/L) against effluent feed contaminated with TOC, inorganic ions and humic acid. The effluent containing more than 20 types of different pollutants was treated in a commercial RO spiral wound membrane for up to 40 days with ~99% rejection of the pollutants, resulting in high-quality water reclamation. Researchers further combined FO and RO in two different configurations (Fig 9) to assess the process efficiency and quality of water recovered.

Figure 9. Schematics of hybrid FO-RO pilot systems adopted from [112], showing hybrid FO-RO process in two configurations: (a) closed loop DS in which FO recovers water from an impaired stream and RO re-concentrates DS for reuse and simultaneously produces purified water and (b) an open loop DS in which FO recovers water from an impaired stream that dilutes the pre-treated seawater from seawater RO.

Hau et al. [114] explored the potential of EDTA sodium salt as a novel draw solution in a hybrid FO for dewatering high nutrient containing sludge. They tested simulated feed mixture of sodium alginate (2 g/L), BSA (2 g/L), natural organic matter (2 g/L), which was collected from a river stream and 7 different salt mixture concentrations ranging from 0.6 to 2.0 mM. Comparative studies on draw solution were also performed on raw seawater and RO brine to obtain pure water flux of ~ 7 and $9.5 \text{ L.m}^{-2}.\text{h}^{-1}$, respectively. The focus of the study was also to monitor fouling layer caused by the organic pollutants on membrane surface. With extensive data analysis, authors proposed chemical methods of cleaning by controlling physical and operating parameters which included three approaches in order to minimize the fouling viz., (i) varying the feed and draw solution cross-flow velocity, (ii) introducing the turbulence through feed channel spacers and (iii) operating the FO in a pulsed flow mode. Commercial CTA membrane was used to concentrate the synthetic sludge comprising glucose, NH_4Cl , K_2HPO_4 and sediment raw sludge from the secondary sedimentation tank using EDTA sodium salt (0.1 to 1M) as the draw solution. In the mentioned study, NF having less energy consumption than RO was also used to separate water from the diluted EDTA sodium salt. However, the mentioned study did not intend to calculate the energy consumption of the studied systems. On the other hand, the study focused on the developing novel draw solutions and proposing possible draw solutes recovery methodologies. There is a scope in future to use similar draw solute (EDTA) in both pristine and modified forms for different applications and can come up with comparative data to show the possible advantages of FO over other pressure driven membrane processes. In this regard, sustainability criteria for the scaling-up the FO technologies dictates the development of

solutes recovery technologies with low energy requirement to satisfy the economic considerations, as a main pillar of sustainability. Therefore, there is an urgent need for the economic evaluation of the already developed technologies in order to investigate the feasibility of such technologies for large-scale applications and the role of solute recovery to make the FO technologies appropriate for real wastewater treatment purposes. The FO has been also used to recover ammonia, nitrate and phosphates (>90 %) from the concentrated sludge, wherein NF has helped to recover Na-EDTA from the diluted draw solution. Kumar et al. [115] performed water recovery feasibility tests from the raw coke oven wastewater obtained from ammoniacal-N liquor tank. In the first step of FO, a maximum flux of $\sim 48 \text{ L.m}^{-2}.\text{h}^{-1}$ was obtained from NaCl draw solution, whereas a min flux of $24 \text{ L.m}^{-2}.\text{h}^{-1}$ was achieved in their study with 2.5 M NaCl, MgSO_4 and CaCl_2 as draw solutions. Overall, > 96–98% of cyanide, phenols, $\text{NH}_4^+\text{-N}$ and COD were rejected from the real coke-oven. Furthermore, NF process produced reusable water at a rate of $45 \text{ L.m}^{-2}.\text{h}^{-1}$ with 92 to 98.5% NaCl rejection from the diluted draw solution. Coal mining normally consumes high amounts of water, which is contaminated with high concentrations of compounds such as sulphates, silica and other toxic materials at different stages of mining process. Thiruvengkatachari et al. [116] explored the efficiency of hybrid FO technologies to deal with highly polluted coal mining effluents. The process successfully recovered and recycled the fresh water by an integrated FO process using di-sodium hydrogen phosphate (DHSP), sodium hexametaphosphate (SHMP) and sodium lingo sulphonate (SLS) as draw solutions, further, RO was used to recover draw solutes.

Unlike the standalone RO processes used for wastewater treatment applications with limited product water recovery potential, FO-RO combinations can represent up to >70% higher water recovery potential. Enhancing the water recovery potential may compensate for a part of the energy used for FO process. These FO-RO hybrid processes can also reduce the economic costs in terms of OPEX cost by $\sim 10\%$ compared to only RO process used for product water or reusable water recovery. Sustainability parameters also requires cost-effective performance of hybrid systems with minimum energy imprints and maximum productivity. In this direction, Cath et al. [117] employed a FO and low-pressure RO (FO-LPRO) hybrid systems to demonstrate the economic feasibility of such a combination for desalination and wastewater treatment. Large set of data collected showed the advantages of combined hybrid system in saving both capital (CAPEX) and operating (OPEX) costs while treating some complex effluents in a FO compared to other conventional wastewater treatment processes (see Figure 10). Extensive data evaluated by Valladares Linares et al.

[118] also concluded that FO-LPRO system has 7% lower CAPEX and 9% higher OPEX compared to all other hybrid systems including FOMBR.

Figure 10. Operational expenses (OPEX) for SWRO and FO-LPRO units in % from the total annual cost of a 100,000 m³/d plant, 39% of the cost represents amortization of CAPEX, adopted from [118].

4.3 FO-NF

As discussed before, the economic considerations are among the most important criteria in membrane technology applications. The development of NF in place of RO along with FO appears to be more economic and technically viable. So only NF is gaining more momentum compare to RO in recent times for many application including ground water desalination. Phuntsho et al. [119] designed a pilot scale FO-NF system (Figure 11) using ammonia and sulphate-based fertilizer as the draw solutions for investigating ground water desalination and inorganic contaminant removal from the coal mining wastes. The process provided the required rejection for NH_4^+ and SO_4^{2-} and the low rejection for feed salts (Na^+ , Cl^-) was observed by FO, while the CTA-FO membrane suffered from serious draw solute cake formation, resulting in a reduced flux.

Hybrid NF-FO systems are investigated for treating high nutrients sludge concentration by dewatering the wastewater. In a study by Hau et al. [114] used EDTA-Na salt to concentrate the sludge using FO-NF integrated system to demonstrate low reverse salt flux to concentrate the sludge up to 32,000 mg/L in 16h. Further, NF was employed for efficient recovery of EDTA. Kumar and Pal [115] also treated the coke-oven wastewater by FO-NF to observe the system in terms of flux and effects of operating parameters such as pressure, cross-flow rate, pH of the feed solution and the duration to achieve 96–98% removal of cyanide, phenols, NH_4^+ -N and COD from the real coke-oven using 1.5 M NaCl as the draw solution at 45 LMH.

Figure 11. Pilot-scale FO-NF desalination testing center located at the Centennial Coalmine site, state of NSW, Australia (up). Schematic diagram of FO-NF desalination system used for pilot-scale studies (down), reprinted with permission from Phuntsho et al. [119]. This pilot-scale plant uses ammonia and sulphate-based fertilizer as the draw solutions.

4.4. FO-MD

Conventional membrane distillation (MD) normally uses heat and vapor pressure to drive the separation and purification of wastewater. MD is a thermally driven process where only vapor molecules transfer through the microporous hydrophobic membrane. Vapor pressure difference across the membrane drives the transfer and separation of product water. A schematic of the process is shown in Fig. 12. Highly metabolic and biological wastewater can be safely treated using MD as the vapor selectively passes through hydrophobic membrane. On the other side of the membrane, chilled water condenses vapor to produce fresh water and the process also known as dual membrane contractor and is energy intensive.

Figure 12. Schematic view of membrane distillation process.

Martinetti et al. [120] employed osmotic process in combination with MD to recover high concentration brines to treat metabolic wastewaters. This process used FO with direct osmosis (DFO or DO) and the modified MD, called membrane osmotic distillation (MOD) required the use of temperature and concentration gradient as the driving forces. The FO-assisted MD showed remarkable increase in flux (~25 times) with a slight increase in temperature (3-5 °C) and >99.99% rejection of urea. Realizing the potential of FO-MD process, Ge et al.[121] designed another system to deliver effluents to FO from the MD by vacuum assistance. For FO, poly(acrylic acid) sodium (PAA-Na) salt was used as the draw solutes to enhance dewatering of sewer mining effluent. This configuration also extended to recover high quality water from the RO rejected brine with TDS ranging from 7500 to 17,500 mg/L. However, vacuum MD did not achieve enough water recovery, restricting to 81% due to precipitated salt onto the membrane surface causing the flux decline, while FO produced >90% water recovery in FO-MD hybrid process. The PAA-Na dewatered feed waste and diluted PAA-Na were recovered in the MD process. The efficiency of PAA-Na in dehydrating wastewater in the FO process showed a regeneration capacity of 0.48 g m/L at 66°C in MD step making it as one of sustainable hybrid processes for wastewater treatment.

The recovery studies on elements from the effluent are considered as one of the main parameters needed to increase sustainability of a desired method. In this regard, Xie et al. [122] utilized FO-MD hybrid process to extract the valuable phosphorous from the digested sludge. The robustness of the hybrid FO-MD was tested in continuous lab scale mode using raw sewage feed with >80% recovery of water. However, modifications in MD increased the TOrCs concentration in draw solution to recover water (see Fig. 13). Alternatively, it was proposed to add granular activated carbon for adsorption or UV oxidation to prevent the contaminant accumulation.

Zhang et al.[123] used the FO-MD process for sustainable water recovery from the oily wastewater in which both phosphorous in the form of struvite ($\text{MgNH}_4\text{PO}_4 \cdot 6\text{H}_2\text{O}$) and reusable water were successfully extracted. Here, researchers used MD as the recovery step for draw solution (MgCl_2). During feed concentration in FO, a significant amount of acetic acid was retained, which was further used as an additive to crude oil production in the MD process. This work demonstrated that not only water, but also organic additives in wastewater may be effectively recovered by the FO-MD systems.

Figure 13. Schematics of FO-MD hybrid system with a mitigation loop for wastewater treatment. Reproduced with permission from [122].

Husnain, et al. [124] integrated the FO-MD using hollow fiber membranes for the treatment of oil-water emulsions having the droplet sizes $<1 \mu\text{m}$. This hybrid FO-MD system was implemented for the removal of ammonium, COD, arsenic, and combined solutes in both synthetic and real wastewaters to offer $\sim 99.9\%$ rejection for all the contaminants. In this study, FO was performed as an up-stream pre-treatment process for the removal of foulants from the feed helping the downstream MD for water recovery with a minimal membrane fouling. Secondly, the integrated process successfully recovered the draw solution for FO process. The oily wastewater with a relatively high salinity was effectively recovered (90%) by the FO-MD hybrid system.

Liu et al. [125] used the FO-MD integrated system for human urine treatment in which a series of NaCl solutions at different concentrations were adopted as draw solutions and MD was designed for NaCl recovery. At the end of individual draw solute studies, four stable equilibrium conditions were optimized to match the water transfer rates of individual FO and MD processes. It was found that the integrated system is stable, sustainable and works synergistically when water transfer rate of FO subsystem is equal to that of the MD

subsystem. However, the authors did not discuss exactly which pillars of sustainability can be counted as the potential advantages of the hybrid system.

Besides the benefits of such integrated systems, some drawbacks also existed such as the case of integrated FO-MD system showed relatively smaller rejection for TOC, total nitrogen (TN) and ammonium nitrogen ($\text{NH}_4^{(+)}\text{-N}$) from the human urine compared to FO process[125]. Nguyen et al. [126] employed the highly charged phosphate as the draw solute to generate fluxes as high as 12.5 LMH in FO-MD system for concentrating high-nutrient sludge. The draw solution and product water from the diluted Na_3PO_4 were achieved using four different types of polytetrafluoroethylene membranes in a MD operation. Highly polluted waters were studied for waste treatment using such hybrid systems. In this venture, water reclaiming from the shale gas drilling flow-back fluid (SGDF) was investigated by Xue-Mei et al.[127] using a hybrid FO-vacuum-MD system to achieve ~90% water recovery by implementing FO as the pre-treatment step to remove the most contaminants and constituents that would otherwise cause membrane fouling in subsequent MD process.

4.5 FO-ED

Electrodialysis (ED) is a technique that is based ion exchange membranes to transfer ionic species from the concentrated to dilute chamber by applying an electric potential difference[128]. Combination of FO with ED as configured in Figure 14 can be an effective system for effluent treatment. In this study, high quality reusable water was recovered from the secondary wastewater effluent concentrating TOC, carbonates, and cations. However, draw solute can be recovered using solar energy assisted ED in a cost effective step, which would meet the sustainability criteria[129]. On the other hand, a study was conducted [130] to concentrate the trace pharmaceutical contaminants from the bio-waste effluents using FO in combination with electrochemical oxidation (FO-EO) process to recover trace antibiotics. At a current density, $J=1 \text{ mA/cm}^2$, >98% rejection of antibiotics was achieved and the method was proved safe for disposal.

Figure 14. Water and salt balance in FO-ED system adopted from [129].

4.6 FO-MFC combinations

Microbial fuel cell (MFC) is an emerging area to generate clean energy satisfying the sustainability criteria. Recently, FO assisted MFC (FOMFC) was developed that gained much significance in the treatment of effluents from various origins. The module design of FOMFC where FO membrane performs as a separator is shown in Fig. 15. Here, two different modes are followed: (A) to recycle water from the waste for reuse and (B) direct seawater desalination known as microbial desalination cell (MDC)[50]. The performance of FOMFC was examined using both NaCl and seawater as a catholyte (draw solution). However, FOMFC can produce more electricity than MFC in batch experiments, which may be attributed to improved proton transport with water flux through FO membrane[25].

In a similar study, an air-cathode FOMFC design was developed to improve the energy recovery[50] where the use of an appropriate FO membrane could restrict the internal resistance, resulting in a high maximum power production (43 W/m^3). The optimum removal of carbon-based compounds (acetate) ($\sim 90\%$) was reached in the FOMFC with a stabilized flux of 4.1 LMH . The MDC system was also adopted for wastewater treatment in an energy-efficient design [131]. Organic solutions treated in a MDC process was subjected to water recovery in a FO mode, which resulted in reducing the wastewater volume by 64% and improved the conductivity reduction in saline water by two-times. However, MDC is currently in its early developmental stage and more studies are needed to enhance its sustainability criteria for adoption to solve practical applications.

Figure 15. Proposed applications of forward osmotic microbial fuel cells (FOMFCs): (A) for water reuse and (B) for seawater desalination (MDC-microbial desalination cell), reprinted with permission from Zhang et al. [25].

In a completely different approach[132], microbial electrolysis (ME) was designed in combination with FO to recover ammonium and water from a synthetic effluent, in which with ME, >60% ammonium was recovered at the cathode aeration using 2 M of NH_4HCO_3 as the draw solution, while in FO, >50% water recovery was achieved from the ME anode effluent, which is higher than the raw leachate. The study successfully demonstrated usefulness of ME-FO process for the recovery of ammonia and re-uses it as a draw solute in FO for successful water recovery from the treated leachate. For instance, Yuan et al.[133]employed mathematical model for pre-data assessment of an actual domestic wastewater treatment amended with glucose and NaHCO_3 . The FO-MDC system thus generated desalinated effluent containing 1130 mg TDS/L.

4.7 MF-FO-MBR and UF-FO-MBR

Generally, MF and UF membrane operate at low pressures, resulting in high water fluxes. The use of MF or UF membrane filtration can therefore be efficiently used to remove contaminants, which may cause serious membrane fouling during high pressure operations. MF-assisted membrane photo-bioreactor (MFMPBR) and FO-based membrane photo-bioreactor (FOMPBR) have been designed by Praveen et al. [134]with *Chlorella vulgaris* for tertiary wastewater treatment. The FOMPBR achieved high nitrogen and phosphorus removal (86-99% and 100%, respectively), while MFMPBR process resulted in 97% and 46% removal

efficiency, respectively. Promising results make the process suitable for microalgae-based tertiary wastewater treatment.

Figure 16. Schematic of (a) FOMBR system used for long-term evaluation during Phase 1 and (b) UFO-MBR system used for long-term evaluation during Phase 2 of the study, reprinted with permission from Holloway et al. [135].

Qiu et al. [136] used a combination of MF and FO bioreactors (MF-FOMBR) to recover >70% of the nutrients including PO_4^{3-} , Ca^{2+} , Mg^{2+} , etc., from the municipal wastewater using sea water as the draw solution. Utilization of sea water draw solution not only resulted in nutrient enrichment (>97% PO_4^{3-}P), but also resulted in the TOC (>90%) and $\text{NH}_4^+\text{-N}$ removals. Further, the authors [113] designed a similar MF-FO process by manipulating contaminants retention time and hydraulic retention time to remove TOC and $\text{NH}_4^+\text{-N}$. Holloway et al. [135] used FOMBR process combined with UF to extract the reusable water from the activated sludge and to concentrate the dissolved constituents such as total nitrogen (TN) and total phosphorus (TP), inorganic salts and other nutrients. The authors designed FO integrated systems with (a) aerobic and (b) UF for sludge treatment by simultaneously recovering water. Thus, application of FO not only helped to recover >90% of contaminants and total removal of COD, but also significantly reduced the fouling of MBR.

In conclusion to this section, it can be stated that hybrid FO systems can demonstrate a remarkable potential compared to standalone processes in terms of lower energy

consumption, especially for the treatment of highly impaired feed streams. FO-MD and FO-MBR configurations have proven to be quite promising for fouling mitigation and maintaining system's integrity over prolonged operation times. In case of MD, low grade waste heat from power plant's cooling towers, solar energy and even geothermal latent heat can be the driving force to achieve separation. However, from the point of view of commercialization, several bottlenecks still plague the MD process including membrane wetting, low recovery rate and scaling up capabilities. For seawater desalination application, hybrid FO-RO processes have shown better recovery rates and lower brine production ratio. Standalone FO has also proven to be effective pre-treatment step to reduce bio-fouling and scaling in subsequent SWRO process. Due to low and often reversible fouling propensity, FO processes have an edge over conventional pressure-driven pre-treatment processes viz. MF and UF.

In order to be economically viable with long-term operational sustainability and low OPEX, better FO modules along with efficient and fouling resistant membranes will have to be commercialized. As mentioned in earlier sections, nanomaterials like carbon nanotubes, graphene & its composites and various metal-based nanoparticles have been used in membranes to mitigate organic and bio-fouling. But leaching out of these fillers during long-term operation remains a challenge.

An in-depth comparison between various hybrid FO processes has been tabulated in the supporting information section (Table S1). Various sustainability parameters including draw solution recovery method, energy consumption and value-addition prospects have been compared.

5. FO as a pre-filtration step

Several studies have shown ways to implement FO as an intermediary or pre-filtration step to the existing wastewater treatment techniques. This approach is the widely regarded as feed systems with highly concentrated solutes and contaminants like high BOD and TPs in wastewater are expected or anticipated to spoil the main treatment method set-up like NF and RO process. However, there is a need for further studies to overcome the still existing scientific gaps in this regard. One of the major concerns and challenges in the development of FO processes and their commercialization is the lack of suitable FO membranes with suitable pore structures. So far, popular TFC RO membranes have been explored for many FO process. There is currently a need to develop the FO membranes with two active layers to perform (facing feed and draw solutions) independently. Also, finding suitable draw solution and recovery protocols are also of high importance in this regard. Development of the membranes with less fouling potential can also promote the application of the membranes for such applications.

Some feasibility studies have been previously conducted using commercial FO membranes (such as HTI, TFC-PA, etc.) to study the membrane fouling properties in the FO membranes used for pre-treatment of the effluents. Development of high efficiency FO membranes can also mitigate the fouling in the subsequent steps in case a combination of membrane processes is desired to be applied for the combined treatment of polluted waters and wastewaters. In this regard, there have been several attempts [137] to develop novel FO membrane that are resistant to fouling in order to sustain long term operations. In addition to conventional cleaning methods employed so far, identifying the interaction between (i) pollution load, foulants and membrane, (ii) chemical change during concentration or removal process of different contaminants, (iii) chemistry of contaminants and process parameters, and (iv) membrane functionality, intermediate contaminants and cleaning chemicals are to be identified.

In efforts to use FO as a pre-treatment step to maintain its chemical, structural and mechanical integrity, FO process was used [138] as a preliminary step for the coagulation/flocculation (CF) of textile wastewater. Here, polyamide TFC membrane was used to concentrate synthetic textile wastewater from the textile dyes, inorganic salts, surfactants and organic additives, which are normally used in textile dying processes. By using 2M NaCl as the draw solution, a water flux of 36.0 LMH with a contaminant rejection

of ~99.9% was achieved. Thus, the use of FO as a dye enrichment step could remove >95% of dyes with a small dosage of coagulants and flocculants.

In another study[139], pre-treated municipal wastewater was treated using a commercial membrane from aquaporin and HTI-CTA at the lab scale. The study documented a stable flux with a high efficiency for BOD, TP and other inorganic salts removal regardless of the type of wastewater. Another study discussed about pilot scale units designed to treat raw municipal wastewater using FO as pre-treatment step, wherein coagulation, flocculation, micro-sieve and MF were used as pre-treatment step in different configurations with FO satisfying the regulatory norms for BOD, TP and salts [140]. Recently, municipal wastewater and synthetic feed water containing naturally occurring foulants such as humic acid, protein (BSA) and sodium alginate were employed to study the FO membrane fouling to demonstrate that these foulants could damage the membrane structure more severely than the real wastewater contaminants [141].

In recent times, extensive use of scanning electron microscopy (SEM) and zeta potential technique for in-situ or online monitoring fouling and solute deposits on the membrane surfaces opening new windows to understand the fouling phenomenon. Understanding solute deposition dynamics and solute/membrane interaction is the way forward to mitigate the one of the major issues FO facing. This will also lead to design efficient membrane for future. As in case of CTA and PA-FO membranes surfaces that were continuously monitored and characterized for salt deposition by measuring variations in zeta potential.[142] Polyamide membrane showed more affinity towards hydrocarbons, iron, silica, calcium, sodium, and chloride than the CTA, but TFC had a higher rejection for all aliphatic and polycyclic aromatic hydrocarbons than the CTA membrane. A study to assess fouling pattern on CTA and PA-TFC membranes for the analysis of oily wastewater against 2M NaCl as the draw solution [143]. The accumulation of organic foulants on the membrane surface was predominant whereas inorganic species caused concentration polarization in both the membranes. CTA membrane showed higher flux recover (~95%) upon back washing than PA membrane. The mentioned study also revealed some of important aspect regarding the choice of cleaning agents and protocols. Among all, osmotic back wash resulted in higher flux recovery on CTA membrane.

Generally, scaling of high molecular weight solutes and their scaling phenomenon is largely understood. The smaller size molecules may also play important roles in blocking the permeation path of membrane over time which has been to some extent ignored in the performed studies due to the importance of concentration polarization, intrinsic polarization

(which refers to the behavior of the draw solutes as potential foulants which will effect overall performance of FO process), and surface fouling. However, there is not enough information available in the literature on the reverse solute flux in connection with nature of polymers, membrane structure and pore morphologies. High concentration draw solutes/solutions may effectively act as foulants on the other side of membrane in the absence of suitable dual active face FO membranes.

Some studies have tried to address this issue. In a recent study, solute scaling on membrane surface and its impact on draw solution and membrane performance was thoroughly studied on a FO membrane [144]. General scalants like common salts, calcium carbonate (CaCO_3) and ammonia-carbon dioxide ($\text{NH}_3\text{-CO}_2$) in draw solution were used to understand the effect of solute concentrations on scaling. The insignificant changes in flux were noticed with NaCl draw solution whereas a drastic flux decline was recorded when $\text{NH}_3\text{-CO}_2$ was used as draw solution due to scaling phenomenon. Regular SEM analysis was also performed to analyze CaCO_3 scale formation on selective layer due to the reverse diffusion of carbonate species from the draw solution. Even though cleaning agents like EDTA was used to maintain constant flux, however study recommended following proper cleaning procedure while using $\text{NH}_3\text{-CO}_2$ as draw solution.

6. Conclusions and prospects

High quality water recovery rate using FO reveals recognizable technological advances in this area and its extraordinary growth prospective in the near future. However, there is an urgent need for sustainability analysis of the existing FO-based technologies to promote their application for the treatment of highly polluted industrial. Technical considerations are among the most important parameters, which may attract industries to select efficient methods which are able to deal with the produced effluents and to satisfy the stringent environmental standards.

There are some reports in the literature which indicate that membrane technologies are the leading methods for the treatment of the effluents from various origins. Although some technical bottlenecks still exist restricting the wider application of membrane technologies, yet they have repeatedly shown acceptable performance at the lab and pilot scale studies. Hitherto various configurations of semi-permeable FO membranes have been used for the treatment of effluents from different origins. Also, with the advent of nanotechnology, a new class of functionalized materials has been tested in FO applications

with special emphasis on sea and brackish water desalination. Promising examples include those based on the aligned carbon nanotubes, hybrid protein-polymer biomimetic membranes and thin film nano-composite membranes. Inorganic-organic, bioactive species like *aquaporins* and functionalized inorganic nanoparticle composite membranes are uniquely interesting because they exhibit enhanced water permeability, good solute rejections and improved interfacial properties like antimicrobial and photo-catalytic activities. Efficiency can be considered as the most important technical criteria when sustainability is the goal of selecting the most suitable technology to deal the highly polluted effluents. An overview on the literature published on the application of forward osmosis processes can indicate that such technologies, especially in the form of FO-based hybrid processes wherein FO is used as a pre-treatment step, can be successfully applied to treat various types of wastewater streams. Among the membrane desalination techniques, hybrid FO processes have performed better than other membrane-based technologies for the separation of concentrated water pollutants and sludge dewatering. In addition, there is a need to pay attention towards the importance of environmental and energy consumption of various designs. Future FO processes have to include sustainable and green chemistry origin draw solutions integrated with the recovery process for both product water and draw solution. Therefore, green, renewable and cost-effective draw solutes/solutions for easy recovery and reuse are to be further explored. In terms of ease of implementation, hybrid FO processes have proven to be economically facile for the treatment of highly polluted wastewaters. Finally, hybrid FO processes will have enormous value addition to the concentrated wastes as these processes do not affect chemical composition of the feed solutions. Future studies should focus on detailed economic analysis for those technologies which have proven their efficiency to promote commercialization. Studies are also required to compare the cost-effectiveness of FO processes with methods such as RO to aid the industries to select the most sustainable option among the existing technologies to deal with their effluents. More efforts are also welcome to scale-up some novel technologies which have successfully been demonstrated in lab-and pilot scales. Nanomaterials coated membranes are good examples in this regard but still require more deep studies of the applicability in real wastewater treatment applications.

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