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Comprehensive review of osmotic dilution/concentration using FO membranes for practical applications

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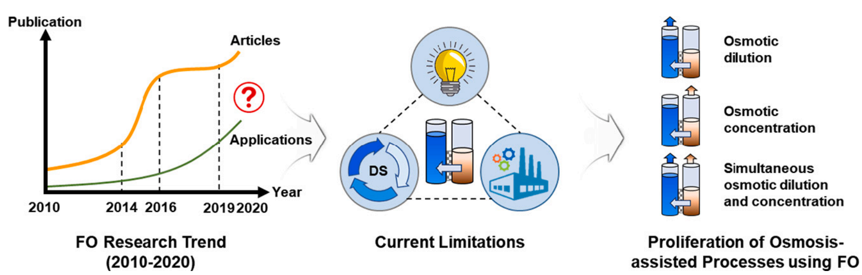
HIGHLIGHTS

- Critical review on the current state of osmosis-assisted processes using FO.
- FO applications limited by DS regeneration, high energy use, and scale-up.
- Osmotic dilution and concentration by FO without DS reconstitution.
- Commercial applications of osmosis-assisted dilution/concentration processes.

GRAPHICAL ABSTRACT



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ABSTRACT

Forward osmosis (FO) has attracted growing attention in the field of membrane-based separation technology over the last two decades. Despite recent advancements in various osmosis-assisted processes, few studies have succeeded in commercialization. This paper reviews the state-of-the-art developments of FO membrane, limitation analysis, and commercial proper applications. First, the development of FO technology in terms of FO membranes, FO draw solution (DS), and FO systems is reviewed. Based on a literature database survey spanning 1965–2020, current limitations of FO, particularly in terms of DS regeneration, energy consumption, and scale-up implementation, are identified to overcome the obstacles to commercialization. The key applications of the FO membrane process in commercial sectors are further classified into three configurations (i.e., osmotic dilution, osmotic concentration, and simultaneous osmotic dilution and concentration), and their successful applications are discussed. Since industrial demonstrations of simultaneous osmotic dilution and concentration are in progress, we believe that FO technology with no need for DS regeneration will be commercialized soon in the future.

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1. Introduction

The forward osmosis (FO) process has been receiving growing attention over the last two decades. Unlike reverse osmosis (RO), which is a pressure-driven membrane process, FO is driven by the natural osmotic gradient between two solutions with different salinities (i.e., feed solution (FS) and draw solution (DS)) for permeation of water through a semipermeable membrane [1]. FO exhibits a high fouling reversibility [2,3], and has a high rejection rate of undesired solutes and pollutants [4,5]. Despite early hype as a low energy process [6], FO application has been limited mainly due to the lack of ideal draw solution which produces high osmotic pressure and gets easily recovered [7]. Therefore, many studies have been conducted for the development of high-performance FO membranes, ideal draw solutes, and more efficient systems for successful applications, as presented in Fig. 1.

Extensive research has been conducted to resolve the challenges encountered by the FO process. Meanwhile, advances in technology have led to an improved performance of the FO process with regard to FO membranes, DS, and FO systems. Fig. 2 presents the state-of-the-art FO technologies in terms of publication numbers and research articles on the application of the FO process in the last decade. Overall, the number of published papers increased dramatically, although a slight stagnation was observed from 2016 to 2019. In addition, an analysis of the categories of all the research publications, research articles focusing on applications in various industries have increased steadily every year. This indicates that the FO technology has drawn the attention of researchers working for the commercialization of the FO process and has been applied in various industries. Notably, the highly scientific research output caused by FO application proliferation has accounted for the increasing proportion of the total academic article output, research into FO application showed a significant influence on the publication growth trend in 2020. Therefore, a critical assessment and review of the current state of commercial applications is necessary to further promote the FO process.

During an FO working process, FS is continuously concentrated and DS is diluted simultaneously, which can be called the concentration-dilution mechanism. Therefore, FO systems can be classified into four cases (Fig. 3): (i) the concentration of FO feed with additional DS regeneration, (ii) the dilution of DS to improve its value, (iii) the concentration of FO feed and discharge of diluted DS with no reuse, and (iv) the simultaneous concentration and dilution of FS and DS, respectively, to increase their values.

The first case (Fig. 3a) has been intensively studied because it can successfully achieve both water treatment and clean water production. Therefore, many researchers have proposed various FO configurations combined with novel DSs. Inorganic chemicals have been mostly examined as DSs for FO systems because they can be rejected by RO

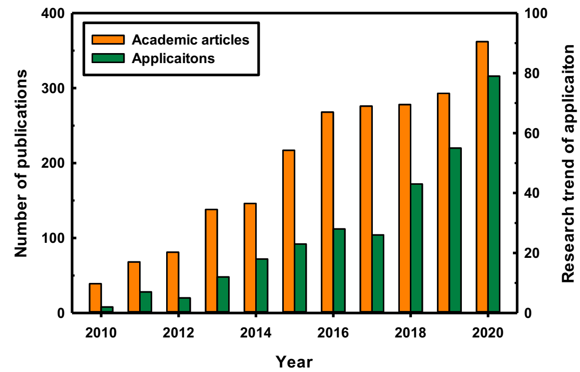

Fig. 2. Academic literature database from 2010 to 2020 extracted from Scopus with keywords (i.e., "forward osmosis" and "direct osmosis").

[4,8]. Thermo-responsive DSs have also been proposed because of their easy separation by distillation [9]. The second case (Fig. 3b-1) is osmotic dilution (OD), which has been receiving increasing attention owing to its low energy consumption [10]. In OD, pure water is transported from the FS to the DS because of the osmotic pressure gradient, and hence, diluted DS can be obtained without external energy. Therefore, OD (for example, in emergent devices, medical products, and direct fertigation) can eliminate one of the major issues (i.e., DS recovery) that impede the commercialization of the full-scale FO process. Furthermore, the absence of an external DS supply or a re-concentration process for the diluted DS could be another effective solution to facilitate the real-world commercialization [11].

The third case concerns osmotic concentration (OC) to dewater or concentrate the FS, including food and beverages, food additives, and bioproducts, as presented in Fig. 3b-2 [12,13]. Compared with conventional evaporative concentration techniques, FO is advantageous for maintaining the physical properties (e.g., color, taste, aroma, and nutrition) of the products without deteriorating their quality. The fourth case regards simultaneous FS concentration and DS dilution, as shown in Fig. 3b-3. The representative technology involves an indirect desalination process coupled with RO for simultaneous wastewater treatment and desalination [14,15]. In this hybrid system, FO concentrates wastewater and dilutes seawater, and then diluted seawater is desalinated by RO with a reduced energy consumption. Among publications on these four cases, a fraction reports on the application of FO hybrid systems (Case 1) in various industries. However, most of the performance data have been collected from laboratory-scale testing; therefore, there is still a lack of comprehensive data from pilot-scale tests or real applications to facilitate the commercialization.

Despite the recent advancements in various FO processes, only a few

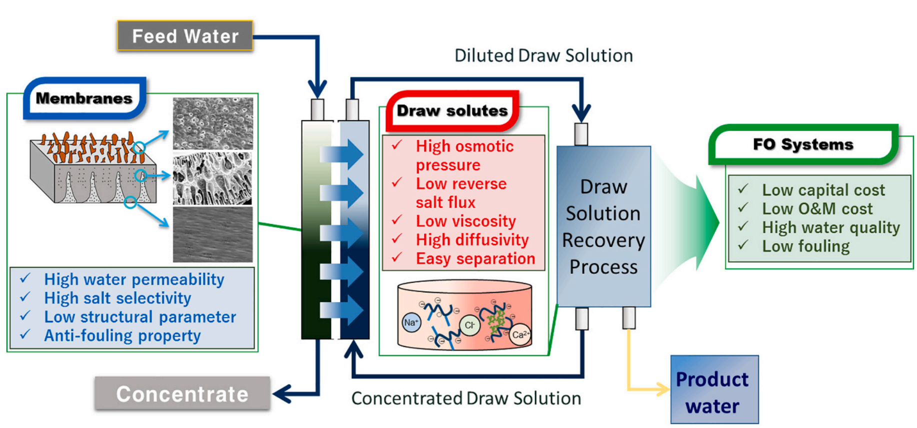

Fig. 1. Three major components of forward osmosis (FO).

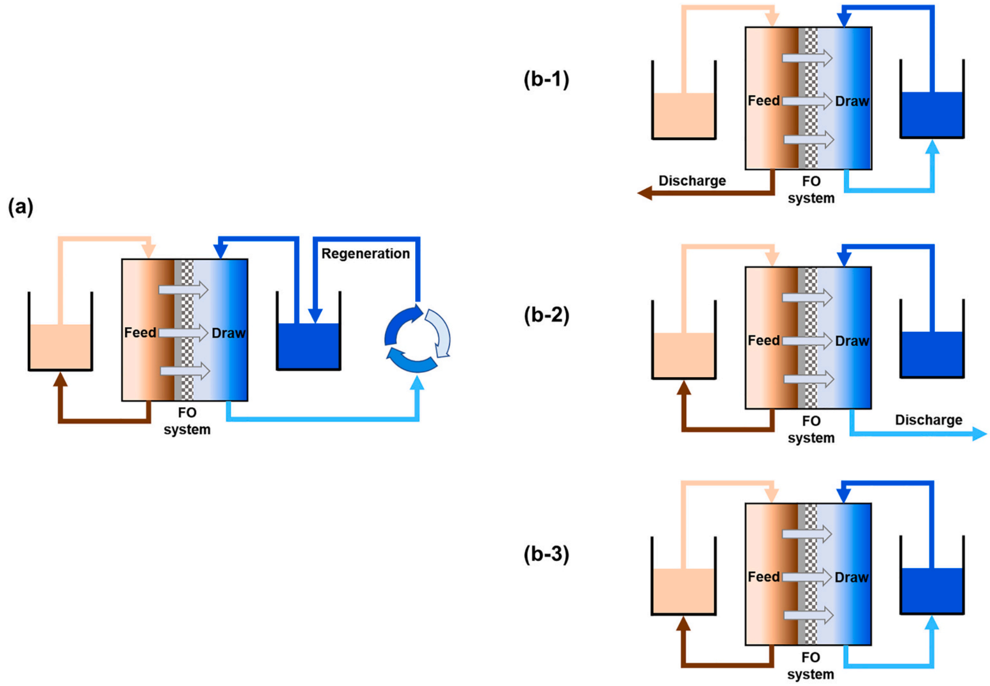

Fig. 3. Classification of FO applications based on fundamental working mechanisms of FO systems. (a) Conventional FO system with DS regeneration, and (b-1) osmotic dilution; (b-2) osmotic concentration; (b-3) simultaneous osmotic dilution and concentration, without DS regeneration.

studies have succeeded in the commercialization of their approaches. This is because the need for DS recovery can still be a significant obstacle to economic feasibility owing to an additional process [16]. Nevertheless, the development of better draw agents is still in progress; hence, the practical application of FO-integrated systems as filtration systems in the future is anticipated. To ensure continuity in these research efforts, the inherent challenges (e.g., DS recovery) of FO systems must be overcome to realize economically viable applications.

Therefore, this review aims to explore the recent developments in FO systems without DS recovery and the potential to expand their applications for commercialization. First, the historical development of FO in terms of FO membranes, DSs, and FO systems is reviewed. Based on the understanding of FO, current limitations in terms of DS recovery, energy issues, and a lack of pilot-scale evaluations to overcome the obstacles against commercialization are identified. From the discussion, it is found that FO systems without DS recovery have a high potential for commercialization. Therefore, FO systems are further classified into three configurations (i.e., OD, OC, and simultaneous osmotic dilution and concentration (SODC)), and their successful applications are discussed. Finally, the hype cycle of FO applications is analyzed based on the literature to address the strategies and identify future research directions of FO applications.

2. Where does FO process stand?

2.1. Historical development

The FO process was first introduced by Loeb et al., who proposed the application of osmosis for dewatering of brine from the Dead Sea in 1975 [17]. In early 2000, the development of a novel ammonia–carbon dioxide (NH_3-CO_2) DS for an FO-based desalination system resurrected academic and industrial interest as an alternative to the existing desalination technologies [18]. Since then, FO has been considered as a filtration system to remove undesired pollutants and produce clean water.

A cellulose triacetate (CTA) FO membrane was first commercialized in the 1990s by Hydration Technology Innovations (HTI) for commercial products (e.g., LifePack, Hydrowell, etc.). However, this membrane exhibited a low performance (i.e., low water flux and low salt rejection) due to the intrinsic properties of the active layer and the large structural parameter of the support layer [19]. To enhance the water permeability and salt selectivity, a high-performance thin-film composite (TFC) polyamide (PA) was first reported by Yip et al. in 2010 [20]. Subsequently, the TFC PA flat-sheet FO membrane and an FO spiral wound module were commercialized by HTI in 2012. Porifera introduced a novel plate-frame module using their TFC PA FO membranes in 2014. Aquaporin fabricated an aquaporin-incorporated TFC FO flat sheet membrane in 2015. To further reduce internal concentration polarization (ICP), many studies focused on developing ideal support layers using, for example, nanofibers for a low structural parameter [21–24]. Furthermore, a free-standing FO membrane with no support layer was also proposed for zero ICP [25–27].

A spiral wound module and a plate-frame module have a lower packing density compared to a hollow-fiber module. Therefore, many researchers have attempted to fabricate hollow-fiber FO membranes owing to the low hydraulic pressure of FO, similar to that of microfiltration (MF) and ultrafiltration (UF). Wang et al. examined a polybenzimidazole (PBI) hollow-fiber nanofiltration (NF) membrane for FO in 2007 [28]. To prevent fouling on the support layer, a double-skinned FO HF membrane was developed in 2012 [29]. Industrial companies have also developed hollow-fiber FO membranes. Toyobo succeeded in fabricating CTA hollow-fiber FO membranes in 2014, and Aquaporin fabricated an aquaporin-incorporated TFC hollow-fiber FO membrane in 2017.

Several studies incorporated nanomaterials in FO membranes to achieve a high performance as well as specific properties such as antifouling. Fan et al. fabricated a TFC FO membrane with a support layer incorporated with carbon nanotubes (CNTs) to reduce ICP and provide anti-fouling properties via electroconductivity [30]. The incorporation of hydrophilic nanomaterials in the substrate of a thin-film composite forward osmosis membrane will increase membrane porosity, hydrophilicity and decreases the tortuosity of the support layer, that collectively alleviate the effect of ICP [31]. In the future, FO membranes will be further developed for anti-fouling and hydrophilic properties using novel nanomaterials or polymers.

In FO, DS is a key component for extracting pure water from FS. The ideal draw solute should satisfy several important criteria, including (i) high osmotic pressure, (ii) low reverse solute flux (RSF), and (iii) easy and cost-effective recovery from the diluted DS [8,32]. Therefore, several draw solutes have been examined for FO applications. In 2005, McCutcheon et al. suggested ammonia-carbon dioxide (NH_3-CO_2) as a draw solute for FO desalination owing to its easy separation [18]. Ling et al. synthesized highly water-soluble magnetic nanoparticles (MNPs) for FO in 2010 because MNPs can be readily extracted from diluted DS [33]. In 2011, Phuntsho et al. evaluated fertilizer candidates as DSs to supply di luted fertilizer solution for irrigation [10]. Li et al. tested ionic polymer hydrogels for FO owing to their high water release rate under a combination of pressure and thermal stimuli in 2011 [34]. In 2013, Stone et al. used switchable-polarity solvents (i.e., mixtures of carbon dioxide, water, and tertiary amines) to use their polar and nonpolar phases for the separation of draw solutes from a diluted DS [35]. In 2014, Zhao et al. employed dendrimers (i.e., poly(amidoamine) terminated with sodium carboxylate groups) for seawater desalination because of their high osmotic pressure, low viscosity, and large molecular size [36]. In the same year, Guo et al. investigated Na + −functionalized quantum dots as draw solutes because of their unique properties (i.e., ultrasmall size and rich ionic species) [37]. Kim et al. synthesized and examined thermo-responsive copolymers containing ionic groups in the same year by adjusting the ratio of [2-(methacryloyloxy)ethyl]trimethylammonium chloride and 2-(2-methoxyethoxy)ethyl methacrylate as monomers for FO [38].

In addition to the development of FO membranes and DSs, several FO systems have been proposed, as shown in Fig. 3. The concept of FO desalination was first patented in 1965 [39]. Sidney et al. first proposed the concept of FO for desalting water from the Dead Sea water 1975 [17]. However, this concept required an additional process for the production of pure water and regeneration of the DS. Thus, FO was coupled with distillation column to recover volatile draw solutes (e.g., NH_3-CO_2) [9,18]. McGinnis et al. proposed an NH_3-CO_2 FO membrane brine concentrator [40]. In 2009, Achilli et al. proposed an osmotic membrane bioreactor (i.e., a bioreactor coupled with an FO membrane) for wastewater treatment owing to the low fouling property of FO [41]. For the effective reconcentration of inorganic DS, RO was integrated because of its high rejection rate [42]. However, RO has a high energy requirement for DS regeneration; hence, NF and UF were proposed as a DS recovery technology when using divalent ions and macromolecules as the DS, respectively [43,44]. Thermal desalination technologies (i.e., membrane distillation (MD) and multi-stage flash distillation (MSF)) have also been utilized for the recovery of inorganic DSs [45,46].

OD was first proposed in 1975. It does not require DS regeneration and uses the dilution phenomenon of the DS during FO operation. Therefore, an osmotic hydration bag (i.e., the first commercial product using OD) was developed to make a sugar drink for an emergency by extracting pure water from the surrounding water body. An osmotic pump was developed in 1982 to deliver drugs in its surroundings continuously [47]. After 30 years, Phuntsho et al. proposed an FO system for fertigation, which uses diluted fertilizer DS for irrigation [10,48]. This concept was adapted in 2015 to dilute dialysate concentrate for kidney dialysis from impaired water sources [49].

Researchers have proposed the opposite concept to OD (i.e., OC). Because FO uses the osmotic pressure gradient as a driving force, it can concentrate a high-TDS solution. Therefore, FO is also suitable for the food and beverage industries. Garcia-Castello et al. concentrated sucrose by a factor of 5.7 using FO [50]. Esperanza et al. examined the dewatering of orange peel press liquor and increased its concentration 3.7 times using 4 M NaCl as the DS [51]. The application of this concept was expanded to high-salinity brine treatment. Bartholomew et al. proposed an osmotically assisted RO (OARO) process for high-salinity brine treatment in 2017 [52]. This system fills up the permeate channel with high-salinity water, such as RO brine, to concentrate RO brine with low hydraulic pressure.

To utilize both OD and OC, FO was first proposed as a pretreatment for RO desalination in 2005 [53]. FO extracts pure water from the water source with high fouling potential using DS, and then RO produces clean water from the diluted DS with reduced energy consumption. The OD of highly saline water that is regenerated by RO or direct discharge into the sea provides another method to reduce the energy demand and environmental impact during brine discharge using a co-located source of impaired water. A secondary goal may be the volume-minimizing treatment of the impaired water stream [14]. With regard to wastewater reclamation, waste solutions needing either concentration or dilution are generated in several industrial fields (e.g., municipal wastewater, manufacturing industry, produced water, and biorefinery). Waste solutions having a high potential for recycling are usually treated separately for resource recycling. Therefore, from an economic and technical point of view, FO can be applied to industrial applications as a potential wastewater reclamation technology. To this end, the osmotic gradient that is established by two different waste streams generated from the facilities can be used as the driving force [11,54–61] (Fig. 4).

In addition to academic research, the industrial development of FO systems is important because it is directly related to FO commercialization. FO industrial companies can be classified into three groups: (i) FO membrane manufacturers, (ii) FO system manufacturers, and (iii) both FO membrane and FO system manufacturers. Detailed information

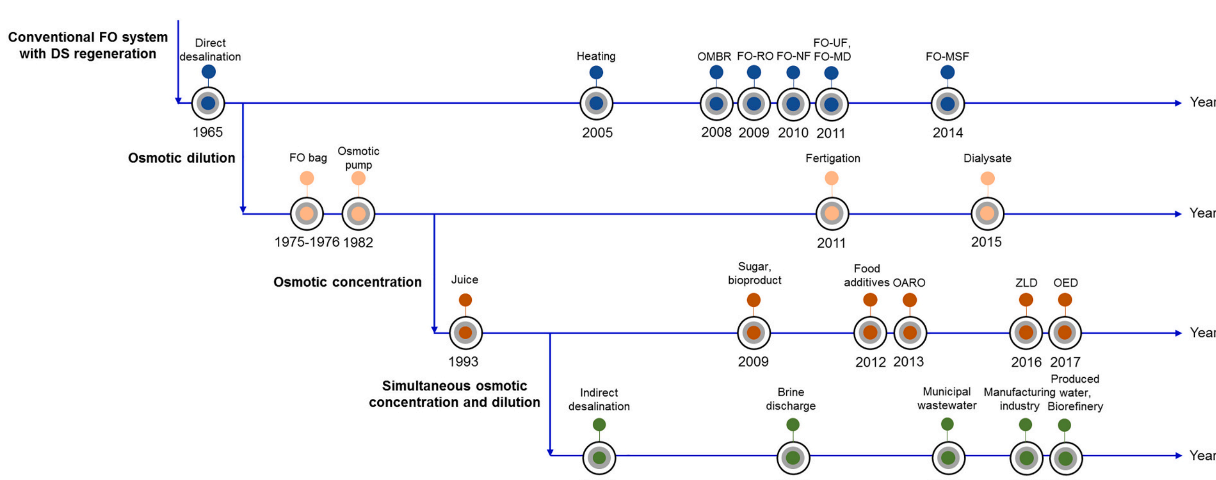

Fig. 4. Historical development schematic diagram of FO industrial applications.

on FO companies is summarized in Appendix 1. The representative FO membrane manufacturer is the HTI company, founded in 1986. HTI first developed commercial CTA FO membranes for the production of tomato paste. Similarly, FTSH2O, which was founded in 1989, has been manufacturing both FO membranes and FO systems. Two types of CTA FO membranes are used: (i) an FO industrial membrane and (ii) an FO sanitary membrane. FO industrial membranes can be applied for very dirty, high-TSS and -TDS wastewaters without extensive pretreatment, such as landfill leachate, because of the less cleaning frequency. Oasys Water, which was founded in 2008, offers FO membranes and unique DS and FO systems. The company commercialized TFC PA FO membranes in 2010 [62]. It introduced two FO systems: (i) ClearFlo Membrane Brine Concentrator (MBC)™ and (ii) ClearFlo Complete. It also developed unique thermolytic salts (ammonia–carbon dioxide salts) as DSs for FO systems [40]. Porifera has been providing a TFC PA flat-sheet FO membrane and a unique plate-frame FO module and FO systems for product concentration and wastewater treatment for zero-liquid discharge (ZLD) applications since 2009. In particular, the module exhibits high efficiency and low head loss owing to its unique module design, enabling it to achieve high surface velocity across the entire membrane area.

These companies manufacture both FO membranes and FO systems. Few manufacturers produce only FO membranes and modules. Toyobo successfully fabricated a CTA hollow-fiber FO membrane and supplied its membranes to a Danish osmotic power plant in 2018. Aquaporin prepared biomimetic hollow-fiber FO membranes and modules by incorporating aquaporin proteins with a TFC PA FO membrane. Trevi Systems developed FO hybrid systems using unique draw chemistry to couple FO with a coalesce. The company conducted many case studies, such as the FO pilot system at Abu Dhabi, UAE in 2013, a 10-m^3/day pilot system at Kuwait City, Kuwait, and a 500-m^3/day FO plant at Kailua-Kona, Hawaii, USA in 2020.

In addition to companies mentioned above, several companies produce medical devices (osmotic pumps) developed based on the principle of the osmosis. ALZA (currently Durect) first developed osmotic pumps in 1970's for internal uses and then launched their first commercial products for an animal research in 1977. Currently, they have eleven models with reservoir volumes (i.e., 100 μL, 200 μL and 2 mL) and medicine release durations ranging from 1 day to 6 weeks. RWD also produces eight osmotic pump models with reservoir volumes (i.e., 100 μL, 200 μL and 2 mL) and medicine release durations ranging from 3 days to 4 weeks.

2.2. Current limitations

2.2.1. Draw solution performance and recovery

As discussed above, the selection of appropriate DSs is crucial in an FO process because the properties of the DS help determine the FO performance (i.e., in terms of water flux and RSF) and the method for DS regeneration. For high water flux, DS should have a high osmotic pressure because its driving force is the effective osmotic pressure gradient between the FS and DS [8]. Therefore, there have been many studies on

Table 1
Overview of DSs and recovery methods in FO.

Recovery methods	Process	Typical DSs	Remarks	Ref.
Thermally driven process	Distillation	NH_3 and CO_2, switchable polarity solvents (SPS), etc.	Energy-intensive	[18,35]
	MD	2–methylimidazole based solutes, organic ionic salts, etc.	High energy cost unless using waste heat	[63,65,80]
Pressure-driven process	RO/NF/UF	Organic acids and inorganic salts, polyglycol copolymers, polyelectrolytes, etc.	Low water recovery and poor solute rejection	[8,43,44,64,76,81]
Precipitation method	Chemical reaction	CuSO, Al_2SO_4	Not economical and feasible	[82,83]
Stimuli-responsive method	Changes in the magnetic field, temperature, light, and pressure	Magnetic nanoparticles, hydrogels, etc.	Complex and ineffective	[33,34,84,85]

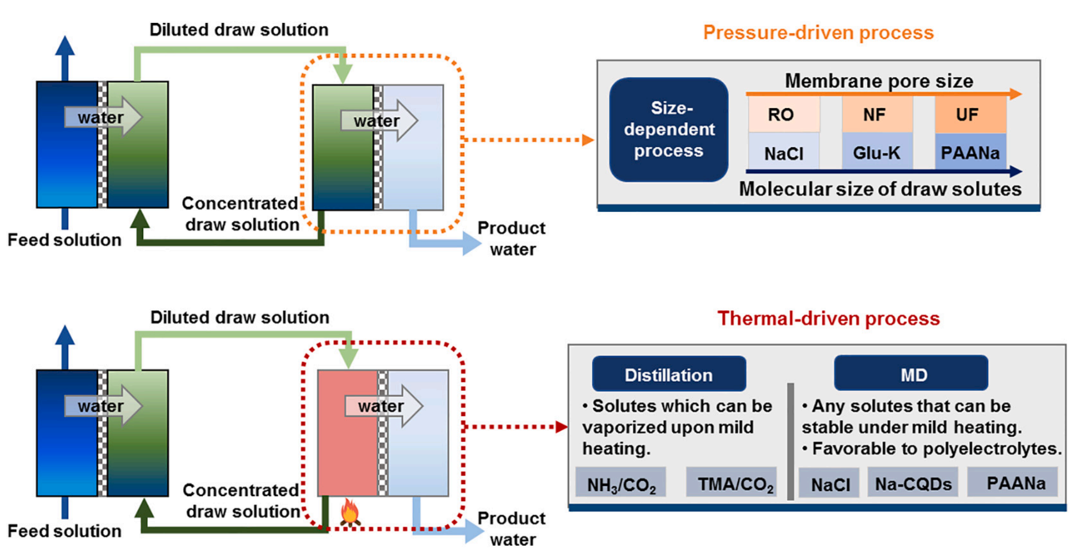

Fig. 5. Schematic diagram of a system combining FO and pressure-driven membrane processes (including RO, NF, UF, and MF) and thermal-driven process (including distillation and MD) [73].

developing novel DSs with a high osmotic pressure to enhance water flux in FO [63]. Moreover, RSF is a key component in FO because a low RSF is favorable for stable operation and economic feasibility [6,32]. Thus, a DS should have a high water flux and negligible RSF. Based on this consideration, various DSs (e.g., $HMTA^{+}$-SO_3^{-}, phytic acid salt, P(SSA-co-MA)-Na, oxalic acid complexes, ferric-lactate complex, and organic phosphonate salts) have been developed [64–72].

However, an important issue with DS is that it must be recovered from the diluted DS. In recent years, various novel DSs have been proposed in combination with various remarkable recovery technologies to advance FO technology for various applications, as summarized in Table 1. Pressure-driven membrane processes such as RO, NF, and UF have been widely investigated as representative technologies for the reconcentration of diluted ionic or macromolecular DS [73]. As shown in Fig. 5, RO was first proposed as a post-treatment process for draw solute recovery among the different membrane systems because of its high water recovery rate as well as high rejection rate of various salts. However, it requires a high hydraulic pressure, typically 35–100 bar, and thus, the operating costs are still high [14,74,75]. NF and UF were therefore suggested for the recovery of DSs because they require less energy input than those in RO and other thermally driven processes [76–79]. Although the experiments were mostly carried out at a much lower concentration than the actual DSs used in FO, UF and NF processes still suffered from the low rejection rate of the draw solutes during regeneration [76,78].

Thermal separation is mostly used for volatile compounds and dissolved gas-based DSs. Other alternatives for thermal recovery, such as distillation, MD, and thermosensitive materials, are categorized as membrane processes and stimuli-responsive separation [63,65,80,86,87]. With only a temperature difference of 10–20 °C between the feed and permeate solutions, the separation of draw solutes from water is theoretically achievable; thus, high cost-effectiveness can be obtained from the utilization of low-grade heat such as industrial waste heat [18,35,45,88–90]. To overcome the limitations of classical DSs, synthetic materials such as stimulus-responsive polymers, nanogels, polyelectrolytes, hydrogels, and MNPs, have been developed [33,34,84,85,91]. The requirements for selecting an appropriate draw solution are subject to an FO application's purpose; thus, the priority

Table 2
Summary of previous energy consumption analysis in FO membrane process.

Application	Feed solution	Draw solution	FO/PRO SEC (kWh/m^3)	Total SEC (kWh/m^3)	Comparison to conventional technologies	Evaluation method	Year	Ref.
Conventional direct desalination	Seawater	NaCl	N.A.	FO-RO: 4.9	RO: 8.5	Pilot-scale	2011	[98]
	Synthetic seawater	$MgSO_4$	N.A.	FO-RO: 2.93	RO: 3.79	Modeling	2012	[99]
	Synthetic seawater	NaCl	0.1	FO-RO: 3.58	RO: 3	Modeling	2014	[16]
	Synthetic seawater	NaCl	0.09–0.18	FO-RO: 6–10	RO: 4–5.5	Modeling	2014	[94]
	Seawater	Thermo-copolymer	N.A.	FO-thermal: 35–40	MED: 62	Pilot-scale	2019	[100]
Indirect seawater desalination	Secondary effluent	Seawater	N.A.	FO-RO: 1.5	RO: 2.5–4	Modeling	2011	[75]
	Synthetic wastewater	Synthetic seawater	N.A.	FO-RO: 1.4–2.1	RO: 2.4–3.9	Modeling	2015	[101]
	Impaired water	Synthetic seawater	N.A.	FO-RO: 2.5–2.8	RO: 3.7	Modeling	2015	[102]
	Impaired water	Synthetic seawater	N.A.	FO-RO: 9–9.8	RO: 6.3–14	Bench-scale	2015	[103]
			N.A.	FO-RO: 4.37–4.8	RO: 2.5–6.2	Modeling		
	Treated wastewater	Synthetic seawater	N.A.	FO-RO: 2.26	RO: 2.92	Modeling	2016	[104]
	Wastewater effluent	Synthetic seawater	N.A.	FO-RO: 1.86–2.41	N.A.	Modeling	2016	[105]
	Secondary effluent	Seawater	0.55	FO-RO: 2.85	RO: 3.34	Pilot-scale	2017	[7]
	Synthetic wastewater	Seawater	0.2	FO-RO: 1.6	RO: 3.9	Modeling	2018	[15]
	Municipal wastewater	Seawater	N.A.	FO-RO: 1.17–1.46	RO: 1.95–2.28	Modeling	2018	[106]
	Wastewater	Seawater	N.A.	FO-RO: 1.37–1.82	RO: 2.13–4.05	Modeling	2019	[107]
Brine concentration	Produced water	NH_3/CO_2	N.A.	FO-distillation: 21	MVC: 28–39	Pilot-scale	2013	[40]
	Synthetic brine	Sweep	N.A.	OARO: 2.9–19.3	MVC: 10–24	Modeling	2017	[52]
	Synthetic brine	Sweep	N.A.	COMRO: 3.16	RO: 3.79	Modeling	2018	[108]
Brine discharge	Synthetic wastewater	RO brine	0.4–0.6	PRO-RO: 1.2	RO: 2	Modeling	2014	[95]
	Synthetic wastewater	RO brine	N.A.	PRO-RO: 1.08–1.14	RO: 1.79–2.27	Modeling	2016	[109]
	Algae	Brine	0.13	UF-FO: 0.36	UF: 0.47	Bench-scale	2020	[112]
	Municipal wastewater	RO brine	N.A.	PRO-RO: 0.98–1.55	RO: 1.95–2.28	Modeling	2018	[106]
Wastewater reclamation	Synthetic wastewater	NaCl	N.A.	FO-RO: 15–30	Bioreactor: 50–370	Modeling	2005	[53]
	Wastewater effluent	Sodium Polyacrylate, $MgCl_2$, $MgSO_4$	0.1	FO-NF: 2.23–2.51	RO: 1.66	Pilot-scale	2018	[96]
Concentration of liquid foods	Acid whey	Potassium lactate	0.32	FO-evaporator: 0.6	RO: 0.25	Modeling	2019	[97]
	Skim milk	NaCl	5–10	FO: 5–10	RO: 9–20	Pilot-scale	2019	[111]

order of evaluation criteria is obviously different for each application area. However, the practical applications of these methods in continuous FO processes have not been demonstrated.

Despite recent advances in effective DS regeneration, the requirement of additional processes may hamper commercial applications because of increases in both capital and operating expenses (i.e., CAPEX and OPEX) [92,93]. Even though inexpensive/free energy sources (e.g., waste heat and solar energy) can be secured for low OPEX, high CAPEX will be an obstacle adoption. Furthermore, there are no free energy sources in reality. Many studies considered low-grade heat as waste heat, but most industries utilize low-grade heat or sell it to other companies. Moreover, solar energy is not free energy because it can also be used to generate electricity or heat energy. Furthermore, the water recovered by sunlight was not in liquid form but mostly in a vapor state; thus, additional condensation to replace the evaporated water may be needed, which can increase energy consumption.

2.2.2. Energy consumption in real-scale applications

Energy consumption is a critical evaluation factor in pilot-scale operation and is closely related to economic feasibility. Based on the

Table 3
Overview of pilot-scale FO applications.

Application	Feed solution	Draw solution	Membrane configuration	Module	Capacity (m^3/d)	Operating period (day)	SEC (kWh/m^3)	Final water quality (mg/L)	Year	Ref.
Conventional direct desalination	Seawater	NaCl	Hollow-fiber (–)	Modern Water, Oman	100	365	4.9	TDS: < 200	2015	[98,116]
	Seawater	Thermo-copolymer	Hollow-fiber (336 m^2)	Trevi Systems Inc., USA	10	30	35–40	TDS: 143	2019	[100]
Agricultural fertigation	Brackish groundwater	$(NH_4)_2SO_4$ fertilizer	Spiral-wound CTA 8040 (9 m^2)	HTI, USA	N.A.	3	N.A.	Nitrogen: 831	2015	[113]
	Saline coalmine water	$(NH_4)_2SO_4$ fertilizer	Spiral-wound CTA 8040 (20.2 m^2)	HTI, USA	15	180	N.A.	Ammonia: 75	2016	[115]
	Synthetic wastewater	Commercial liquid fertilizer	Spiral-wound TFC 8040 (15.3 m^2)	Toray Industries, Korea	72	N.A.	N.A.	Ammonia: 17.5	2017	[114]
Indirect seawater desalination	Tertiary effluent	Synthetic seawater	Flat-sheet CTA (0.266 m^2)	HTI, USA	N.A.	5	N.A.	Ammonia: 0.3	2010	[14]
	MBR effluent	Synthetic sea salt	Spiral-wound CTA 4040 (1.58 m^2)	HTI, USA	4.4	55	N.A.	Nitrate: < 1.57	2013	[117]
	Brackish surface water	Synthetic seawater	Spiral-wound TFC 2521-MS-P-3H (–)	HTI, USA	1.8	7	N.A.	N.A.	2016	[118]
	Oil and gas wastewater	NaCl	Hollow-fiber CTA (31.5 m^2)	Toyobo, Japan	1.9	2	N.A.	N.A.	2020	[119]
	Synthetic wastewater	Synthetic seawater	Hollow-fiber CTA (31.5 m^2)	Toyobo, Japan	1.4	2	N.A.	N.A.	2020	[120]
	Secondary effluent	Seawater	Flat-sheet TFC PFO-100 (7 m^2)	Porifera, USA	21.8	150	2.85	Ammonia: 0.01	2017	[7]
Wastewater reclamation	Domestic sewage	NaCl/$MgSO_4$	Flat-sheet CTA (0.1 m^2)	N.A.	13	8.3	N.A.	Nitrate: 3.36–6.8	2010	[121]
	Domestic wastewater	NaCl	Flat-sheet CTA (1.2 m^2)	HTI, USA	N.A.	249	N.A.	Nitrogen: 6.3	2015	[122]
	Wastewater effluent	Sodium Polyacrylate, $MgCl_2$, $MgSO_4$	Flat-sheet TFC PFO-100 (84 m^2)	Porifera, USA	4.3	480	2.23–2.51	Conductivity (μS/cm): 853	2018	[96]
	Synthetic wastewater	NaCl	Flat-sheet TFC (0.05 m^2)	Toray Industries, Korea Porifera, USA	N.A.	16	N.A.	TrOCs rejection: >90%	2018	[123]
Brine concentration	RO brine	NaCl	Flat-sheet CTA (–)	HTI, USA	N.A.	4.5	N.A.	water recovery: 98%	2009	[124]
	Produced water	NaCl	Spiral-wound CTA	HTI, USA	926	N.A.	N.A.	TSS: 22	2010	[125]
	Produced water	NH_3/CO_2	Spiral-wound TFC (59.4 m^2)	Oasys Water, Inc., USA	21.8	4.1	21	TDS: < 300	2013	[40]
	Produced water	NaCl	Spiral-wound CTA (–)	HTI, USA	N.A.	7	N.A.	N.A.	2014	[126]
	Produced water	NaCl	Spiral-wound CTA (4.9 m^2)	FTS-H_2O, USA	8.6	28	N.A.	DOC: 2.4	2018	[127]
Brine discharge	Tertiary effluent	Brine	Flat-sheet CTA (0.266 m^2)	HTI, USA	N.A.	4	N.A.	Ammonia: 0.3	2010	[14]
Concentration of liquid foods	Skim milk	NaCl	Spiral-wound CTA 8040–45-SDS (12 m^2)	FTS-H_2O, USA	N.A.	N.A.	5–10	N.A.	2019	[111]
	Whey	NaCl	Spiral-wound CTA 8040–45-SDS (24 m^2)	FTS-H_2O, USA	N.A.	N.A.	N.A.	N.A.	2020	[128]

applications of different feed and draw solutions, extensive investigations of previous FO energy consumption have been conducted. An overview of the studies concerning conventional direct desalination, indirect seawater desalination, brine concentration and discharge, wastewater reclamation, and concentration of liquid foods is presented in Table 2. Most specific energy consumption (SEC) values of FO systems are typically modeled by calculating the assumed pressure differences, water recovery, and ideal pump efficiency. Most of these studies reported energy consumption values inferring that FO is indeed a low-energy separation process. The fact that water in FO permeates spontaneously through a semi-permeable membrane does not mean that FO is more energy efficient as a separation process than other membrane processes [81]. Numerical modeling exhibited SEC values of 0.1–0.6 kWh/m^3 [7,15,16,94–97]. However, a crucial error often made in most of the studies is the fact that the energy used in FO was assumed to be almost 0 kWh/m^3, and those modeling SEC results were all obtained by given assumptions; as a result, the energy in FO was assumed to be negligible [40,52,53,75,98–110].

A comparison of overall energy consumption between the FO and RO processes reveals that conventional FO direct desalination coupled with synthetic DS regeneration by RO introduces extra capital and operating costs with much higher energy consumption compared to those in RO processes [16,94,98,99]. A pilot-scale FO-NF study demonstrated that FO is a low-fouling technology that can achieve a stable, high-quality permeate for wastewater reuse in agriculture in the long term, and the total energy consumption is approximately 40% higher than that in the UF-RO process [96]. Recent research on the nonthermal concentration of liquid foods revealed that finding an economical solution for DS regeneration is key to reducing energy consumption in FO [97,111].

In addition, owing to the SODC working principle, FO processes have been proposed to have low energy consumption for indirect seawater desalination, brine concentration, and discharge prior to seawater RO desalination systems or other technologies [7,15,40,52,75,95,101–109,112]. Preliminary studies demonstrated that FO/PRO-RO hybrid processes can have positive economic benefits compared to stand-alone SWRO. This is because the hybrid process has a lower fouling tendency as estimated from laboratory or pilot-scale testing, which leads to lower costs in terms of energy (OD) and maintenance. The FO/PRO-RO hybrid process is advantageous compared with two independent and established water treatment streams (i.e., water reuse and/or desalination) or simply mixing these streams before treatment.

2.2.3. Performance assessment of pilot-scale FO studies

To address the limitations in the full realization of FO, more pilot-scale evaluations of FO studies, or even full-scale trials, must be conducted on site in the long term. Although FO pilot studies are important, the operating data from such studies are either limited or are not yet open to the public. Various pilot-scale FO applications have been investigated, as summarized in Table 3. Most of pilot-scale FO research has focused on the treatment of wastewater using synthetic seawater or RO brine (NaCl or other inorganic salts), including municipal wastewater and high-salinity produced water. These studies have emphasized the advantages of the FO process in treating highly impaired water without substantial irreversible membrane fouling. In addition, higher concentration or dilution results were obtained for agricultural fertigation and concentration of liquid foods; however, quality evaluations of the final products are unavailable or not yet open to the public [111,113–115].

Most pilot research works listed in Table 3 typically adopted CTA FO membranes although TFC FO membranes outperform CTA FO membranes. Only limited pilot-level data on TFC FO membranes have been reported in the literature. Moreover, these FO pilot tests were executed over relatively short periods (1–480 days) with low operating capacity (1.8–100 m^3/d); thus, there is a high demand for long-term operating data. Furthermore, FO energy use, total SEC value, CAPEX, and OPEX cost were mostly evaluated using modeling methods. Real-scale, long-term operation of FO systems is highly needed. Further verification of these data is needed because of the lack of critical operating information, such as limited produced water flux and low water recovery. In conclusion, the concepts of both concentration and OD systems have not been realized at the pilot- and full-scale levels, although many attempts are currently being implemented.

3. Commercial application of forward osmosis membrane

3.1. Osmotic dilution (OD)

OD is used to dilute highly concentrated DS via FO with natural extraction of water from impure water. Therefore, several applications have been developed based on the concept of OD, as summarized in Appendix 2. An FO bag (FOB) is an emergent device for the production of energy drinking water from human urine and wastewater [129–134]. Fertilizer-drawn forward osmosis (FDFO) is used for the production of diluted fertilizer solutions for fertigation [10]. Furthermore, an osmotic pump and the OD of dialysate concentrate have been proposed for medical applications. An osmotic pump is a device used for continuous drug delivery in its surroundings [135]. Dialysate concentrate is diluted using OD from various water sources (e.g., impaired water sources and tap water) and used for dialysis of the kidneys [49,136–139]. Among these applications, osmotic pumps and FDFO have been considered the most feasible applications of OD; hence, we will focus on these applications in this section.

3.1.1. Osmotic pump

Osmotic pumps can also be considered for the successful application of OD in the pharmaceutical industry. For instance, Neely and Hake used an osmotic pump to maintain tritiated thymidine exposure to cells in culture [47]. Stringer et al. employed osmotic pumps for the sustained release of 1-aminobenzotriazole and the inhibition of cytochrome P450 enzymes in mice [140]. Osmotic pumps, also called osmotic drug delivery systems, consist of three components (i.e., osmotic agent, solvent, and drug) [135]. Depending on the design of the three components, osmotic pumps are classified into three types: (i) single-compartment intracorporeal pumps, (ii) two-compartment intracorporeal pumps, and (iii) multicompartment extracorporeal pumps. A single-compartment pump has two components (i.e., drugs and a semipermeable membrane), as shown in Fig. 6a. In this system, the drugs act as osmotic agents and absorb water from the surrounding body fluid. The pressure inside the pump increases, and thus, the drugs escape through an outlet. A two-compartment pump is composed of four components (i.e., drugs, osmotic agents, a movable barrier, and a semipermeable membrane), as shown in Fig. 6b. When exposed to body fluid, the osmotic agents are activated and absorb water from the body fluid. The pressure of the osmotic agent increases, which pushes the movable barrier to the drug chamber. As a result, the drugs are delivered into the body fluid. A multicompartment pump has three chambers, one each for water, osmotic agents, and drugs. A semipermeable membrane is placed between chambers for the water and osmotic agents, and a movable barrier is installed between the chambers for the osmotic agents and drugs. Once the osmotic agents absorb water through the semipermeable membrane, the movable barrier starts to squeeze the drugs, which are delivered to the surroundings of the osmotic pump.

Rose and Nelson patented the first osmotic pump for the delivery of drugs to the gut of sheep and cattle [141,142]. They used capsule-based osmotic pumps consisting of three chambers: a drug chamber, a salt chamber containing excess solid salt, and a water chamber [143]. The drug and water chambers were separated by a semipermeable membrane. The Higuchi–Leeper pump, which was the first simplified design of the Rose-Nelson pump, was developed by the Alza Corporation in the early 1970s [144,145]. The Higuchi-Leeper pump has no water chamber, and the device is activated by water absorbed from the surrounding environment. In the early 1970s, Higuchi and Theeuwes developed a simpler version of the Rose-Nelson pump [146]. This device is similar to

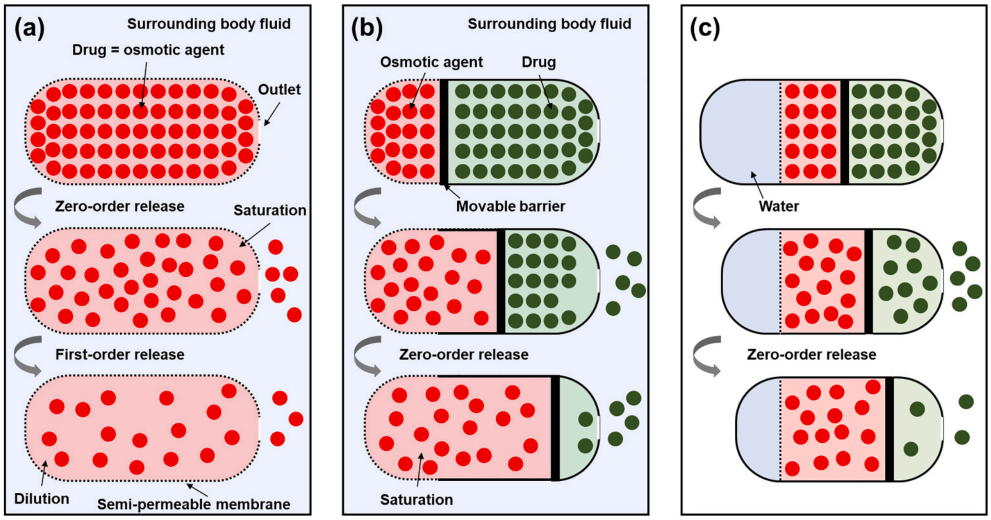

Fig. 6. Illustrative principle of (a) an intracorporeal single compartment pump, (b) an intracorporeal two-compartment pump, and (c) an extracorporeal multi-compartment pump [135].

the Higuchi-Leeper pump, but the rigid housing is replaced with a membrane that is strong enough to withstand the pumping pressure developed inside the device. In 1991, Wong et al. proposed a liquid osmotic pump (similar to the Rose-Nelson pump) to deliver liquids, lipid–lipid emulsions, and solid dispersions [147].

The elementary osmotic pump, which is the first unitary-core osmotic pump, was developed by Theeuwes in 1974 [148]. This device is a further simplified form of the Higuchi–Theeuwes pump. It does not have a separate salt chamber, and the drug is used as the osmotic agent. A controlled-porosity osmotic pump was patented in 1980 to decrease the risk of extremely localized drug-induced irritation at the site close to the orifice of the pump [149]. A multilayer-core osmotic pump was also proposed. A push-pull osmotic pump consisting of a push layer and a drug layer was first developed by Cortese and Theeuwes in 1982 [150]. In 1988, a push-stick osmotic pump consisting of three layers (i.e., a push layer and two drug compartments) was proposed by Ayer et al. to prolong the therapy period [151].

In addition to patents, there have been many publications on the development of osmotic pumps. Theeuwes and Yum designed and tested osmotic pumps in 1976 for the delivery of semi-solid or liquid drug formulations [152]. Liu prepared an osmotic pump system by coating the core tablet with an indentation for the delivery of prazosin hydrochloride; thus, there was no need for laser drilling [153]. Yang et al. manufactured a monolithic osmotic pump tablet for traditional Chinese

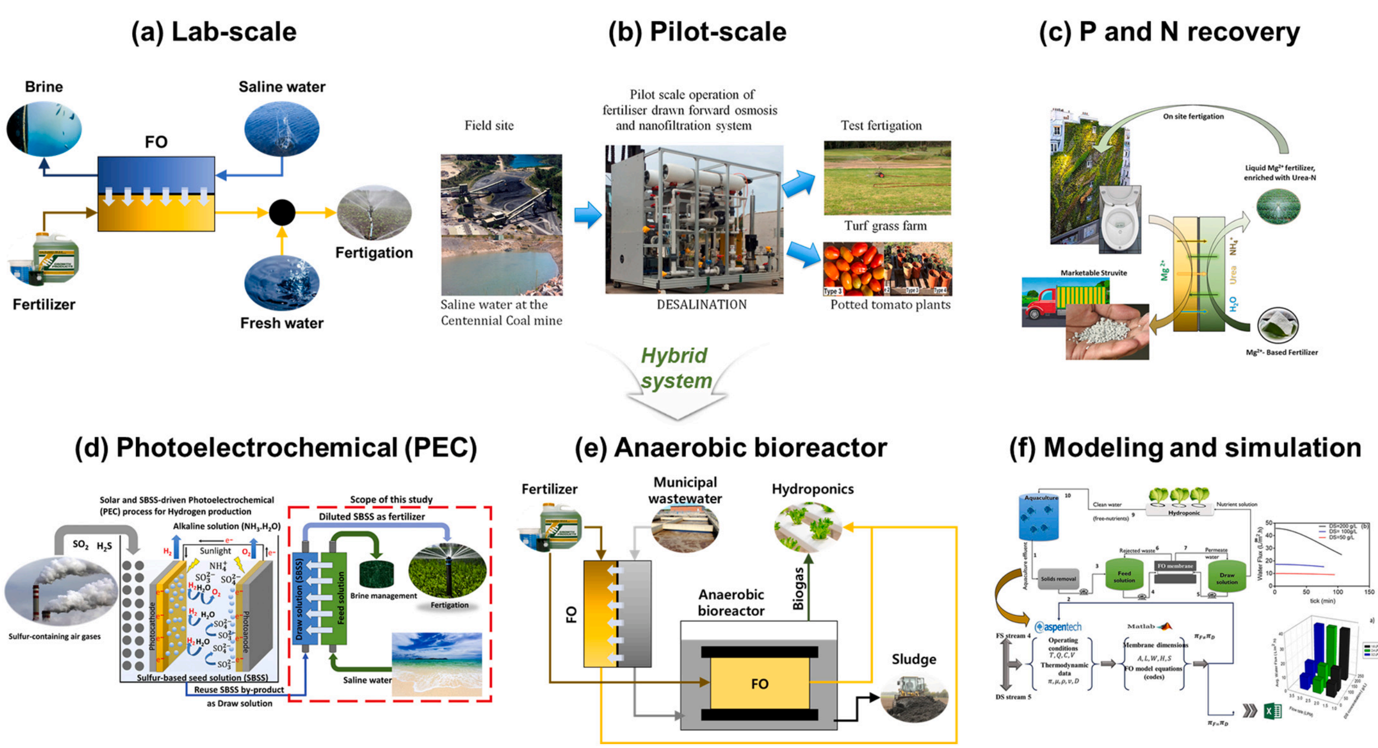

Fig. 7. Development stage of fertilizer-drawn forward osmosis (FDFO). (a) Conceptual illustration of a laboratory-scale FDFO [10], (b) pilot-scale FDFO system [115], (c) FDFO for nutrient recovery [172], (d) FDFO combined with photoelectrochemical process as pre-treatment [173], (e) anaerobic bioreactor and FDFO integrated system [32], and (f) modeling and simulation of FDFO [174].

medicine compound recipes [154]. Xin et al. evaluated time-released monolithic osmotic pump tablets containing diltiazem hydrochloride [155]. Tablets were coated with Kollidon® SR-polyethylene glycol mixtures via compression-coated technology rather than by spray-coating methods. Wang et al. evaluated a drug-resin complex (DRC) core incorporated with an elementary osmotic pump tablet to increase the lag time [156,157].

In addition, many researchers have utilized polymer swelling as an osmotic agent. Thombre et al. proposed a swellable-core technology to use osmotic pressure and polymer swelling to deliver drugs to the gastrointestinal tract in a reliable and reproducible manner [158]. Nokhodchi et al. designed a swellable elementary osmotic pump and optimized influencing factors such as the concentrations of the swelling, osmotic, and wetting agents, as well as the orifice size and membrane thickness [159]. Rabti et al. developed a swellable polymer osmotic pump tablet to deliver carbamazepine-containing solubility enhancers [160].

Controlled-porosity osmotic pumps have been extensively investigated to utilize the whole surface of the osmotic pump for drug delivery. Bi et al. developed a controlled-porosity osmotic pump system, which consisted of a tablet-in-tablet core and a controlled-porosity coating membrane, with biphasic release of theophylline for the nocturnal therapy of asthma [161]. Kumaravelrajan et al. designed controlled-porosity osmotic pump tablets by incorporating drugs in the core and coating with various types (PVP, PEG-400, and HPMC) and levels (30, 40, and 50% *w*/w of polymer) of pore former at weight gains of 8, 12, and 15% to deliver multiple drugs (i.e., nifedipine and metoprolol) at a prolonged rate up to 12 h [162]. Shahi et al. evaluated a controlled-porosity osmotic pump to deliver diltiazem hydrochloride [163]. Huang et al. developed a controlled-released osmotic pump capsule based on nimodipine-loaded self-microemulsifying drug delivery systems to improve the low oral bioavailability of NM [164].

3.1.2. Fertilizer-drawn forward osmosis (FDFO)

In 1976, Moody and Kessler first proposed a novel FO extractor for agricultural applications by using a fertilizer as a high-salinity DS to extract water from a brackish source solution [165]. Phuntsho et al. resurrected the concept of FDFO for direct fertigation from brackish groundwater in 2011, as shown in Fig. 7a [10,166]. In FDFO, a highly concentrated fertilizer solution is employed as the DS, and a low-salinity solution (e.g., brackish water or municipal wastewater) is used as the FS. A low-salinity solution is concentrated, and the fertilizer solution is diluted simultaneously. Diluted fertilizer solution can be supplied for irrigation after meeting the nutrient standard requirements for plant growth. To meet the water quality requirements for direct fertigation, several blended fertilizers were investigated [48]. However, reducing the final nutrient concentration for direct fertigation remains a challenge because of osmotic equilibrium, particularly when using high-salinity feedwater [167].

Therefore, Phuntsho et al. suggested NF as either a pre-treatment for a reduction in TDS of FS or a post-treatment for the partial recovery of draw solutes and the production of acceptable diluted fertilizer solution for direct fertigation [168]. Altaee et al. suggested that a dual-staged low-pressure RO is effective for concentrating the diluted fertilizer solution and producing a suitable fertilizer solution [169]. The concept of the FDFO and NF hybrid process was evaluated and demonstrated in a pilot-scale system for direct fertigation, as presented in Fig. 7b [115,170]. This system has lower environmental impacts (i.e., 21% less energy consumption and 14% lower OPEX cost) than UF-RO hybrid systems [92]. Kim et al. proposed a new scheme to couple RO with FDFO to achieve a higher recovery rate, lower energy, and more suitable fertilizer solution compared to a two-stage RO system [6]. To overcome osmotic equilibrium, hydraulic pressure was applied to FS, and the process is called pressure-assisted fertilizer-drawn forward osmosis (PAFDFO) [171]. This system further dilutes DS beyond the point of osmotic equilibrium and is more energy-efficient than simple FDFO [169].

Single or blended reagent-grade fertilizer chemicals may differ from commercial fertilizers, possibly leading to different performances. Commercial liquid fertilizers such as DS were investigated for the OD of wastewater for various applications, such as fertigation of green walls [175,176]. A high initial draw concentration leads to low energy consumption, whereas the primary effluent induces a high energy demand owing to a significant reduction in water flux by serious membrane fouling and enhanced external concentration polarization (ECP) [177]. Fertilizers containing less urea were more appropriate for FDFO because of the higher water flux and lower nutrient loss, as shown in Fig. 7c [178]. In addition to general fertilizers, novel fertilizers (e.g., sodium lignin sulfonate, sulfur-containing air pollutants (Fig. 7d), and molasses) have also been explored [173,179,180].

For the water–energy–food nexus, some researchers integrated FDFO with biological processes. Kim et al. proposed an FDFO and anaerobic bioreactor hybrid system, as presented in Fig. 7e, for wastewater treatment and a greenhouse hydroponic application consisting of two parts: (i) FDFO for concentration of municipal wastewater and dilution of fertilizer solution, and (ii) an anaerobic bioreactor coupled with FDFO for the treatment of concentrated municipal wastewater, production of biogas, and dilution of fertilizer solution [32,181,182]. However, the reverse diffusion of inorganic draw solutes has a negative impact on methane production [183]. Therefore, MF or UF were further installed for salinity build-up mitigation, but their effect was restricted [184,185]; thus, novel RSF mitigation method (e.g., surfactant-blended fertilizer DS) must be developed [186].

For hydroponic applications, Chekli et al. conducted pilot-scale experiments to obtain a diluted fertilizer solution [114]. They demonstrated that lettuce can be successfully grown with a fertilizer solution diluted by FDFO. They found that higher DS concentrations produce a higher water flux that is suitable for sustainable reuse, and increasing the recirculation flow rate leads to an increase in the energy consumption of FDFO for hydroponic applications [187]. They also simulated FDFO for hydroponics using the Aspen Plus-MATLAB model, as shown in Fig. 7f [174]. The type of feed solute (e.g., seawater or wastewater) impacted the external concentration polarization in the AL-FS mode, whereas concentrated DS promoted the ICP. The presence of organic micropollutants (OMPs) in fertilizer solution can affect the utilization of fertilizer solutions for fertigation because OMPs can have a negative impact on plants. Therefore, Kim et al. investigated the effect of fertilizer properties on OMP transport in FDFO [5,188]; however, the impact of OMPs in fertilizer solution on the growth of plants has not yet been investigated.

3.2. Osmotic concentration (OC)

The concentration of liquids is necessary in many industrial fields, such as the food industries and oil and gas industries, to either reduce the volume or increase the value in terms of nutrition or content. Several desalting technologies have been employed to realize liquid concentration. For example, pressure-driven membrane technologies such as NF and RO can be used to concentrate orange juice [189]. In addition, thermal technologies such as MSF and MD can be applied for brine concentration [190]. Although these technologies are highly effective in liquid concentration, they are limited due to huge energy consumption. FO has great potential for use in liquid concentration as a solution can be readily concentrated without high energy input because of the driving force of the osmotic gradient. Therefore, many researchers have focused on various FO applications including liquid foods (e.g., juice) and brine (e.g., produced water).

3.2.1. Concentration of liquid foods

The concentration of liquid food is one of the major unit operations in the food processing industry and improves product stability by reducing the water activity. It is performed before the product preparation process and involves processes as drying and reducing the weight and volume of liquid food, thereby reducing packaging, storage, and transportation costs [191]. Liquid food is generally concentrated by thermal-based processes. However, it is difficult to maintain the nutritional value of the food in such processes. Most nutrients are sensitive to heat; thus, exposing these components to high temperatures can cause a degradation of biologically active compounds [192]. With regard to the concentration of liquid foods, research has focused on pressure-driven membrane processes such as MF, UF, NF, and RO. However, there are many limitations to these membrane processes, such as the requirement of high pressure, limit to maximum attainable concentration, concentration polarization, and membrane fouling [193].

FO has attracted significant attention over the last decade for liquid food concentration because it can increase the value of liquid foods [13]. The key idea is to develop liquid food concentration systems in which the FO process can add value. Compared with conventional evaporative concentration techniques, FO is advantageous because it retains the physical properties of liquid foods (such as color, taste, aroma, and nutrition) without compromising their quality. Moreover, FO results in increased rejection and less membrane fouling compared to those of pressure-driven membrane processes such as RO. Therefore, diverse applications of the FO process have been studied. The results obtained in several studies on FO are summarized in Appendix 3.

FO has been used for the concentration and recovery of many products in the food and beverage industry (juice, sugar, food additives, etc.) [13,50,51,194–217] and bioproduct processing (protein, whey, etc.) [97,128,213,218–228]. It is expected that highly dehydrated liquid foods can be obtained through FO without affecting the nutrition value. In previous studies, FO has been investigated for use in various juices and sugars to assess osmotic dehydration potential [13,50,51,194–209]. The dewatered products were found to be highly dehydrated through FO, and no significant nutrient loss was observed. In addition, FO has been successfully used for the concentration of other food products and ingredients, including food additives and bioproducts. In comparison with pressure-driven membrane processes such as RO, FO offers lower energy requirements with low hydraulic pressure operation demand and can concentrate feed at ambient temperature without significantly losing nutritional and bioactive components [50,97,202,210,220,222].

Previous studies on the dewatering performance of FO concentration systems have mainly focused on the variation of DSs, membrane types, water flux, and concentration rate of the final products. A more dewatered product (approximately 14.4- to 26-fold) was observed using FO for juice and sugar concentrations, whereas the conventional evaporation method and pressure-driven membrane processes have much lower concentration rates (approximately 7.9- to 14.1-fold, Appendix 3). However, a low water flux condition (below 18 LMH) led to a less dewatered food additive and bioproduct. Owing to the lack of cost-effective DS and suitable commercial membranes, a limited water flux performance challenges the applicability of FO for practical use in the food industry. Thus, high water flux and high product concentrations would be expected if the need for membrane development and the designing of DS can be guaranteed.

3.2.2. *Brine concentration*

As the energy consumption of SWRO is lower than that of conventional distillation techniques, including MSF and multi-effect distillation (MED), it has become the most desirable technology [229]. However, the suitable management and disposal of brine represents a major environmental challenge for most desalination plants. These waters generally comprise waste flow with high concentrations of salts, organics, and other contaminants [230]. As summarized in Appendix 3, two methods of advanced brine management are possible by performing simultaneous concentration (volume reduction) and OD (concentration reduction) [6,40,52,59,108,231–236].

The first approach is to concentrate brine further to decrease its volume. Several ZLD and near-ZLD (feedwater recovery up to 95–98%) technologies have been used in the past to reduce brine volume and management costs. ZLD technology is considered uneconomical and has been used in only a few cases in the past [237]. The FO process avoids phase transitions and can simply carry out salt exchange over the entire membrane, such that the brine is dehydrated without the need for a second membrane, thermal, or solvent-induced separation step [52]. The FO process can also treat high-TDS water, in excess of 70,000 mg/L,

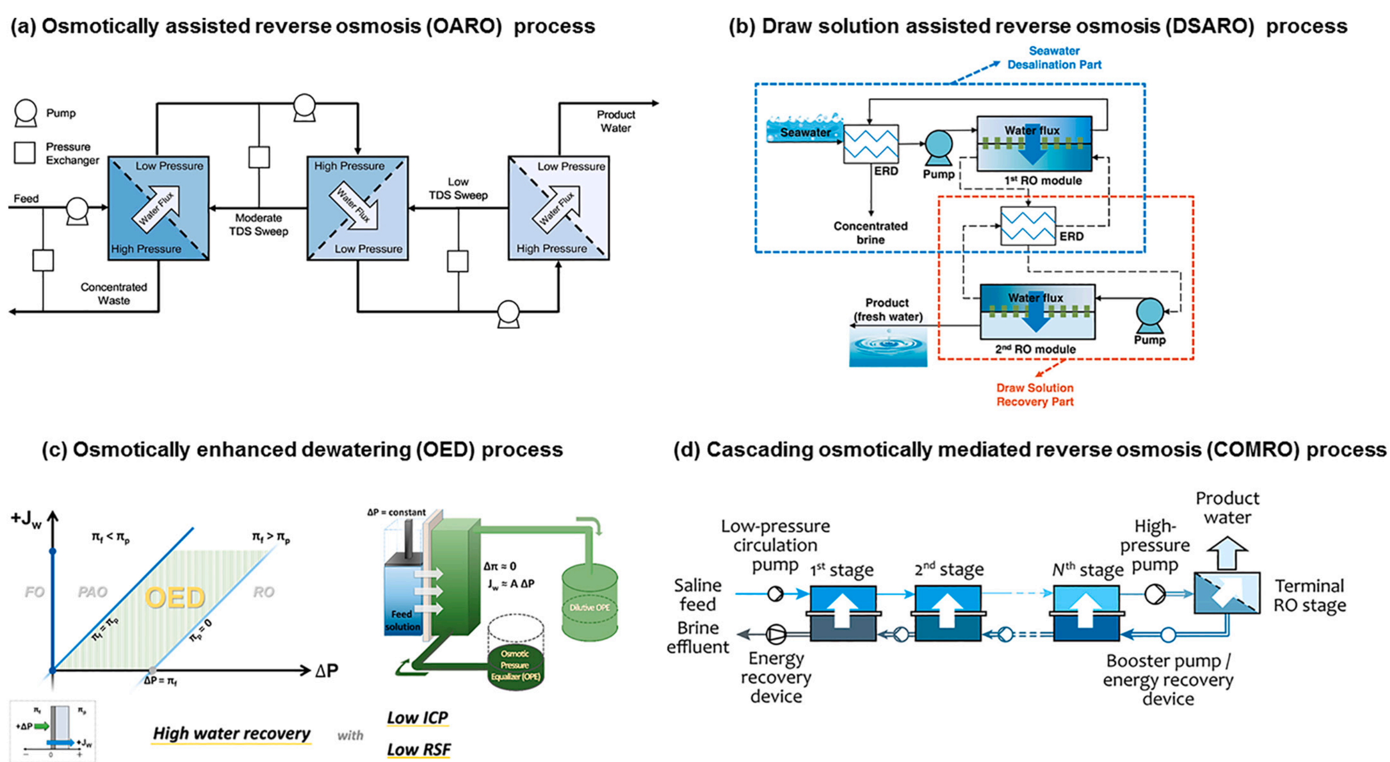

Fig. 8. Conceptual illustration of brine concentration with the energy use of hydraulic pressure and osmotic pressure.

making it suitable for brine treatment. It demonstrates a lower membrane fouling propensity than pressure-driven membrane processes. Membrane fouling in FO is relatively low and more reversible than in conventional methods, and it can be minimized by optimizing the hydrodynamics [237].

Several studies have employed the balance of two different driving forces (hydraulic pressure and osmotic pressure) to achieve high water recovery and energy efficiency. This particular operating mode has variants referred to as osmotic-assisted reverse osmosis (OARO), draw solution-assisted RO (DSARO), osmotically enhanced dewatering (OED), or cascading osmotically mediated reverse osmosis (COMRO) process, as shown in Fig. 8.

In OARO, as shown in Fig. 8a, the osmotic pressure difference across a membrane decreases with a permeate side saline sweep. A series of OARO stages can be used to sequentially reduce the feed concentration until fully desalinated water can be obtained with the conventional RO process. The OARO process was used to obtain reasonable recoveries energy consumption that was less than or comparable to that in the MVC process, which is a widely used high-salinity brine treatment technology [52].

The DSARO process uses the draw solution to induce the transfer of spontaneous water molecules through a low-concentration draw solution, rather than a high-concentration draw solution, to reduce the osmotic pressure difference in Fig. 8b. Therefore, the DSARO process can decrease the high-pressure requirement in the conventional RO process by utilizing a two-stage low-pressure RO process. Although the SEC is higher than that of the conventional RO process, the cost estimation results revealed that the specific water production cost of the DSARO process is 10% lower than that of the conventional RO process [238].

In OED, which is similar to DSARO, a DS with a lower concentration than the FS concentration replaces a fraction of the hydraulic pressure with the osmotic pressure as the driving force, as shown in Fig. 8c. Therefore, compared with the traditional RO process, the OED process can be operated at a lower hydraulic pressure, thereby improving energy efficiency and improving the ability to extract water from saline wastewater [239]. According to a recent study, the OED process was found to have a water recovery rate of 35%–50% with an energy consumption of 6–19 kWh/m^3 of product water for a feed solution of 100–140 g/L NaCl.

To overcome the limitations of conventional RO for hypersaline brine desalination and high-recovery seawater desalination, COMRO adopted a novel design of bidirectional countercurrent RO stages to decrease the hydraulic pressure requirements by reducing the transmembrane osmotic pressure difference, thus simultaneously achieving energy conservation, as shown in Fig. 8d. Compared to the pressure of 137 bar required by conventional RO to desalinate a 70,000 mg/L TDS hypersaline feed, the highest operating pressure in COMRO was only 68.3 bar. In addition, COMRO can realize energy savings of up to 17% [108].

3.3. Simultaneous osmotic dilution and concentration (SODC)

FO technology is generally considered to be an “advanced pretreatment technology” that is not suitable for direct use in desalination, OMBR, or other membrane separation processes of liquid wastewater because FO has an impact on suspended constituents, dissolved organic and inorganic substances, as shown in Fig. 9a. However, none of them have been successfully applied in practical fields, mainly because the proposed recovery method of the diluted draw solution is still inefficient and energy-intensive. FO is applicable and reliable when the diluted draw solution does not need to be recovered.

Herein, we propose an alternative approach for FO membrane separation and the use of feed and draw solutions. As discussed above, finding industrial applications that do not require an external supply of draw solution or a reconcentration process for the diluted draw solution could be an effective solution to promote the real-world commercialization of FO. FO-based systems can be simplified in the application fields (e.g., indirect desalination and brine discharge) by eliminating the recovery process of the draw solution; thus, several economical and technical problems of the draw solution can be easily solved [11].

Based on this, most promising applications of FO should meet the following criteria as shown in Fig. 9b: (1) more than two types of logistics with essentially different osmotic pressures are locally available; (2) a lower saline solution needs to be concentrated; (3) a liquid with a higher osmotic potential does not need to be recovered after its dilution. FO can potentially be a feasible technique as an intermediate process to resolve the current issues as it is energy-effective and operable with a simple configuration.

Furthermore, we found that there may be several industrial applications in which SODC systems are beneficial. The previous research results listed in Appendix 4 show that higher-concentrated feed (approximately 11.5 to 17.2-fold) and higher water recoveries of DS (approximately 70–80%) were obtained for industrial wastewater reclamation. For municipal wastewater treatment, owing to the high fouling potential of foulants in FS, most studies were conducted under a low-concentration condition. Thus, the diluted seawater and brine stream were then regenerated at a lower water recovery. Therefore, to benefit from the simultaneous treatment of two fluids in the FO process,

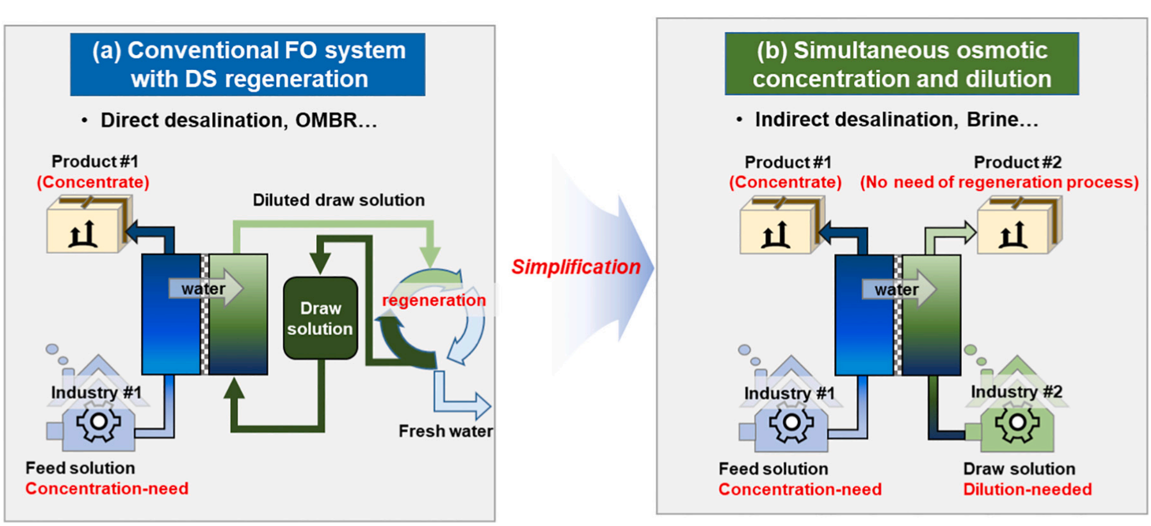

Fig. 9. Conceptual diagrams: (a) conventional FO system consisting of two separate processes for FO and draw solution recovery, and (b) simplified FO that does not require reconcentration process for the diluted draw solution [11].

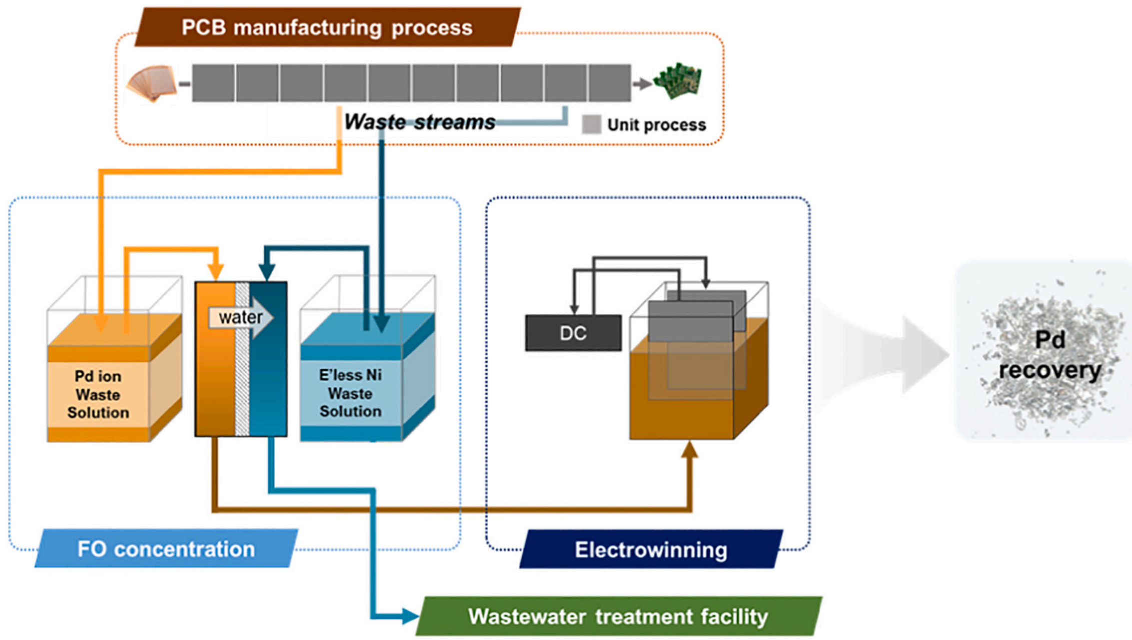

Fig. 10. Conceptual diagram of a new FO-based system for precious metal (Pd) recovery using high-salinity industrial wastewater as the draw solution [11].

water and wastewater streams produced in industrial parks with numerous industry branches should be investigated.

3.3.1. Wastewater reclamation

Given its high fouling reversibility, FO can be directly applied to treat a complex solution without extensive pretreatment, which helps facilitate the development of several system configurations for wastewater treatment and water reclamation. To date, only a few studies have searched for new industrial fields for FO applications that can meet the aforementioned criteria. As summarized in Appendix 4, these studies were inspired by compelling ideas. This approach can revolutionize the field and offer a sustainable solution to the problems of water recovery, draw solution regeneration, and product up-concentration [11,54–59,112]. It will help satisfy the techno-economic feasibility demands with its cost-effective simple designs and high throughput and help address environmental concerns owing to its low energy consumption and minimal waste generation.

Gwak et al. suggested FO as a promising industrial application for precious metal recovery from printed circuit board (PCB) wastewater (Fig. 10) [11]. Palladium-containing wastewater as FS was concentrated by 17.2 times. Palladium could thus be regained efficiently by a subsequent electrowinning system. Nickel-containing wastewater from electroless nickel plating was used as the DS. The diluted DS was disposed in a wastewater treatment plant. This recovery-free FO system driven by a waste stream offers attractive economic advantages including (1) zero cost for the supply and reconcentration of the draw solution and (2) enhanced energy efficiency in the metal recycling process.

Owing to different experimental settings, operating conditions, and data interpretation, it is impossible to compare the efficiency of different application experiments. Thus, it is difficult to assess the potential for FO applications in industry. One approach is to consider the relatively low reported performance of commercial FO membranes, which may be a key barrier to the practical application of FO in the industry. However, the performance cannot be compared based on a given permeate flux

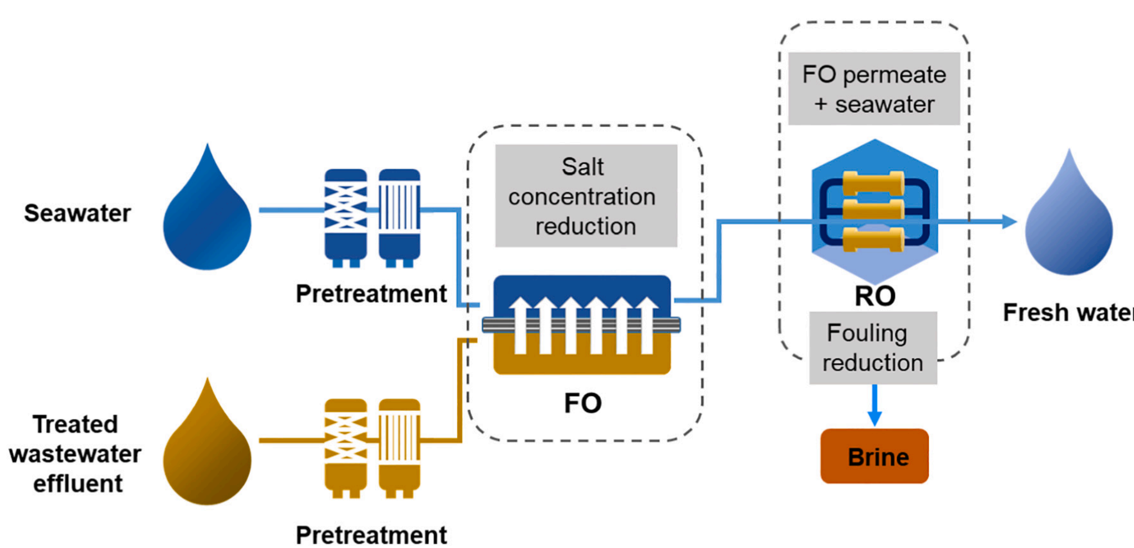

Fig. 11. Schematic diagrams of the FO-RO dilution process for integrating wastewater reclamation and seawater desalination.

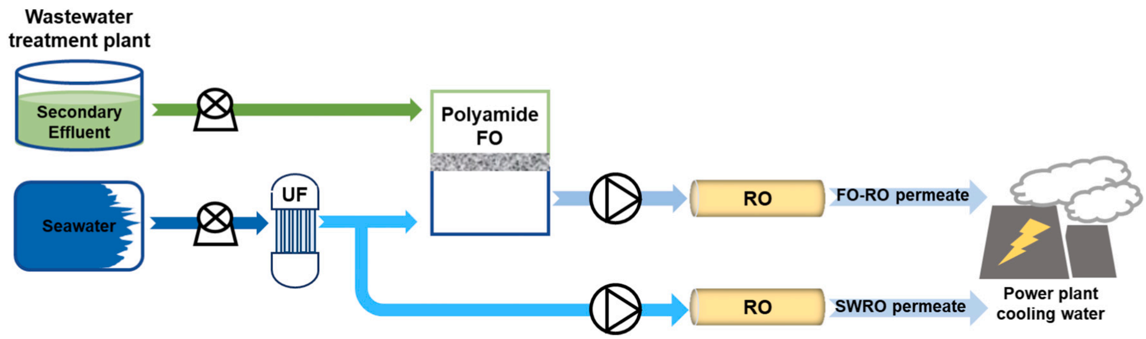

Fig. 12. Schematic summary of FO-RO (OD process and SWRO process) pilot plant [7].

range from the initial short-term permeate flux to the long-term average permeate flux. In addition, because more aspects must be considered, the FO potential must be evaluated separately for each application scenario. These aspects include economic benefits, alternative processing techniques, and legal requirements. With regard to industrial applications, only basic principle verification studies have been carried out on a laboratory scale. Upgrading to pilot-scale or full-scale tests will be the next step in optimizing operations and implementing FO in industrial water and wastewater treatment.

3.3.2. Indirect seawater desalination

The FO process itself can directly desalt a seawater FS by employing a DS with a higher osmotic pressure than that of seawater. However, the energetics of product water recovery and DS component recirculation are not suitable. Recently, researchers have started to explore and assess the treatment performance and economics of the FO-RO dilution process for the simultaneous treatment of impaired/reclaimed water and seawater for acquiring alternative water resources (Fig. 11) [3,7,240–242]. In this configuration, the osmotic pressure induced by the salt in seawater or brackish water is utilized as the driving force to extract fresh water from the low-salinity feed side. In addition to the free-of-charge DS, the attractiveness of this process is to extract clean water from the feed using free osmotic pressure, leading to partial desalination (diluted seawater or brackish water) that can be further desalinated by a subsequent RO step to produce clean potable water. Concentrated impaired water can be further dewatered to recover nutrients for use in fertilizer or bioenergy (biogas) recovery by an anaerobic process. This process is expected to greatly enhance the sustainability of both wastewater reclamation and seawater desalination by reducing energy consumption as well as the environmental impact [7,241].

The FO-RO dilution process decreases the energy demand for desalination and the cost of wastewater treatment. Several studies have investigated different types of quality-impaired water as the feed, including primary and secondary wastewater effluent, and industrial wastewater (Appendix 4) [2,7,14,15,53,75,101,102,104–107,117,119,120,122,240,243–259]. As FO acts as a pre-treatment of wastewater effluent prior to the RO stage, the fouling potential shifts from RO to FO. Coupled desalination processes may achieve higher system recoveries by operating at higher pressures or temperatures without the risk of scaling. The FO-RO dilution process can be applied as a practical and sustainable technology for the full-scale direct potable reuse of impaired water sources for industrial purpose, for example, as feed water to electronics industry or as a cooling water for power plants which are typically located in the coastal areas.

In 2017, Choi et al. used pilot plant consisting of an integrated FO-RO utilizing wastewater from a coal-fired power plant as a feed solution to dilute the seawater (draw solution). The plant was operated over a period of five months. (Fig. 12) [7]. The energy consumption analysis revealed that the energy consumption for desalinating the diluted seawater by FO was 23.3% less than that for typical seawater desalination by RO. The SEC for the integrated FO-RO was 2.85 ± 0.05 kWh/m^3 and that for seawater RO was 3.34 ± 0.05 kWh/m^3. This led to a lower total energy consumption by approximately 15%.

However, the FO-RO dilution process is still in its early stages of development, and many challenges must be overcome to realize commercialization. Because the osmotic pressure difference between wastewater and seawater draw solution is extremely small, a low membrane flux is one of the most critical problems in the indirect FO desalination process. The economic sustainability of the FO-RO dilution process remains uncertain because of the additional investment costs associated with integrating FO units and the larger membrane area required to increase the process water recovery, which increase the process footprint and investment costs.

3.3.3. Brine discharge

As discussed above, FO is advantageous to remove salt from brine and has a low environmental impact after discharge. Conventional disposal alternatives for brine include surface water discharge, deep well injection, evaporation ponds, and land application. The disposal of brine is of great significance both from environmental and economic standpoints, particularly for inland cities. However, the salt concentration in the brine can be reduced by utilizing osmotic power, which also allows the recovery of energy. One of the approaches that can simultaneously solve both problems is the application of pressure-retarded osmosis (PRO) to utilize osmotic energy in SWRO brine. Osmotically diluted RO brine can be either recycled back to the RO process or discharged to the environment because of its low environmental impact [2,14,95,101,106,109,239,255,260–264].

Minier-Matar et al. proposed a novel "single-step" FO process concept, in which waste brine from a thermal desalination plant was used as a DS to remove water from the production/process water (PPW), thereby reducing its volume, as shown in Fig. 13 [255,260,262]. The water from the "pumping" solution does not need to be recovered; thus, no external energy is required, except for the energy required to pump water through the system. The process runs in OD mode and thus consumes low energy.

FO provides an effective solution for brine treatment to avoid the potential risks of brine management using common disposal options and treatment processes. This is because (1) most conventional treatments are ineffective when considering climatic conditions, geographical location, and the environment; (2) conventional treatments can only remove organic matter. As a result, these treatments have a negative impact on the aquatic environment and on humans, and it is challenging to employ environmentally friendly management options. The advantage of saltwater is that it is made up of many precious and rare metals, and recycling them can bring potential profits to the industry. The applications of FO for brine treatment are attractive, reliable, and environmentally friendly because they produce high-quality wastewater and resource recovery. However, fouling is considered to be a major barrier to membrane applications, which leads to decreased efficiency. Therefore, a fouling control strategy in the FO process is necessary.

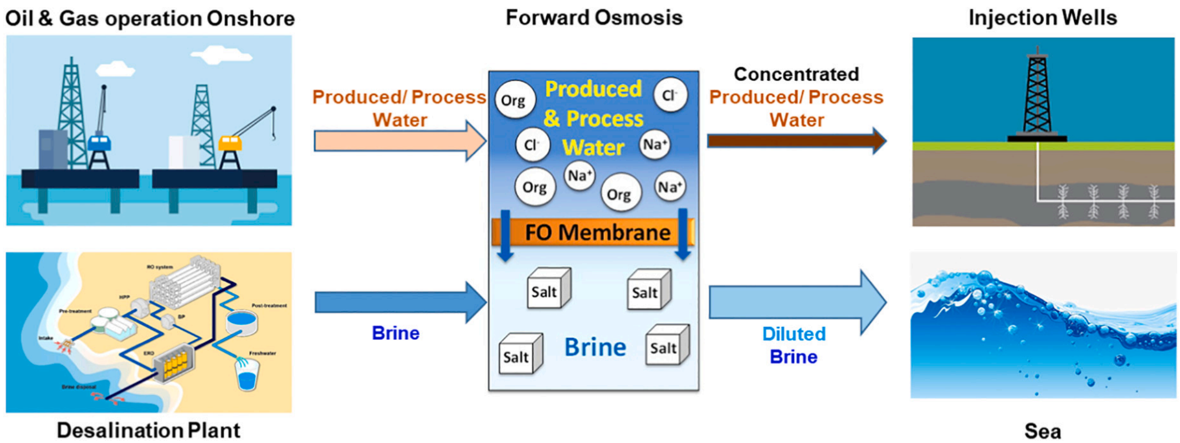

Fig. 13. Application of FO to reduce PPW using brine from desalination plants as draw solution [260].

4. Concluding remarks: future prospects

In this study, we reviewed the state-of-the-art developments of FO membranes, draw solutes, and FO applications, and then classified FO applications into two primary categories: (i) conventional FO systems with DS regeneration and (ii) osmotic dilution/concentration systems without DS regeneration including OD, OC, and SODC. Based on the literature review, current limitations of FO in terms of DS regeneration, energy consumption, and scale-up implementation were identified to overcome the obstacles in commercialization. DS utilization influences the FO performance; the DS must be reconcentrated for its reuse and the production of clean water. This results in an increased energy consumption and hence renders FO an energy-intensive process. Thus, we concluded that an FO process without DS regeneration (i.e., OD, OC, and SODC) has a high potential for the commercialization of the FO process in the future.

A detailed discussion of the three applications was provided. First, we suggested osmotic pumps and FDFO as the most successful applications for the OD process. In particular, osmotic pumps have been intensively utilized in the medical field because of their ability for continuous drug release. In contrast, FDFO application is limited because it cannot sufficiently dilute the fertilizer DS for irrigation owing to osmotic equilibrium; thus, a breakthrough is required for the commercialization of FDFO.

We found that the concentration of liquid food and bioproducts, as well as brine concentration, can be the most feasible for the OC process in the current stage, because FO can readily concentrate a high-concentration solution owing to its driving force (i.e., osmotic pressure gradient). These properties have been applied for concentrating high-value products; such applications have facilitated the use of FO for applications in the production of concentrated organic compounds such as pigments, enzymes, and pharmaceuticals, which need high purity and concentration. However, the development of novel FO membranes with low RSF is required for high-purity production. If RSF can be controlled, it may be beneficial for providing essential components (e.g., salts and sugars) to a concentrated solution.

The SODC concept could be the most successful application of the FO process because the values of both FS and DS can be improved. An example is wastewater reclamation in which FS is concentrated for resource recovery and DS is diluted for discharge. Indirect seawater desalination is similar to wastewater reclamation, but diluted seawater as

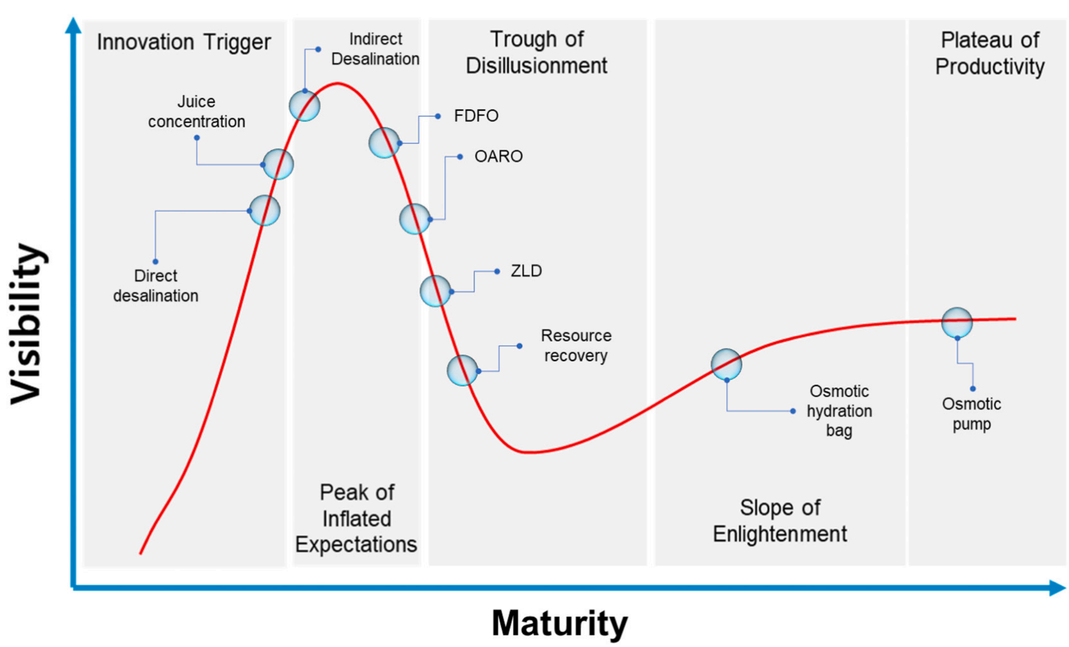

Fig. 14. Hype cycle of FO applications. FO systems were evaluated based on the definition of hype cycles. Detailed information of the Gartner hype cycle is provided in other studies [265].

a DS is further desalted by SWRO, resulting in a low energy consumption. Currently, OARO is receiving growing attention because it can concentrate as well as dilute the RO concentrate or a high-salinity solution with low hydraulic pressure. Therefore, OARO may be the most feasible because there is no necessity for FS posttreatment or DS regeneration.

Based on the review of these FO applications, the hype cycle of the FO process is proposed (Fig. 14) to understand the current status of the FO process. Research on the FO process without the need for DS regeneration has remained in the stage of Trough of *Disillusionment*. FO applications such as OARO have been continuously studied for ZLD and the recovery of valuable resources. However, the engineering of the FO applications is relatively scarce, as most of the research on FO is still at the laboratory scale, and progress in practical applications requires research at the pilot and full scales. Recently, several industrial companies have attempted to demonstrate the SODC concept by pilot-scale and plant-scale evaluations for successful commercialization. Hence, the commercialization of FO applications may thus be possible in the near future.

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Appendix A. Supplementary data

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