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# Current advances in membrane technologies for saline wastewater treatment: A comprehensive review

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#### ABSTRACT

The saline wastewater from various sources including agriculture and industrial activities, appears to have high salt concentration, organic content and other pollutants which can harm the environment. Thus, saline wastewater treatment has become one of the major concerns in many countries. Membrane technology offers great potential in saline wastewater treatment due to its high permeate quality, flexibility, and desalination capability. This paper highlights the current development in various types of membrane processes such as pressure driven-based membranes, forward osmosis, membrane distillation, electrodialysis and membrane bioreactor, either as a stand-alone or integrated process for saline wastewater treatment. The membranes performance in terms of water reclamation as well as resource recovery is discussed. Besides, the membrane fouling issue is highlighted, and the efficiency of various fouling mitigation strategies when dealing with real/challenging saline wastewater are reviewed. Finally, the future challenges and outlook in the context of membrane application for saline wastewater treatment are discussed.

# 1. Introduction

Saline wastewater is considered waste effluent containing a rich amount of salts (NaCl) and organic matter. While the composition of saline wastewater can be very complex, the high salt content is the feature that distinguishes saline wastewater from other types of wastewater. It posed a serious threat to the environment due to the high salt, organic content, and other pollutants. The salts in wastewater can harm the aquatic species and suppress plant germination as well as seed growth. This process will consequently interfere with the ecosystem due to decreasing diversity of species. Furthermore, the irrigation activity using saline effluent can also lead to secondary salinization since water evaporation will leave the salt residue in soils [1]. In some cases, heavy metals might be detected in the saline effluent, depending on the discharge source. It is known that certain heavy metals have carcinogenic and toxic properties. They are also non-degradable and can accumulate in living organisms via the food chain [2]. Thus, heavy metals' presence makes the saline effluent more harmful to the environment if not treated properly prior to discharge.

Over the last ten years, various processes have been reported to treat different types of saline wastewater coming from a wide range of industries. Among the major processes, these can be categorised as physicochemical and biological treatment methods. The physicochemical methods include mainly the advanced oxidation process, membranes, coagulation-flocculation, thermal technique, electrochemical and ion exchange process [3,4]. The biological treatment includes aerobic and anaerobic methods such as the activated sludge method, sequencing batch reactor, biofilm and biofilter reactors, up-flow anaerobic sludge blanket, up-flow anaerobic filter and membrane bioreactor [3]. Recently there have also been increasing interest to use constructed wetlands for saline wastewater [4–6].

This review focuses on the membrane processes that have been used

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for saline wastewater treatment. Fig. 1 shows the distribution percentages of membrane techniques that have been reported since the year 2000 based on a quick analysis of papers published as listed in the Scopus database that contain the terms "saline wastewater" and "membranes". The major techniques that have been reported for saline wastewater treatment are membrane bioreactor (MBR) and membrane distillation (MD). Other membrane techniques such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), reverse osmosis (RO), forward osmosis (FO), electrodialysis (ED) and membrane contactors (MC) have also been reportedly used. It should be noted that the types of membranes used in MBR are mostly of UF type, and thus, UF plays a major role in terms of saline wastewater treatment as well.

The main objective of this review is to discuss the contribution of membrane processes for saline wastewater treatment that have been reported in the literature over the last decades. In this work, we focus on the papers which dealt with pilot-scale study or those that investigated the membrane performance using real saline wastewater rather than synthetic wastewater. The review will start with an overview of saline wastewater followed by an analysis of the membrane technologies that have been used, including those used within an integrated processes framework with other membrane/non-membrane techniques. The review will also touch on fouling and mitigation strategies when dealing with real saline wastewater treatment. This approach is important to provide an insight into how efficient the fouling mitigation techniques are when working with complex or challenging wastewater. Finally, the future challenges and outlook in this area of study will also be discussed.

# 2. Overview of saline wastewater

Generally, saline wastewater can be referred to wastewater containing numerous types of pollutants and dissolved salts. The salinity can be expressed in terms of total dissolved solids (TDS) concentration (mg/ L) which represents the level of soluble salts. It is sometimes expressed in percentage (%); for example, it was reported that the salinity level of the saline wastewater could be in the low range (<0.2%) or vary from 3 to 15%, depending on the source [7]. Besides TDS and salinity percentage, the electrical conductivity (EC) value can also be used to characterize the level of soluble salts.

Various sources of saline wastewater have been identified in the

literature, including those from agriculture activities, industrial sectors (e.g., food, textile, tannery, pulp/paper, mining, petroleum, and coal chemical industries) and secondary sources which originated from water treatment plants. Agricultural drainage coming from saline farmland generates saline wastewater which not only contains salt ions and fertilizer nutrients but also contains pesticides and herbicides [8]. In food industries, salt and brine application in food processing plants associated with seafood products, pickled vegetables, meat canning, and dairy products leads to saline effluent production [3]. For example, the dairy wastewater with a TDS range of 1800–2700 mg/L contains salts that originate from cheese-salting and whey-processing [9]. In Australia (Victoria region), almost 10 billion L of dairy effluent are discharged annually. This type of wastewater needs to be treated properly to meet the water effluent standard quality required by Australian authorities [9].

Meanwhile, the olive oil mills also discharge the saline wastewater (0.5–2% mineral salts) with a strong odour and rich with organic pollutants [10,11]. The presence of phenolic compounds in olive mill wastewater explains why this effluent type is difficult to treat [12]. Besides food industries, the textile industries also contribute to the saline wastewater discharge due to dveing and washing activities [13]. A typical constituent of textile wastewater comprises dyes, suspended solids (SS) (e.g., fibre), phenols and inorganic pollutants, including chloride, nitrogen, heavy metals and sulphate [4,13]. For instance, a wastewater sample taken from Tirupur, a popular textile city in India, displayed the pH, TDS, total suspended solids (TSS), total organic carbon (TOC), chemical oxygen demand (COD) and biological oxygen demand (BOD) values of 8.2, 9960 mg/L, 1340 mg/L, 309 mg/L, 1970 mg/L and 540 mg/L, respectively [14]. In the tannery industry, a substantial amount of saline wastewater is produced from the tanning process, which requires a salt application. It is considered the most polluting industry due to the toxic feature of the wastewater which contains various pollutants including chromium [15,16]. The conductivity of 11.71 mS/cm, the chromium content of 570 mg/L, COD of 2200-3000 mg/L and chloride content of 1691 mg/L characterized the tannery effluent from Istanbul [15]. In another report, the COD of tannery unhairing process wastewater can go up to 20,000-80,000 mg/L [17], while the NaCl concentration of chromium tanning discharge can achieve up to 80,000 mg/L [3].



# Membrane processes for saline wastewater treatment

Fig. 1. Percentages of membrane techniques used for saline wastewater treatment.

The oil and gas industries also generate saline wastewater known as produced water or flowback water during the oil drilling process [18]. It has been estimated that the shale reservoirs could generate 1.7–14.3 million L of produced water per well during the first 5–10 years of production [19]. The produced water composition is associated with the reservoir age, hydrocarbon types and geographic location [20,21]. A typical salinity of produced water from the oil field is higher than 100,000 mg/L (TDS), while the TOC value is 500-2000 mg/L [22]. Also, produced water may contain dispersed oils, heavy metals, and grease [23]. Besides the wastewater from the industries mentioned above, the landfill leachate also has the characteristic of saline wastewater since it contains inorganic salts, ammonia, biological organisms, xenobiotics, heavy metals, and organic pollutants [24]. It was reported that the organic constituents of landfill leachate will be more chemically stable and become more complex as the leachate ages [25]. Thus, the treatment of this type of wastewater is very challenging to fulfil the standard discharge qualities required by the authorities (Table 1).

### 3. Membrane technologies for saline wastewater treatment

In view of the negative influence that saline wastewater has on the environment, its treatment involving the salts and pollutants removal is crucial before effluent discharge. The treated wastewater could be further reused if the water quality fulfils specific standard criteria, thus providing an alternative to minimize the burden on fresh water supplies. Currently, membrane-based technologies have attracted vast interest due to their high permeate quality, flexibility, and desalination capability in water treatment [45]. The following section discusses various membrane-based treatments that have been developed to treat saline wastewater.

Table 1

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Sources	Common constituents in wastewater	Wastewater characteristics	Effluent discharged standards <sup>a</sup>
Textile industries	Dyes, suspended solids (e.g., fibre), sulphate, phenols, salts, nitrogen, heavy metals (e.g. Cr, Sb, Cu, Pb and Cd) [13]	-Bleaching process [26]: TDS = 2500-11,000 mg/L COD = 1200-1600 mg/L TSS = 200-400 mg/L -Dyeing process: TDS = 1500-4000 mg/L COD = 400-1400 mg/L TSS = 50-350 mg/L	-Industrial effluent discharged standard A (US) [27]: BOD <sub>5</sub> = 20 mg/L, COD = 80 mg/L, TSS = 50 mg/L -Indian textile industry standards [28]: pH 6.9, BOD 30 ppm, COD 250 ppm, TDS 2000 ppm, Sulphide 2 ppm, Chloride 500 ppm, Calcium 75 ppm, Magnesium 50 ppm
Tannery/ leather industries	Chromium, salts, organic nitrogen, sulphide, phosphorus, ammonium [29]	Soaking process [29]: TDS = 22,000–33,000 mg/L COD = 3000–6000 mg/L BOD <sub>5</sub> = 2000–5000 mg/L	-Effluent discharged standard (Turkey) [15]: COD = 400 mg/L Chromium = 0.5 mg/L NH <sub>3</sub> -N = 40 mg/L Colorless -New Delhi permissible limit [30]: pH 7.6–9.0, COD = 250 mg/L, TDS = 2100 mg/L, Chromium = 2 mg/L, Chloride = 1000 mg/L, Sulphate = 1000 mg/L
Food industries	-Dairy wastewater: lactose, soluble proteins, lipids, mineral salts, and detergents [31].	Dairy wastewater (salty whey) [9] TDS = 48,000 mg/L COD = 29,000 mg/L	Discharge limit for food industry effluents (India) [36]: Reactive phosphorus <10 mg/L, chloride <750 mg/
	-Seafood wastewater: suspended solids, fats, oils, and grease (FOG), excess nutrients (nitrogen and phosphorus), proteins, sodium chloride	Fish canning and marination effluent [33]: 2–5% (w/v) salt. Fish processing wastewater [34]: COD = 13,180 mg/L	L, sulphate <750 mg/L, ammonia nitrogen <1 mg/L, nitrate <10 mg/L
	-Olive mill wastewater [32]: Suspended solids, phosphorus, phenols, sugar, protein, fats, oils, salt, carbohydrate, pectin, inorganic compound	BOD = 3250 mg/L Olive mill wastewater [11,35] EC = 11.30-23.5 mS/cm COD = 55,730-156,000 mg/L Total phenol = 2439-8300 mg/L	
Oil and gas industries	Typical produced water contains dispersed oil, grease, salts, organic compounds, heavy metal, radionuclides, benzene, toluene, ethylbenzene and xylene (BTEX), carboxylic acid and polycyclic aromatic hydrocarbon [23].	$\label{eq:produced water from oilfield [22]} TDS > 100,000 \mbox{ mg/L} TOC = 500-2000 \mbox{ mg/L} \\ pH = 5.6 \end{tabular}$	Permissible surface water discharge (US) [37] TDS = $500 \text{ mg/L}$ pH = $6.5-9.0$ Oil and grease = $0.01 \text{ mg/L}$
Pulp/paper mill industries	Lignin, stilbenes, phenols, dioxins, chlorides, furans, sulphur compounds, chloride, phosphate, acetic acid, propionic acid	-Paper and paper recycling mill effluent [38] TDS = 395–2500 mg/L COD = 480–4450 mg/L pH = 6.1–8.3 -Kraft mill effluent [39] COD = 1124–1738 mg/L SS = 37–74 mg/L pH = 10.1	-Permissible limits for bleached Kraft mill effluent [40] US standard SS: 3.86 kg/adt, BOD:2.41 kg/adt France standard SS: 6.5–10 kg/adt BOD:3.3–30 kg/adt COD:48.95 kg/adt -Permissible limit for paper mill effluent (Malaysia) [41] COD = 80 mg/L
Landfill leachate	Inorganic salts, ammonia, biological organisms, xenobiotics, heavy metals (e.g.: Cd, Pb, Cr) and organic pollutants [24].	EC = 10.74-35.8 mS/cm COD = 2042-69,470 mg/L [42,43]	Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (China) [44] COD = 100 mg/L, BOD <sub>5</sub> = 30 mg/L, Ammonia = 25 mg/L. Total phosphorus 3 mg/L, Total nitrogen = 40 mg/L, SS = 30 mg/L $C_{2}^{6+} - 0.05$ mg/L

<sup>a</sup> Only selected parameters are displayed here due to many parameters in certain effluent discharge standards.

#### 3.1. Pressure-driven membranes

The pressure-driven membrane processes refer to the membrane filtration techniques driven by a pressure exerted on the solution at one side of the membrane to separate the stream into a permeate and retentate. Four categories of membranes that typically operated under pressure-driven mode are MF, UF, NF and RO, in the order of decreasing pore size and increasing rejection capability. The dominant separation mechanism of MF and UF is mainly by sieving, where both are normally used to remove particulates and bacteria from the water [46,47]. On the other end of the spectrum, solution-diffusion has been reported as the main separation mechanism for RO. It can prevent dissolved mineral ions' permeation, apart from the impurities rejected by the other three classes of membranes [48]. In between UF and RO is the NF membrane, where its charged surface plays a critical role in exerting electrostatic repulsion towards impurities, especially multivalent ions and relatively small charged organic compounds [49,50].

# 3.1.1. Microfiltration and ultrafiltration

The application of these membranes in saline wastewater treatment heavily relies on their characteristics and separation mechanism or rejection capability (Table 2). For instance, MF has been used to remove the suspended solid content and turbidity of dimethylformamide (DMF) wastewater from polyurethane synthetic leather factory [51]. The presence of suspended particulates was found to cause blockage of equipment pipes and distillation columns and subsequently hindered the DMF recovery from the wastewater. In this case, Zhang et al. [51] showed that ceramic MF membrane could reduce the turbidity of DMF wastewater by 99.62% and remove the suspended solids by 99.99%. This process produced permeate with low turbidity and suspended solid content at 1.7 NTU and 0.01 mg/L, respectively, where the wastewater

#### Table 2

Application of pressure-driven membranes in saline wastewater treatment

quality was much improved and could benefit the subsequent DMF recovery processes. The high filtration efficiency could be attributed to the smaller membrane pore size (0.2  $\mu$ m), capable of retaining the larger impurities (average size of 1.39  $\mu$ m). Though membrane fouling was inevitable, the foulant cake layer (formed from the accumulation of impurities on the membrane surface) could be removed via a combination of flushing, ultrasonic, NaOH, and NaCIO cleaning processes. The pure water flux was successfully recovered to 608.2 LMH, with a recovery rate of 96.8%.

Similarly, UF has also been employed in pulp and paper mill wastewater treatment, as the effluent normally consists of high-molecular-weight compounds that could be easily retained by UF. Gonder et al. [52] demonstrated that UF could attain high rejection/reduction for total hardness (83%), sulphate (97%), spectral absorption coefficient (95%), and COD (89%) for the treatment of pulp and paper mill wastewater. However, the membrane could not efficiently reduce the effluent's conductivity, as rejection lower than 50% was reported. This result could be due to the larger pore size of UF that fails to retain the dissolved ions from permeating through the membrane. The observation signifies the role of UF as a membrane process to remove high-molecular-weight impurities rather than dissolved ions, where NF or RO would be more suitable for such ion rejection.

# 3.1.2. Nanofiltration and reverse osmosis

The removal of coarse particles and large organic compounds in wastewater is not the only desired aim for the treatment process, as some effluents contain a high concentration of dissolved ions that pose hazardous salinity levels to the environment for reuse purposes. Unfortunately, MF and UF membranes are not particularly good in removing dissolved monovalent and multivalent ions in the water due to their larger pore size. This sets the boundary where MF and UF's role is more

Membrane	Saline wastewater type/source	Wastewater characteristics	Membrane performance	Reference
MF	DMF wastewater from the leather industry	$EC = 698 \ \mu S/cm$	Pure water flux = $608.2 \text{ LMH}^{\text{b}}$	[51]
	(Jiangsu, China)	SS = 408  mg/L	Recovery rate = $96.8\%$ .	
		pH = 7.68	SS removal = 99.99%	
		Turbidity = $452.50 \text{ NTU}^{a}$		
UF	Pulp and paper mill wastewater (Turkey)	$EC = 1200 \ \mu S/cm$	Removal efficiency:	[52]
		Total hardness = $480 \text{ mg/L}$ as CaCO <sub>3</sub>	Total hardness = $83\%$ , sulphate = $97\%$	
		Sulphate = $150 \text{ mg/L}$	COD = 89%	
		COD = 850  mg/L	EC = 50%	
		pH = 7.0		
NF	Olive mill wastewater (Spain)	EC = 3.2 - 3.6  mS/cm	Flux = 69.9 LMH	[53]
	-	$Cl^- = 875.8 - 1045.1 \text{ mg/L}$	Feed recovery $= 90\%$	
		$Na^+ = 534.0 - 728.7 \text{ mg/L}$	Removal efficiencies:	
		COD = 150  mg/L	EC = 55.5%, COD = 88.5%	
NF	Landfill leachate concentrate (China)	EC = 10.74  mS/cm	Desalination efficiency $= 99.5\%$	[42]
		COD = 2042.9  mg/L	Recovered humic substance purity $= 98.3\%$	
		$Cl^- = 2887.1 \text{ mg/L}$	Rejection:	
		Humate substance = $1473.9 \text{ mg/L}$	Humic substance $= 98.9\%$	
		Humic acid = $285.2 \text{ mg/L}$	Salt = 6.5–7.5%	
NF	Textile wastewater (Spain)	EC = 2.6 - 2.8  mS/cm	Permeate:	[57]
		$Cl^- = 200-365 \text{ mg/L}$	COD = 15  mg/L	
		$Na^+ = 179-190 \text{ mg/L}$	$EC = 2740 \ \mu S/cm$	
		COD = 200-315  mg/L	Hardness = $6.69 \text{ mg/L}$	
		Hardness = $133-171 \text{ mg/L}$	Dye 100% removed	
RO	Textile wastewater (Turkey)	$EC = 5500 \ \mu S/cm$	Permeate	[63]
		COD = 1000  mg/L	Flux = 19 LMH	
		SS = 276  mg/L	Water recovery 70%	
		$\mathrm{Cl}^-=1000~\mathrm{mg/L}$	$EC = 300 \ \mu S/cm$	
RO	Landfill leachate (anaerobic landfill in	EC = 28.1 - 31.8  mS/cm	Permeate	[64]
	Southwest China)	Total nitrogen (TN) = 2911.70 mg/L	EC = 0.15 - 0.22  mS/cm	
		COD = 5690.70  mg/L	Water recovery >83%	
		$Cl^- = 6460.60 \text{ mg/L}$	Removal of organic matter >80%	
RO	Mining industry wastewater (Victoria,	TDS = 5660-7910  mg/L	Rejection efficiency:	[66]
	Australia)	Turbidity = 39.4–160 NTU	Turbidity 85%, TDS 96%, Antimony 95%, arsenic	
		Antimony = $36.4-50.2 \text{ mg/L}$	66%, nickel 82%	

<sup>a</sup> NTU stands for Nephelometric Turbidity unit.

<sup>b</sup> LMH unit represents L/m<sup>2</sup>·h.

to remove larger impurities as sole treatment process or pre-treatment for other processes. In this context, an NF membrane with a tighter pore size and charged surface could be employed to reduce the wastewater's EC value. For instance, Ochando-Pulido et al. [53] deployed NF as the tertiary treatment for olive oil mill wastewater to bring down the hazardous EC value from 3.2-3.6 mS/cm to 1.5 mS/cm. With the adoption of NF, the dissolved ions have been halved, which could not be removed by the primary and secondary physicochemical processes (precipitation, Fenton, flocculation-sedimentation, and olive stone filtration). The additional benefit was the further reduction of COD value in the treated effluent down to 17.3 mg/L. The reduction of EC and COD enabled the treated water to be re-used for irrigation purposes and complied with the water quality standard values for discharge into public waterways. In another study, Ochando-Pulido et al. estimated the necessary overdesign of the membrane operation was 9.42-17.53%, which indicated a maximum required membrane area of 61.82 m<sup>2</sup> for the treatment plant [54]. This result implies that just two membrane modules should be installed for the operation, boosting the proposed NF treatment process's economic feasibility both from an operational and capital costs perspective.

Another study of pilot-scale NF treatment of olive mill wastewater has demonstrated that NF was technically and economically feasible. Sanches et al. [55] reported that NF treatment, combined with other processes (dissolved air flotation and Fenton), recorded a total operation cost of 4.17–4.69  $\notin/m^3$ . This value was considered feasible as it only represented approximately 54–60% of the total operation cost associated with the treatment processes (coagulation/flocculation and Fenton) currently applied in the olive mill wastewater system. Such analysis demonstrated the competitiveness of the NF process in both technical and economic feasibilities.

The paradigm shift in sustainable wastewater management has encouraged the water operators to investigate the possibility of extracting useful resources from the wastewater instead of just treating and disposing it to the environment. In this context, Ye et al. [42] presented an interesting finding on using NF as an efficient process to fractionate the humic substances and inorganic salts from landfill leachate. The NF membrane surface was tailored by mussel-inspired modification through dopamine self-polymerization, which conferred the membrane a superior selectivity between humic substances and salts from the landfill leachate. The NF membrane demonstrated promising separation of the two components, recording a high rejection of humic substances (98.9  $\pm$  0.5%) while possessing low rejection (6.5–7.5%) towards the inorganic salts (represented by conductivity reduction). Further dialysis using the same NF membrane successfully concentrated the humic substances from 1779.4 to 17,247.1 mg/L with 96% recovery and attaining 99.5% desalination efficiency. This process produced a high purity (98.3%) of humic substances potentially used as organic fertilizer application.

The same resource recovery concept has also been reported for tannery wastewater treatment. Galiana-Aleixandre et al. [56] employed NF membrane to separate and recover sulphates from tannery wastewater. The NF membrane displayed high sulphates rejection (around 97%), producing a concentrated sulphates stream that possesses a high potential to be reused in the tanning process. The high rejection towards sulphates ions was attributed to size exclusion mechanism separation. Based on the experimental data, it was postulated that as much as 14.79 kg sulphates/ton of raw hide could be reused in the tanning process. The reuse of sulphates can lead to savings in chemicals and water, where savings up to 10  $\epsilon$ /ton of raw hide can be achieved. The additional benefit was that the NF-filtered wastewater, which contained lower sulphates concentration (<1000 mg/L), can be discharged to a municipal wastewater treatment plant. The lower amount of sulphates will generate less H<sub>2</sub>S in the biogas from the anaerobic sludge stabilization and subsequently minimize the gas engine corrosion risk.

Another application of NF for resource recovery is in the textile industry. The textile industry is a water-intensive industry and generates a huge amount of wastewater laden with hazardous dyes pollutants. Recovery of water from the effluent for reuse has been deemed a sustainable practice to cut down the textile industry's water consumption. In this scenario, NF with a smaller molecular weight cut-off between 150 and 300 Da appears to be a good candidate to remove most of the dyes from the textile wastewater since the pollutants possess molecular weight ranged 300-1000 Da [57]. In a pilot-scale study conducted by Ong et al. [58], the hollow fibres NF demonstrated superiority in reducing COD (reflecting organic textile dyes' presence, ranged 3000-8000 ppm) in industrial textile wastewater and maintained above 95% rejection rate throughout the 45 days operation. Such a high rejection rate indicates the prevention of the release of dyes to waterways. While preventing the permeation of dyes, the NF membrane allowed the passage of NaCl (80%) and Na<sub>2</sub>SO<sub>4</sub> (90%) through the membrane. This process offers the potential to recover and reuse these salts in the subsequent dyeing process. However, membrane fouling is a challenge for long-term operation, and it should be addressed to minimize the operation cost of frequent membrane cleaning.

Compared to NF with high selectivity in rejecting impurities, RO has been known for its capability to reject almost all impurities in the water, especially the monovalent ions that other membranes fail to reject. This capability leads to the commercial deployment of RO to desalinate seawater and brackish water for the production of clean drinking water [48]. However, the accumulation of a high concentration of salt ions in the rejected stream, also known as hypersaline brine, with a TDS concentration above 70,000 ppm, poses a handling challenge for the inland desalination operators. A suggestion has been proposed to treat the hypersaline brine with high-pressure RO that can be operated above 80 bar [59]. Though such group of RO membranes is not as diverse as other polymeric RO membranes and it has not been widely adopted in the industry, there are a few high-pressure RO membranes developed for the market, such as the Pall Disc Tube™ Module System by Pall Corporation, Specialty Membranes XUS180804 and XUS180802 Ultra-High Pressure RO Element by DuPont<sup>™</sup>, and TM800M by Toray [60–62]. The manufacturers claimed that the RO membranes could be operated at above 80 bar. However, data of such operation has been rare and limited. Nonetheless, RO is generally employed for water recovery in various industrial wastewater treatments due to the effluent's high conductivity. For instance, even though high colour and COD reduction is possible with the combination of biological and chemical treatment processes, water recovery for the textile industry is not practical due to the high conductivity. In this context, Sahinkaya et al. [63] have demonstrated the adoption of RO could reduce the conductivity of textile wastewater from 5500  $\mu$ S/cm to 300  $\mu$ S/cm and attain 70% water recovery. The substantial removal of dissolved ions enabled the reuse of permeate in the dyeing process.

Apart from the salinity, the additional benefit of employing RO for water recovery is its capability to reject other contaminants that are difficult to be removed by other technologies. Wu and Li [64] reported that the two-stage disk tube-RO treatment system was capable of reducing the EC value of mature landfill leachate from 28.1–31.8 mS/cm to 0.15–0.22 mS/cm while at the same time removing the refractory organic matter (99.8%) and nitrogenous (99.97%) pollutants to a level suitable for discharge. The RO system performed well in rejecting the pollutants and attained a high-water recovery rate (>83%).

In conventional tannery wastewater treatment, the treated effluent will be discharged into the municipal sewer system for further treatment before being released to the environment. The role of RO as a potential tertiary treatment method for the tannery wastewater after pre-treatment has been explored to determine if the water could be recycled instead of being discharged into the municipal sewer system. George et al. [65] reported that the pre-treated tannery wastewater which was then filtered by RO has attained the good effluent quality. The chromium concentration, turbidity and conductivity were reduced by 99%, 100% and 95%, respectively. This water quality was on par with the well water that the tannery processes were currently using,

indicating the feasibility of adopting RO for water reclamation and reuse in the tannery industry. Cost analysis also supported this concept, where the total maintenance and operations cost for the RO system (\$101.26) was less than the total operations cost for withdrawing and disposing of the water through conventional methods (\$142.16). The potential cost savings of using RO treatment to recycle tannery wastewater could offset the capital costs of RO system within a short period of time. Nonetheless, a full-scale study is necessary to determine if such application is cost saving with an initial capital cost for implementing a sustainable process (water reclamation and reuse).

RO has also been well-accepted in the treatment of mining wastewater. Untreated or poorly treated mining effluent will cause disastrous impacts to the environment since it contains toxic pollutants that may cause reduced reproduction and deformities. Samaei et al. [66] presented the performance efficiency data of RO as a post-treatment process in mining operations in Victoria, Australia, within the timeframe of 2015-2018. In general, the RO process achieved satisfactory rejection efficiency for the entire evaluation period, particularly for turbidity (85%), TDS (96%), antimony (95%), arsenic (66%), and nickel (82%). The permeate met the limits specified by the Environmental Protection Authority of Victoria. Several anomalies occurred during the observed period where the RO permeate failed to comply with the discharge limits. The diagnosis indicated that the RO feed quality's inconsistency was the prime factor contributing to RO membrane fouling and the noncompliance of TDS and antimony levels in RO permeate. Hence, having a reliable and extensive pre-treatment is crucial for RO, especially in harsh operating conditions where the effluent impurities may result in membrane degradation and physical damage. Since pressure-driven membrane processes are susceptible to fouling and given the diverse constituents of wastewater, a standalone membrane process would not be sufficient to remove all the pollutants efficiently. Other treatment processes must be combined with the membrane processes through integration or hybridization.

# 3.2. Forward osmosis

Forward osmosis (FO) is a membrane separation process that operates based on the osmotic pressure difference. In FO filtration, the chemical potential gradient across the membrane allows the water permeation from the feed solution to the draw solution (DS), i.e., from low to high osmotic pressure region. Hence, the DS will be diluted while the feed side will be concentrated due to the water loss. In saline wastewater treatment, the FO process can be applied to extract the water from the feed solution for further reuse and concentrate effluent for other processes such as resource recovery. The FO membranes development has begun to receive much greater attention in the last 20 years since it offers numerous advantages such as lower fouling propensity than that of pressure-driven membranes (Fig. 2). The absence of hydraulic pressure in the FO process may reduce the cake layers formation on the membrane surface [67,68]. Furthermore, the FO process cost could be lowered due to the less energy consumption as no additional pressure is needed to achieve the organic/inorganic rejection [69].

FO process can remove nearly all dissolved ions and suspended solids. Previously, FO treatment has been applied to treat saline wastewater from various sources, as tabulated in Table 3. A recent work by Liden et al. [70,71] reported that FO is applicable for water extraction from produced water with high salinity (16,000-210,000 mg/L TDS). However, the concentration equilibration issues between the DS and feed solution, membrane fouling and temperature swing caused the flux to decrease over time. Sustainable water recovery from tannery wastewater, also known as "dirtiest water", was investigated by Mahto et al. [72]. They found that the use of a deep eutectic solvent (DES) as DS in FO treatment was capable of extracting >90% water from various feed solution, including tannery wastewater. The NaCl content of the feed tannery wastewater increased from 786 to 2732 mg/mL as the feed solution was concentrated after the FO process. Nevertheless, the DES recovery system design needs to be further improved to enhance the DS recovery.

A recent report also demonstrated that FO membrane is efficient in terms of heavy metal rejection [73,74]. The hydrated ionic radius of heavy metal ions is mostly larger than that of seawater monovalent ions. Since the thin dense layer in a composite FO membrane is specially created to remove small ions (e.g., Cl<sup>-</sup> and Na<sup>+</sup>), it is expected that FO would be highly effective to reject heavy metals with a larger ionic radius [73]. In a study conducted by Meng et al. [73], the performance of FO in terms of toxic heavy metal (antimony (Sb)) rejection from the real dyeing wastewater (conductivity >100 mS/cm) was investigated. It was reported that the FO treatment of dyeing wastewater was challenging due to its high conductivity, which resulted in the decrease of the osmotic driving force. This leads to the lower water flux (<9 LMH) compared to that of synthetic wastewater treatment. However, increasing the feed solution's pH (pH 3 to 11) can increase the water flux and Sb rejection. Their finding demonstrated that the aquaporin FO membrane was effective to remove the Sb (>99.7% rejection) as well as other inorganic and organic contaminants (90%) during the treatment of real dyeing wastewater.



Fig. 2. Comparison of FO process with less fouling propensity and RO process.

#### Table 3

Application of FO in saline wastewater treatment.

Saline wastewater types/source	Wastewater/feed solution characteristics	Draw solution	Performance	Reference
Tannery wastewater (Chennai, India)	TDS: 60,000 ppm (pre-treated, organic free)	Deep eutectic solvent	Average osmotic flux of~1.5 LMH NaCl content in feed solution increased from 786 to 2732 mg/mL. Water recovery >90%	[72]
Dyeing wastewater from textile and printing industries (China)	EC >100 mS/cm Sb = 0.20 mg/L COD = 398.11 mg/L TOC = 170.70 mg/L	NaCl	Rejection: Sb > 99.7% COD and TOC $\sim$ 90% Water flux = 8.08–8.78 LMH	[73]
Agricultural wastewater (i.e., livestock wastewater, USA)	EC = 4.66 mS/cm COD = 4166.67 mg/L Cl <sup>-</sup> = 104 mg/L PO <sub>4</sub> <sup>3-</sup> P = 166.50 mg/L Mg <sup>2+</sup> = 15.07 mg/L NH <sub>4</sub> <sup>-</sup> N = 413.33 mg/L	MgCl <sub>2</sub>	Water recovery>50% Water flux = 3.12 LMH Phosphate recovery >99% Ammonium nitrogen removal >93%	[75]
Landfill leachate (Virginia, USA)	$EC = 35.8 \text{ mS/cm}$ $COD = 69,470 \text{ mg/L}$ $NH_{4}^{4}\text{-N} = 2753.0 \text{ mg/L}$ $Mg = 722 \text{ mg/L}$ $Cl^{\circ} = 5957 \text{ mg/L}$	NaCl	Struvite (Mg) recovery $98.6\%$ Leachate volume reduction $37\%$ Water recovery = $36.6\%$	[43]
Shale gas flowback produced water (Sichuan Basin, China)	$\begin{split} EC &= 36.34 \text{ mS/cm} \\ TDS &= 22,530 \text{ mg/L} \\ Cl^- &= 13,020 \text{ mg/L} \\ Na^+ &= 8350 \text{ mg/L} \\ Turbidity &= 0.16 \text{ NTU} \end{split}$	Fertilizer solution (NH <sub>4</sub> H <sub>2</sub> PO <sub>4</sub> )	Flux recovery 92%	[80]
Textile wastewater (textile industry, Slovenia)	EC = 2.2 mS/cm COD = 2862 mg/L TDS = 1944 mg/L TSS = 144 mg/L	MgCl <sub>2</sub> , Concentrated dyeing solution	Water recovery 55% Rejection: Dye 100% COD 95%, TDS, TSS, Cu <sup>2+</sup> , Zn <sup>2+</sup> > 99%	[82]

In the context of resource recovery, several works regarding FO application to promote valuable resource recovery from saline wastewater was reported in the literature [43,75,76]. For instance, Wu et al.  $\left[75\right]$  studied the effect of  $MgCl_2$  as the DS to obtain the struvite from agricultural wastewater (i.e., livestock wastewater) using the FO system. The FO was proposed as an alternative treatment in their work since conventional livestock wastewater treatment such as anaerobic digestion can only reduce the organic concentration without realizing the nutrient recovery. In their study, the pre-treated digested swine wastewater with 4.66 mS/cm conductivity, 4166 mg/L of COD, 166.50 mg/L of PO\_4^{3-}, 413 mg/L of NH\_4^+, 15.07 mg/L of Mg^{2+} and 104.77 mg/L of Cl^ions was fed to cellulose triacetate FO membrane. Unlike other work which added an external magnesium source to promote struvite precipitation in the feed solution [77], they saw the reverse salt flux  $(Mg^{2+})$ from DS to feed side as an opportunity to promote the in-situ struvite recovery. Results revealed that the highest water recovery (>50%) and > 99% phosphate recovery were attained using 0.5 M of MgCl<sub>2</sub>. Their findings suggested that it is possible to recover the struvite via a combination of reverse-fluxed (Mg<sup>2+</sup>) and nutrients such as PO<sub>4</sub><sup>3-</sup>P and NH<sup>+</sup><sub>4</sub>-N in the feed solution, rather than the conventional method, which directly adds MgCl<sub>2</sub> to the centrate for struvite precipitation. In landfill leachate treatment, the chemical pre-treatment in feed solution is essential before being subjected to FO process to enhance the resource recovery. However, it shall be noted that the addition of chemical during pre-treatment will increase the ionic concentration in the landfill leachate, thus reducing the osmotic pressure gradient across the FO. Wu et al. [43] investigated the trade-off between water recovery capability and ionic concentration in feed solution as well as the treatment configuration. The lowest water flux (0.05 LMH) was observed after a 60-h operation when the leachate was subjected to calcium pretreatment and struvite precipitation steps before the FO process. Such observation was due to the increase in leachate conductivity (39.5 mS/ cm) after the addition of Na<sup>+</sup> ions during pre-treatment, further reducing the driving force for water recovery through the FO membrane. However, the application of FO process in between calcium pretreatment and struvite precipitation step was found to be the optimal configuration in their work as that system had successfully achieved

98.6% magnesium recovery and attained the highest water extraction by reducing 37% volume of the landfill leachate.

In developing the FO treatment, the selection of DS is one of the important criteria to ensure the FO process's efficiency. The DS nature should suit the targeted industrial application and should also be inexpensive [20]. Moreover, the diluted DS's re-concentration after FO treatment is required if one intends to recover the clean water trapped inside the DS. In this case, the FO can be integrated with another process, such as distillation to regenerate the DS and water extraction [78]. Various DS types have been implemented in FO treatment, including salt solutions (e.g., NaCl and MgCl<sub>2</sub>), fertilizer solution and brine from the desalination process [79]. Unlike common DS such as NaCl and NH<sub>4</sub>HCO<sub>3</sub> which need to be recovered for re-use purpose, the use of fertilizer solution serves as an alternative DS which does not require any regeneration step and can be used for other application. Chang et al. [80] implemented the fertilizer drawn FO (FDFO) to concentrate the pre-treated real shale gas flowback produced water for irrigation purpose. In their work, different types of fertilizer solution such as KCl and NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> were applied as DS. The best FO performance was observed when NH<sub>4</sub>H<sub>2</sub>PO<sub>4</sub> was used as DS. Meanwhile, severe membrane fouling was reported for the case of (NH<sub>4</sub>)<sub>2</sub>HPO<sub>4</sub> as DS. In another work, Li et al. [81] used the NH<sub>4</sub>HCO<sub>3</sub> fertilizer solution as the DS in the FO unit during the treatment of landfill leachate. They found that the application of  $\mathrm{NH_4HCO_3}\ \mathrm{DS}$  effectively removed the polycyclic aromatic hydrocarbons (PAHs) compounds and attained 91.6% water recovery rate. The toxicity test revealed that the diluted DS has no toxic effect on zebrafish and its composition has fulfilled the local environmental safety standard for liquid fertilizer. Thus, it can be directly used for irrigation. In textile wastewater treatment application, Korenak et al. [82] introduced the idea of using concentrated dyeing solution as the DS to avoid the DS recovery process. The FO treatment process effectively removed the dye (100% rejection) and attained a COD rejection of 95%. Other contaminants, including TSS, TDS,  $Zn^{2+}$  and  $Cu^{2+}$ , were rejected more than 99%. The long-term filtration test accompanied by the cleaning procedure revealed that the FO membrane achieved 100% flux recovery without any significant irreversible fouling. Nevertheless, the DS from dyes exhibited a slightly higher reverse salt flux value than typical DS

such as NaCl and MgCl<sub>2</sub>.

#### 3.3. Membrane distillation

Membrane distillation (MD) is a thermal-driven separation process that has been developed by integrating the concept of thermal distillation and membrane filtration [83]. During the MD process, the vapour transport takes place from the feed side to the permeate side due to the temperature difference. The membrane's hydrophobic property allows only vapour permeation while liquid remains on the feed side and could not enter the membrane pores. This special working principle makes MD feasible for treating high salinity wastewater, which could not be treated by osmotic-driven and pressure-driven membrane processes [84]. Hypothetically, the MD can reject 100% non-volatile components, which can be further used for the next processing step [84,85]. This functionality allows the MD process to recover clean water in the permeate side while at the same time producing the concentrate solution in the retentate side. Moreover, the MD process could also be integrated with other heat sources, including low-grade waste heat and solar energy, minimizing its carbon footprint [86,87]. Since MD is not very sensitive towards salinity level, it can desalinate the highly saline wastewater such as produced water [20,88,89]. These advantages explain why MD has attracted vast attention in the saline wastewater treatment process. Several types of MD configuration have been developed by researchers, such as direct contact MD (DCMD), air gap MD (AGMD), sweeping gas MD (SGMD) and vacuum MD (VMD) [90]. The difference between these configurations can be explained in terms of the setup for vapour condensation. For example, the condensation plates of AGMD are located inside the modules (Fig. 3), while SGMD and VMD utilize external condensers [84].

Today, numerous MD configurations have been applied in the treatment of the saline wastewater, as summarized in Table 4. Among them, the DCMD has been the most researched configuration due to its easy setup [84,91]. In olive mill wastewater treatment, the use of DCMD is promising to recover the phenolic content in the concentrate since it is a non-destructive method towards the phenol compound [92]. Previously, El-Abbassi et al. [92] used commercial polytetrafluoroethylene (PTFE) membranes in their DCMD system to treat the real olive mill wastewater taken from a Morocco region. Membranes with different pore sizes (0.2–1.0 µm) were tested in different temperatures to recover clean water and concentrate the olive mill effluent, which was rich with polyphenol. It was found that the DCMD treatment at high temperature (80 °C) did not affect the phenolic content and its antioxidant feature. However, the low permeate flux and flux reduction over time remain as the limitation of DCMD. In another work, El-Abbassi et al. [35] combined the DCMD with osmotic distillation to further improve the

treatment of olive mill wastewater (conductivity ~23 mS/cm) at the low-temperature difference (20 °C). In this system, an extracting salt solution (CaCl<sub>2</sub>) is applied in the permeate side to provide additional vapour pressure driving force for the filtration process. A satisfactory permeate flux between 2.9 and 4.2 LMH and high phenolic compounds retention in the feed side was achieved after the DCMD treatment using PTFE membranes.

In textile wastewater treatment, several studies related to the DCMD application have also been reported in the literature [93,94]. Mokhtar et al. [95] synthesized the nanocomposite membrane comprised of polyvinylidene fluoride (PVDF)-Cloisite 15A clay to be used in their DCMD system. The real textile effluent was directly fed to the membrane without any pre-treatment process. It was observed that the MD process could reject 75% of COD, 93.6% TDS and 83% of colour from the wastewater during 40-h operation. The results also indicated that MD could attain a comparable result with RO and NF treatment even though low pressure was utilized. In addition, lower energy was consumed during the process since the MD can use free energy provided by the hot textile effluent. Unfortunately, the membrane fouling due to surfactants in textile wastewater has led to initial permeate flux reduction ( $\sim$ 50%) during the long-term MD treatment process. The treatment of textile wastewater also is challenging since the presence of surfactant can cause membrane wetting. Garcia et al. [96] used the hydrophobic PTFE membrane coated with a hydrophilic layer in their pilot MD system to overcome the wetting issue in textile wastewater treatment. They found that the coating has enhanced the PTFE membrane's wetting resistance capability compared to that of the non-coated PTFE membrane. While the coated membrane performed well during this test, it could not survive in the membrane cleaning process involving sodium hydroxide.

In oil and gas wastewater treatment, MD's operating cost could be minimized by utilizing the waste heat source from the natural gas compressor station (NG CS). Lokare et al. [97] assessed the potential of DCMD to treat the real shale gas produced water by exploiting the NG CS exhaust stream. Their energy analysis showed that the produced water could be concentrated up to 30 wt% regardless of initial salinity by integrating the DCMD and waste heat from NG CS. Xu et al. [98] developed the polyoxadiazole hollow fibre membrane for the application of DCMD in the treatment of real produced water. A long-term experiment (100 hr) demonstrated that the DCMD system has attained the TDS rejection more than 99.5%. A stable water flux was recorded and it decreased as the module length was increased. Their DCMD unit also successfully achieved the low salt concentration in the permeate side (0.088 mg/L), which is lower than the China standard limit for discharge produced water. Besides DCMD, other MD configurations such as AGMD and VMD have also been used in the application of oil and gas wastewater treatment [99,100]. For instance, Alkhudhiri et al. [99]



Fig. 3. Illustration of air gap membrane distillation.

#### Table 4

Application of MD in saline wastewater treatment.

MD operation	Saline wastewater types/source	Wastewater/feed solution characteristics	Performance	Reference
$\label{eq:transform} \begin{array}{l} \text{Osmotic MD} \\ T_{feed} = 40 \ ^{\circ}\text{C}, \\ T_{permeate} = 20 \ ^{\circ}\text{C} \end{array}$	Crude olive mill wastewater (from Morocco region)	EC = 23 mS/cm COD = 160 g/L Total solids = 90 g/L Total phenolic content = 5.3 g/L	Initial permeate flux = 3.9 LMH Phenolic separation coefficient $\sim$ 99%	[35]
DCMD $T_{feed} = 90 \text{ °C},$ $T_{permeate} = 25 \text{ °C}$	Textile wastewater (from factory in Johor, Malaysia)	$\begin{split} EC &= 1294{-}1673 \ \mu\text{S/cm} \\ Turbidity &= 22{-}25\text{NTU} \\ TDS &= 774{-}996 \ mg/L \\ COD &= 405{-}477 \ mg/L \\ BOD_5 &= 11{-}15 \\ Colour &= 346{-}526 \ Pt/Co \end{split}$	Rejection efficiency: Colour = 95.3% EC = 93.7% Turbidity 93% TDS = 93.6% COD = 89.6% BOD <sub>5</sub> = 90.8%	[95]
DCMD $T_{feed} = 60 \circ C$ , $T_{permeate} = 45 \circ C$	Textile wastewater (from factory in Australia)	EC = 976 $\mu$ S/cm TDS = 605 mg/L Total nitrogen = 11 mg/L Total nitrate = 8 mg/L COD = 2830 mg/L	Rejection efficiency: EC = 99.5% TDS = 99.5% Total nitrogen = 97.8% COD = 93.4%	[96]
DCMD $T_{feed} = 70 \ ^{\circ}C,$ $T_{permeate} = 20 \ ^{\circ}C$	Produced water (from drilling locations in Saudi Arabia)	EC = 82  mS/cm $TDS = 84,000  mg/L$ $TOC = 41  mg/L$	Salt rejection >99.5%, EC of permeate = 195 $\mu$ S/cm	[98]
$\begin{array}{l} \text{AGMD} \\ \text{T}_{\text{feed}} = 4080 \ ^{\circ}\text{C}, \end{array}$	Produced water (from Arabian Gulf)	TDS = 187,440 ppm Hardness = 53,621 ppm Chloride = 119,437 ppm Sodium = 65,372 ppm Calcium = 14,161 ppm Magnesium = 2773 ppm	Permeate flux = 1.5 g/m <sup>2</sup> ·s Salt rejection >99%	[99]
VMD $T_{feed} = 6585~^\circ\text{C}$	Wastewater from natural gas field (Sichuan, China)	EC = 26,000 µS/cm TDS = 15,700 mg/L Chloride = 8732.6 mg/L COD = 1083 mg/L TOC = 307.5 mg/L	Water recovery = 88.6% Salt rejection = 99.8%	[100]

investigated the effect of operating parameters such as produced water flow rates and temperatures on permeate fluxes and rejection for three different PTFE membranes containing various pore sizes (0.2–1.0  $\mu$ m) in AGMD system. Increasing the feed temperature (40 to 80 °C) and feed flow rates has increased the flux. Meanwhile, the consumed energy was almost independent of the pore dimension of membrane.

Another potential MD application is to desalinate the brine solution discharged from large-scale desalination processes such as RO [101]. Andrés-Mañasa et al. [102] developed the vacuum-enhanced AGMD in a pilot-scale for the brine treatment. Two commercial spiral-wound modules containing different channel length (1.5 and 2.7 m) was used in their work. The vapour transport across the membrane was improved by the air suction from the gap, leading to MD performance enhancement. The vacuum effect resulted in maximum permeate productivity (8.7 LMH) using the shortest channel length (1.5 m). Their result suggested that the vacuum-AGMD system is promising for the treatment of highly concentrated feed solutions since it can attain better salt rejection factor (SRF) values (99.6%) than that of the RO process. Besides water recovery, MD and hybrid MD application to recover salt and valuable resource such as sodium sulphate from brine were also reported in the literature [103,104]. Despite the advantages of MD in treating saline wastewater, MD operation still faces challenges in terms of low permeate flux (in comparison to RO) due to the temperature polarization phenomenon [83]. Besides, membrane fouling and trapped air insides the membrane could further enhance the resistance, reducing the MD flux [99].

# 3.4. Electrodialysis

Electrodialysis (ED) is another promising membrane-based separation that has been applied in saline wastewater treatment. Basically, the ED structure comprises several cation exchange membranes (CEM) and anion-exchange membranes (AEM) in an alternating arrangement between an anode and cathode. The electrical potential drives the ions migration across the ion-exchange membrane (IEM). The capability of ED to separate monovalent ions enables its application in organic/ inorganic matters, heavy metals, and nutrients recovery [105]. Compared to RO, the ED process exhibits a higher water recovery ratio, lower operating cost, simpler operation, and longer membrane lifetime [105]. Nevertheless, ED could not remove viruses and bacteria, and its operation requires electrical safety [106,107]. Also, ED technology in wastewater treatment has not been explored much compared to the other established membrane processes [107].

Recently, several works related to ED application in wastewater treatment such as oil and gas effluent, leachate, textile, and tannery wastewater have been reported and summarized in Table 5. Earlier studies demonstrated that the ED system is applicable to treat highly saline wastewater such as produced water [108]. For instance, Peraki et al. [109] found that the ED treatment can reduce the TDS level in flow-back water (initial TDS ~60,000 mg/L) by about 27%. Various parameters such as test duration, the applied current and types of electrolyte were investigated in their work to assess how those parameters were affecting the desalination process. It was observed that the test duration is the most significant factor, followed by the applied current. Meanwhile, electrolyte types did not significantly affect the results. Previous work also reported that the ED process consumes energy comparable to that of the vapour compression desalination system for treating flowback and produced water containing 40,000–90,000 ppm TDS [108]. Thus, ED is promising for the highly saline water desalination in terms of energy and cost, but the fouling phenomenon under complex wastewater feed still need to be explored. Besides produced water treatment, the ED also is efficient in reducing the salinity level in wastewater from textile industries that commonly use a bulk amount of NaCl salt [26]. A recent work by Annamalai et al. [110] focused on the treatment of groundwater contaminated with textile effluent (initial TDS of 8397 mg/L) using ED setup. In their work, the contaminated groundwater was chemically pre-treated to reduce the solution's hardness before the ED process. Their analysis revealed that ED has successfully decreased the salinity level of the contaminated groundwater by removing 96% TDS, 90% chloride and 98% sulphate after a 12-h operation.

In addition to conventional ED, several configurations of ED have

#### Table 5

Application of ED in saline wastewater treatment.

Membrane	Saline wastewater types/source	Wastewater/feed solution characteristics	Performance	Reference
Lab-scale ED Electrolyte = $Na_2SO_4$ and $NaCl$ solution	Flow-back water (Marcellus wells, Pennsylvania)	TDS = 100,000  mg/L	-TDS reduction $\sim$ 27% after 7 h operation. -NaCl as an electrolyte presented a more steady and more consistent performance than Na <sub>2</sub> SO <sub>4</sub>	[109]
Lab scale ED Current density: 10 mA/cm <sup>2</sup> and 20 mA/cm <sup>2</sup>	Contaminated groundwater from Tirupur (Textile Valley), India	EC = 9.135 mS/cm TDS = 8397 mg/L Hardness = 4300 mg/L Ca <sup>2+</sup> = 2250 mg/L Mg <sup>2+</sup> = 2050 mg/L Chloride = 3828 mg/L Sulphate = 512.48 mg/L	Rejection efficiency: chloride (90%), sulphate (98%), and TDS (96%) after 12 h operation. -ED with 20 mA/cm reduced the EC to 0.795 mS/cm	[110]
BMED Voltage = 10, 15, 20, 25 V	Leachate (Oyaderi Landfill, Istanbul)	EC = 14.61 mS/cm COD = 7200 mg/L BOD <sub>5</sub> = 3740 mg/L Na = 9430 mg/L Cl = 4370 mg/L Ca = 12.5 mg/L Mg = 0.70 mg/L	The optimal voltage 25 V (a single membrane) produced effluent quality with: EC = 1.97  mS/cm COD = 1920  mg/L TKN = 224  mg/L $NH_3-N = 112 \text{ mg/L}$	[107]
Pilot scale BMED Electrode = Fe and aluminium	Tannery wastewater (Organized Tannery Industrial Region, Istanbul)	EC = 23 mS/cm COD = 2800 mg/L Chromium = 570 mg/L	Treated effluent quality: EC = 1.5  mS/cm COD = 364  mg/L Chromium = 0.0  mg/L	[15]

been proposed, including bipolar membrane electrodialysis (BMED) and electro-deionization (EDI) [107]. In the case of BMED, a combination of cation exchange membrane and low-voltage anion is adapted in the formation of bipolar membranes [111]. Unlike conventional ED, the BMED enables the separate removal of anions and cations from feed water. These removed ions can be subsequently coupled with OH- and H<sup>+</sup> ions to obtain alkaline and acidic solution [112]. In terms of landfill leachate treatment, the obtained alkaline and acidic solution after ED treatment can be further used for resource recovery rather than being subjected to disposal. In a study conducted by Ilhan et al. [107], the leachate was treated using BMED method to convert the catholyte and anolyte into alkaline and acidic solutions. Various operational conditions such as membrane set, electrical voltage and treatment time were investigated in their study. The optimized treatment conditions were observed in the case of 25 V electrical voltage with single membrane. The COD of the effluent was successfully reduced from 7200 mg/L to 1920 mg/L while the conductivity reduced to 1.97 mS/cm (~87% reduction) after ED treatment. Besides, the BMED was capable to reduce the effluent conductivity to 0.03 mS/cm after a long treatment time, but this approach was not applicable for large scale operation due to the increase in cost and reduction in removal efficiency.

Besides water recovery, the ED process can be coupled with other processes to remove contaminants such as heavy metals. For instance, Degles and Kurt [15] applied the ED treatment after the electrocoagulation process to remove the colour, Cr, NH<sub>3</sub>-N and COD from the tannery wastewater effluent. The BMED was implemented in the pilotscale ED treatment to assess the efficiency of the contaminant's removal. They found that the iron electrode application in ED can reduce the effluent conductivity from 23 to 1.5 mS/cm after 75 min operation. The ED-treated effluent also has achieved 87% removal efficiency of COD, while 100% rejection was recorded for Cr, NH<sub>3</sub>-N and colour from the tannery wastewater. The high pollutants removal using ED also indicated the pre-treatment of the wastewater is essential to increase ED process efficiency during tannery wastewater treatment. The treated effluent quality complied with that of effluent discharged standard under Turkish legislation, demonstrating the potential of ED in treating the dirtiest saline wastewater.

### 3.5. Membrane bioreactor

Membrane bioreactor (MBR) is a well-recognized promising wastewater treatment technology that involves the hybridization of membrane filtration with the biological sludge process [113]. Its robustness, good efficiency, ease of operation, low sludge production and small footprint characteristics have led to the widespread application of MBR in various wastewater treatment, including saline wastewater [114,115]. However, the salt constituents in saline wastewater could be a stress factor that destabilizes the microbial communities in MBR, reducing their metabolic activities and subsequently affecting their biomass kinetics to remove carbonaceous and nitrogenous compounds [116]. A typical MBR consists of a bioreactor packed with activated sludge and fitted with a membrane filtration system. MBR has two common configurations, namely side stream MBR and submerged MBR (SMBR) [117]. The SMBR design was proposed to overcome the high energy consumption issue of side stream MBR. Unlike side stream MBR, which placed the membrane outside the bioreactor (Fig. 4), SMBR design consists of the submerged membrane inside the bioreactor, enabling the effluent transfer with sludge retention. Generally, conventional MBR is applicable for treating wastewater with low salinity level (<10 g/L NaCl) since the activated sludges contain non-halophilic microorganisms that only can adapt to low salinity condition. Meanwhile, the use of modified/hybrid MBR systems and extremely halophilic microorganisms have been proposed to treat the medium (30–150 g/L NaCl) and high salinity (>150 g/L NaCl) wastewater [118].

Previously, several investigations have been conducted to assess the effect of salinity on MBR performance in saline wastewater treatment. For instance, Rodriguez-Sanchez et al. [119] demonstrated that the increase of salinity severely crippled the capability of the MBR process to remove  $\rm NH_4^+$  and TN (down from 27.7–85.24% to 24.46–38.55% for salinity increased from 4.5 mS/cm to 8.5 mS/cm), indicating the stress and disruption caused by salinity on the performance of MBR. Membrane fouling was also found to be exacerbated due to the irreversible cake deposition on the membrane surface, likely due to the worsening of sludge filterability and high organic cellular constituents (such as soluble microbial products) secreted by microorganisms in responding to salt shock [116]. Hence, the MBR process must be designed and operated properly, especially when the plant receives wastewater with varying degrees of salinity.

A similar observation has also been reported by Mannina et al. [120], where the influence of salinity and the presence of hydrocarbons on the MBR performance was investigated. The increase of salinity (12 g/L NaCl to 20 g/L NaCl) did not have an apparent impact on the COD reduction efficiency (~90%) of MBR during the pilot plant testing, though the biological COD reduction was moderately quenched (87% to



Fig. 4. Illustration of side stream MBR process.

64%). Also, the increase of salinity caused an inhibition effect on the nitrifier bacteria, reflected by the sharp decline in the nitrification efficiency from 83% to 33%. After the addition of hydrocarbon (mimicking the shipboard slops), the average sludge viscosity increased from 3.5 cP to 5 cP, which could be attributed to the deflocculation effect of the sludge flocs as indicated by the increase of soluble microbial products (2.7 mg EPS/g TSS to 13.6 mg EPS/g TSS). This exacerbated the membrane fouling issues as the formation of irreversible cake deposition on the membrane surface caused deterioration of the membrane performance. The findings from this study demonstrated the need to extend the start-up period (biomass acclimation) to prevent salinity shock that disrupted the biomass in MBR and the possibility of considering chemical addition to cope with sludge worsening and thus improve the membrane performance. It must be noted that the role of MBR is to remove carbonaceous and nitrogenous compounds instead of salt ions. Thus, the permeate of MBR should be further treated with other

#### technologies such as RO to remove the salt ions if it is to be reused.

In saline wastewater treatment, resource recovery, particularly energy in the form of biogas (methane), can also be achieved with the MBR process. MBR operated under anaerobic condition can attain two goals simultaneously, i.e., wastewater treatment (removal of carbonaceous and nitrogenous compounds) and biogas generation (anaerobic process). To address the adverse stress created by the salinity of certain wastewater on the biomass, the MBR can be acclimatized with hypersaline anaerobic seed sludge that has better adaptation to saline wastewater, as demonstrated by Umaiyakunjaram and Shanmugam [121]. The team showed that the anaerobic MBR acclimatized with hypersaline anaerobic seed sludge recorded high COD reduction efficiency (90%) and biogas yield (0.16 L/g  $COD_{removed}$ ) for raw tannery wastewater, indicating the minimum adverse impact of salinity on the biomass. Indeed, the biogas produced could turn the wastewater treatment into an energy-positive operation, as presented by Galib et al.

#### Table 6

Appl	ication	of	MBR	in	saline	wastewater	treatment
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MBR operation	Saline wastewater types/source	Wastewater/feed solution characteristics	Performance	Reference
Jet loop MBR	Olive mill wastewater (Balıkesir, Turkey)	EC = 10.67  mS/cm COD = 23,872  mg/L BOD = 18,876  mg/L Total phenol = 1610 mg/L	Treated effluent: average total phenol concentration = 59 mg/L COD removal 91–93% Total phenol removal 80–87%	[11]
Anoxic-aerobic MBR (pilot-scale)	Textile wastewater (woolen textile dyeing factory, Turkey)	$EC = 3680 \text{ ms/cm}$ $COD = 750 \text{ mg/L}$ $BOD_5 = 325 \text{ mg/L}$ $TOC = 260 \text{ mg/L}$ $Chloride = 89.84 \text{ mg/L}$ $Nitrate = 8 \text{ mg/L}$ $Sulphate = 394.6 \text{ mg/L}$	Removal efficiency of aromatic amine with: one aromatic ring >80%. Two aromatic rings<75%.	[123]
Anaerobic MBR	Bleach plant effluent (from paper/pulp industry, South Africa)	EC = 1196 mS/m Total COD = 1700 mg/L Soluble COD = 1330 mg/L Sulphate = 280 mg/L Chloride = 1700 mg/L Iron = 1.1 mg/L Manzanese = 6.3 mg/L	$\label{eq:states} \begin{array}{l} Flux = 6 \ LMH \\ Treated \ effluent: \ COD \ <\!\!45 \ mg/L \\ SS \ < \ 15 \ mg/L. \\ During \ 85 \ days \ of \ operation. \end{array}$	[124]
Submerged-anaerobic MBR	Meat processing wastewater (Conestoga Meat Packers, Breslau, Canada)	TSS = 1640 mg/L Volatile suspended solids (VSS) = 1460 mg/L Total COD = 4398 mg/L NH <sub>3</sub> -N = 77 mg/L $PO_4^3$ -P = 101 mg/L	COD removal 88–95% at organic loading rate (OLR) of 0.4–3.2 kg COD/m <sup>3</sup> ·d	[122]
Submerged anaerobic MBR (pilot scale)	Tannery wastewater (Tamil Nadu, India)	TSS 8400–12,600 mg/L VSS 5900–9760 mg/L Soluble COD = 7560 mg/L	Permeate flux 6.8 LMH Initial permeate TSS = 4140 mg/L Initial permeate VSS = 2210 mg/L COD removal was 90% (49th day)	[121]

[122], where the anaerobic MBR treating meat-processing wastewater produced a net energy benefit of  $0.16-1.82 \text{ kWh/m}^3$ .

The robustness of MBR in treating a wide range of wastewater has accelerated its adoption and application in various industries, as displayed in Table 6. Apart from focusing on the biomass, attention has also been paid to the membrane, especially membrane fouling issues and the further purification of treated water for reuse purpose. Having said that, to address the fouling issues and reclaim water for reuse or safe discharge, MBR has also been integrated with other treatment technologies. The next section will discuss the various integrated MBR process for wastewater treatment and reclamation.

### 3.6. Integrated treatment process

The treatment and handling of saline wastewater are far more challenging than the typical domestic wastewater. The similarity between saline wastewater and typical wastewater treatment is the need to remove typical pollutants found in effluents, such as suspended solids, dissolved ions, microorganism, persistent contaminants, and nutrients. However, the difference lies in the presence of a high concentration of salts in saline wastewater that requires an additional treatment process to remove it such that the treated water can be reused or safely discharged to the environment (especially inland sites). Given the complex composition of saline wastewater which contains various types of pollutants and different salinity level, the integrated treatment process has been proposed to enhance further the wastewater treatment efficiency over the stand-alone membrane process. Various combinations of membrane processes have been adapted in saline wastewater treatment as demonstrated in Table 7. More than a membrane system could be added in the treatment to tackle the drawbacks of certain technologies and further optimize the filtration process performance.

For example, the FO unit has been integrated with MD for the treatment of produced water [125,126], dairy wastewater [127,128] and high salinity landfill leachate [129]. A recent report by Zhou et al. [129] demonstrated that the hybrid FO-MD system performed better than single FO or MD unit in treating the hazardous landfill leachate. They found that the water transfer rate and stability of the FO-MD system can be maintained by supplying the feed solution (25,000 mg/L NaCl in feed) to the MD at temperature of 72.5  $^\circ$ C. The combination of FO and MD also attained the salt and TN rejection rates more than 96 and 98%, respectively, while the heavy metals (As, Sb and Hg) were completely removed from the treated leachate. In another study, the integration of FO and MBR has resulted in the development of the osmotic membrane bioreactor (OMBR) system to overcome the limitations of conventional MBR in terms of high maintenance cost [16]. The presence of FO in OMBR system reduces the fouling tendency and leads to water recovery with high quality, which requires less energy consumption as compared to traditional MBR [130]. Even so, the application of OMBR in treating real saline wastewater is still challenging due to the reverse salt transport phenomenon from the DS of FO to the biological reactor. For instance, Luján-Facundo et al. [130], who applied the OMBR in the tannery wastewater treatment, reported that this hybrid system is effective for COD removal (~80% efficiency). However, the water flux decreased over time due to salinity build-up in the biological reactor, which was contributed by the reverse salt mechanism. This drawback further leads to integrating the OMBR process with either MF or UF as proposed by other researchers [131,132] to mitigate salinity build-up in the biological reactor. Despite its promising performance due to microbial activity improvement, the integrated system's complexity remains one of the major concerns.

In other work, Song and Liu [133] combined the OMBR and MD process to treat real dairy wastewater. Unlike previous works [131,132], Song and Liu [133] proposed the use of salt-tolerant sludge (Bacillus) in their OMBR system to overcome the salt accumulation problem. The nutrient removal efficiency, water flux and sludge properties were investigated in the OMBR-MD process under 40 days of operation. It was

Table 7

Summary of integrated/hybrid process in saline wastewater treatment.

integrated/hybrid process	same wastewater types/source	кетагкs	Kererenco
FO-MD	Landfill leachate (hazardous waste landfill, Shanghai)	The rejection rates of salt >96%, TN and TOC > 98%. -Heavy metals (As, Sb and Hg) were	[129]
OMBR-MD	Dairy wastewater (dairy product industrial facility, China)	completely removed. -Contaminant's removal ~100% -The TN removal efficiency of the bioreactor was 40–79%. -Bacillus-OMBR	[133]
MBR-NF (pilot scale)	Textile wastewater (Textile mill, Tianjin, China)	membrane biofouling. -The MBR-NF effluent quality showed relatively stable trends and complied the water reuse criteria for textile industry. -TDS value of textile wastewater reduced from 3131 mg/L to 904 mg/L.	[136]
Activated carbon- RO-EDR (pilot- scale)	Petrochemical industry wastewater (Brazil)	-Chlorides and alkalinity removal efficiency >90% -The recovered water complied with standards for reuse in cooling towers. -Water recovery rate: 87 3%	[145]
UF-RO-multi-effect distillation (MED) process (pilot- scale)	Coal seam gas produced water (Gloucester Basin, Australia)	<ul> <li>Overall clean water recovery: 95%</li> <li>The recovered water could be blended with UF treated produced water for irrigation purpose.</li> <li>With the integration of MED, brine with sodium bicarbonate concentration up to 25.5 g/L was generated, which could be recovered as valuable mineral.</li> </ul>	[150]
Coagulation- sedimentation- dissolved air flotation-MBR- granular activated carbon	Tannery wastewater (Tannery plant, Vietnam)	MBR removal efficiency: -Dissolved organic matter: 81% -Total nitrogen: 36%	[151]
MBR-NF vs Fenton- MF-NF	Landfill leachate (Sanitary landfill, Brazil)	Fenton-MF is more efficient than MBR in terms of colour, COD, ammonia nitrogen and toxicity removal. -Final removal efficiency of MBR-NF system: colour ~100%, COD~88%, Ammonia 00%	[152]
NF-ED	Tannery wastewater (Tanning Industry, Brazil)	-The final product water (EC of 3.28–3.67 mS/cm) can be reused as process water for beamhouse operations. -With respect to	[153]

#### Table 7 (continued)

Membrane/ integrated/hybrid process	Saline wastewater types/source	Remarks	Reference
		chloride and COD parameters, the ED product water meets the requirements of process water, even for dyeing operations.	

found that the OMBR-MD hybrid system has successfully removed almost all contaminants (~100%) from the dairy wastewater owing to the help of salt-tolerant species and membrane capability. Besides FO, conventional MBR is often integrated with pressure-driven membranes such as RO and NF to treat other saline wastewaters, including textile and food industries effluents [134,135]. For example, a pilot-scale experiment comprised of MBR followed by an NF post-treatment system was investigated by Li et al. [136] to enhance the water recovery from textile wastewater. In the hybrid system, recirculation of NF concentrate to the MBR could promote higher water recovery, but such backflow influenced the contaminants removal efficiency of MBR. In another work, Falizi et al. [137] investigated the hybrid MBR-RO process for irrigation purpose. This hybrid process was proposed to overcome the limitation of MBR treatment, which could not effectively reduce saline wastewater's salinity, hindering the reuse feasibility of the MBR-treated water for irrigation purpose [138,139]. Water with high electrical conductivity would cause physiological drought that disrupts the plant's water intake ability and subsequently affects crop productivity. The findings from Falizi et al.'s study [137] demonstrated that the installation of RO as post-treatment for MBR effluent could effectively reduce the effluent salinity to irrigation standards (Fig. 5). Nonetheless, another issue arose from such an integrated MBR-RO process, where the RO permeate was found to cause severe infiltration (sodicity) problems in soil due to the excessive soil accumulation of sodium. Sodicity causes a decrease in the downward movement of water into and through the soil, affecting the water uptake of the plant's roots. To overcome the sodicity issue, the authors theoretically mixed the RO permeate with MBR effluent at a ratio of 2:1, tuning the quality of mixture water compliance with the irrigation standards. Hence, this integrated MBR-RO treatment process, with a proper mixing of effluent from each stage, managed to address the challenges encountered by each individual treatment process and produced treated water with meeting the quality for irrigation purpose.

One challenge for membrane filtration system is handling the

rejected stream known as retentate or concentrate. Merely disposing of the retentate is no longer feasible for those looking for sustainable management of wastewater. In this context, the retentate from leachate generated in an integrated MBR-NF membrane treatment system, which contains high organic content (specifically humic substances such as humic acid and fulvic acid), possesses the potential to be handled as a resource (organic fertilizer) rather than waste for disposal. However, the leachate retentate's direct application is not feasible due to its high concentration of salts and insufficient level of humic substances. To resolve these issues, Xu et al. [140] deployed a two-stage process of tight UF membranes to concentrate and desalinate the leachate retentate from MBR-NF for the production of organic fertilizer. The proposed treatment concept is illustrated in Fig. 6. The experimental findings show that the COD of leachate retentate from the MBR-NF system was concentrated from 2500-9500 ppm to 71,590 ppm, with 82.3% of the organics accounted as humic substance. On another note, the low rejection of divalent (21-62%) and monovalent (10-26%) ions allowed the separation of salts (permeate) from the humic substance (retentate). Consequently, the concentrated stream from the UF system could be used as water-soluble fertilizer containing humic substances after the addition of macroelements (such as urea, phosphorous, and potassium). Economic analysis postulated from the study indicates that the proposed treatment system is economically feasible compared to the evaporation process that has been industrially applied for the treatment of leachate retentate from the MBR-NF system in China. Considering the large volume of retentate leachate generated (1/6 to 1/8 of the volume of raw leachate) in integrated MBR-NF system and the increasing number of such integrated system for leachate treatment, the sustainable management of retentate, enabled by the integrated treatment process, can greatly benefit the industries.

Besides performance optimization and resource recovery, an integrated water treatment system can also be designed to achieve zeroliquid discharge (ZLD). In wastewater treatment, ZLD aims to remove all liquid discharge from a treatment process and provide clean water for reuse purpose [141]. The ZLD system has been proposed for preserving the environment, fulfilling the environmental regulations, supplementing water sources, and saving cost on wastewater discharge. Therefore, ZLD implementation may reduce waste, minimize possible water quality implications to ecosystems and manage the industrial wastewater more efficiently [142]. One of the techniques to achieve ZLD is integrating thermal-based technologies (e.g., brine concentrator and crystallizers and multi-stage flash distillation unit) with other membrane desalination units for the brine treatment [143]. However, ZLD is particularly challenging for high-salinity wastewater due to the high energetic cost of these thermal-based brine treatment. Thus, alternative membrane



Fig. 5. Integrated treatment system to produce water meeting irrigation standards (drawn with information taken from [137]).



Fig. 6. Integrated MBR-NF-UF membrane process for the recovery of humic substances from leachate effluent (source: redrawn from [140]).

based technologies have been proposed to treat the high-TDS brine, such as osmotically assisted RO (OARO), membrane crystallizer (MCr), FO, and ED-reversal (EDR) [143]. As an example, it was reported that OARA is capable of treating brine with a TDS range of 100,000-140,000 mg/L [144]. Recently, the ZLD concept was implemented in a pilot scaleintegrated system comprised of RO-EDR for real petrochemical wastewater treatment [145]. Since water is mainly consumed for operating the cooling towers in petrochemical industries, Vencke et al. [145] aimed to recover clean water from petrochemical wastewater for cooling tower reuse. The hybrid RO-EDR process has successfully attained more than 90% removal efficiency of chloride, generating clean water (87.3% recovery) that meet the water quality standard for reuse in cooling towers. In China, ZLD implementation is mandatory for coal chemical industries [146]. Fig. 7 shows a simplified ZLD process used in direct coal liquefaction plant in China [147]. The NF-RO integrated system was able to recover about 75% water for reuse. The NF-RO concentrate was sent to evaporator to recover distillate for reuse while the brine was crystallized, closing the ZLD loop. This highlights the role of NF/RO membrane system in water recovery and the importance of integrating it with other treatment processes for a complete removal of pollutants, water reclamation, and achieving ZLD target.

It shall be noted that the pre-treatment step is required in most membrane desalination unit to ensure the water treatment process's stability and alleviate the fouling effect. The addition of the pretreatment unit based on the membrane or non-membrane processes prior to any main membrane desalination unit can also be considered as a form of an integrated process [148,149]. However, further explanation on the pre-treatment step will be discussed in the next section under fouling mitigation strategies.

#### 4. Membrane fouling issue

Fouling remained to be one of the major issues in the treatment of saline wastewater involving membrane technologies. Theoretically, the fouling phenomenon occurs due to the deposition of undesirable matters (foulants) on the membrane's surface, which leads to flux reduction [45]. In some cases, the foulants can also clog the membrane pores, resulting in internal fouling [154]. The fouling can be categorised into inorganic fouling (scaling), organic fouling, and biofouling, depending on the types of the foulants [155]. Scaling is commonly occurred when the concentration of the inorganic ions in water surpasses the saturation level. This phenomenon consequently causes the inorganic ions to enter the nucleation step, resulting in crystal growth and deposition on membrane surface or pores [156]. Several factors could affect the fouling phenomenon, including the membrane properties, composition of the feed water and operating conditions such as pH and temperature [157,158]. Different types of foulants may be present in saline wastewater which lead to complex fouling formation. The biofouling associated with the microorganism adhesion on membrane surface is commonly exacerbated by the salinity of the wastewater. The saline



Fig. 7. Simplified ZLD process in direct coal liquefaction plant in China (source: redrawn and modified from [147]).

condition can induce the bacteria endogenous respiration and cell aggregation, producing more extracellular polymeric substances (EPS) via cell secretion [118]. The high concentration of EPS could further increase the wastewater's viscosity and cause irreversible cake layer deposition on the membrane surface, which is hard to be removed [115,159].

#### 4.1. Fouling mitigation strategies

Previously, various strategies have been proposed to minimize the fouling formation. Although a wide range of fouling mitigation strategies have been introduced, not many of them have been applied to treat real saline wastewater. In this section, the fouling mitigation strategies involving the real saline wastewater is discussed to provide an insight on the efficiency of these strategies when dealing with harsh and challenging wastewater.

# 4.1.1. Pre-treatment process

Installation of other treatment processes prior to the main membrane unit could serve as effective pre-treatment to remove most foulants in feed water before it reaches the membrane unit. The pre-treatment processes can come from a wide range of technologies deemed appropriate for the corresponding wastewater and can be in the form of a membrane or non-membrane processes. For membrane process as pretreatment, MF and UF are typically used due to their capability to remove larger suspended solids that could severely clog the tighter membrane, such as NF and RO [160,161]. For instance, Abdelkader et al. [162] introduced MF and UF before DCMD operation of saline dairy wastewater to minimize fouling. The pre-treatment reduced 17.0% of TOC, 18.5% of total inorganic carbon and 39.2% of total carbon. Hence, the membrane was not severely fouled by lipids in MD. The permeate flux declined slightly due to the precipitation of calcium and phosphorus but could be maintained to be stable. In another work, Streit et al. [153] studied NF as the pre-treatment of ED in treating leather processing wastewater. NF could remove 96% of COD but not more than 42% of ammonium nitrogen and less than 54% of salts (Na, Mg, Cl, SO4). A reduction in the limiting current was observed after 40 h as the anion membrane in ED was significantly fouled by the organic matter. In some cases, anti-scalant was added into wastewater after membrane pretreatment process. As an example, Duong et al. [86] introduced the anti-scalant into UF-treated produced water before entering RO-AGMD pilot setup. It was observed that the MD operation at low permeate flux (1.4 LMH) combined with anti-scalant effect was effective to control the scaling in their study. Nevertheless, the anti-scalant amount should be properly adjusted to avoid additional fouling formation due to negative impacts of overdosage.

Other non-membrane technologies can also be adopted as pretreatment when the membrane process might be unfit. For example, the coagulation process could remove the suspended solids by neutralizing the colloids charge. Chemical coagulation and electrocoagulation are two types of coagulation processes employed as pre-treatment unit [156]. Yin et al. [163] used poly aluminium chloride as the chemical coagulant in the coagulation process to pre-treat the real textile effluent. In their work, the pre-treatment system comprised of coagulation-sand filter-magnetic ion exchange has alleviated the RO fouling better than the coagulation-sand filter-UF pre-treatment system. This finding could be attributed to the more compact fouling layer formed on the RO membrane. The low molecular weight organic substances could penetrate the UF membrane and easily deposited in the RO membrane's valleys and fouled it. Hence, this demonstrated that proper integration of various processes with membrane core filtration could minimize the fouling issues. Besides application in the RO pre-treatment unit, the addition of polyaluminium chloride coagulant before MBR treatment was also found to efficiently alleviate the fouling in the bioreactor containing real textile wastewater [164]. Meanwhile, it was reported that electrocoagulation could remove the microorganisms and organic

matters, thus mitigating the biofouling and organic membrane phenomenon [156,165]. Recently, Valero et al. [166] pre-treated the almond processing wastewater using electrocoagulation and electrooxidation before ED operation. The membranes attained stable voltage values over 100 h of operating duration, signifying that the membranes were not significantly fouled. More than 94% of wastewater was recovered, with the final conductivity less than 1 mS/cm recorded. Despite its promising performance, electrocoagulation's high operation cost has limited its application as the pre-treatment unit.

Besides coagulation treatment, pre-treatment using activated carbon has also been applied in saline wastewater treatment. Even though powder or granular activated carbon can eliminate the dissolved organic carbon, this pre-treatment method cannot remove microorganisms and particles [156]. Thus, the combination of activated carbon and UF as pre-treatment steps can reduce the fouling effects, including the biofouling. Previously, powder activated carbon and UF have been employed as pre-treatment to RO for tannery wastewater reclamation [167]. The powder activated carbon reduced the COD and colour of the wastewater, while UF was responsible for reducing turbidity and salt density index. The pre-treatments combination ensured that the water fed to RO satisfied the water quality indices required for the RO operation.

In a leather processing plant, the photo-electrochemical method was adopted as the pre-treatment process before ED [168]. Rodrigues et al. [168] found that the photoelectrochemical method reduced COD (87.3%), BOD<sub>5</sub> (94.7%), ammoniacal nitrogen (>99%) and calcium (97.4%) effectively for ED. Recently, Pramanik et al. [169] removed pollutants from the biologically-treated effluent of a dairy farm using FO with ultraviolet/persulphate pre-treatment. Ultraviolet pre-treatment removed humic substances and building blocks significantly but could only reduce biopolymers when paired up with persulphate pre-treatment. Overall, various pre-treatment methods have been applied in real saline wastewater treatment. The combination of several pre-treatment units is commonly preferred over single pre-treatment process to achieve optimum wastewater treatment performance.

### 4.1.2. Physical/chemical cleaning

During the wastewater treatment processes, it is important to clean the membranes regularly to alleviate the fouling. Generally, the membrane cleaning procedures can be classified as physical and chemical cleaning. Water flushing is one of the physical cleaning methods frequently used in membrane operation. For example, Wu et al. [75] flushed the FO membrane with deionized water (200 mL/min) to control the fouling during the struvite recovery process from pre-treated agricultural wastewater. It was claimed that no apparent fouling formation was observed in their study, and more than 50% water recovery was achieved. A similar observation was reported by Carnevale et al. [170], who treated olive milling wastewater using DCMD and VMD. In their study, the membrane was cleaned using deionized water at 60 °C for 15 min. The flux recovery up to 92% was achieved while flux reduction was minimized after cleaning. In another work conducted by Gebreyohannes et al. [12], the osmotic backwash technique was applied in the FO process during the treatment of olive mill wastewater. They found that the osmotic backwash method's application was efficient to remove the fouling from cellulose triacetate membrane, which resulted in permeability recovery of up to 95%.

Unlike physical cleaning which is commonly applied to remove reversible fouling, chemical cleaning is required for irreversible fouling [155,171]. Various chemical agents have been used in the membrane cleaning during real saline wastewater treatment, including acid, base, chelating agent, and surfactant. Scoma et al. [172] applied the baseacidic cleaning using NaOH and HCl solution to control the fouling during ED treatment of olive mill wastewater. The ED removed nearly 30–35% of volatile fatty acid with no fouling formation reported in their work. Chloride rejection remained to be high (>95%) although competition between volatile fatty acid anions was observed. In another study, Zhang et al. [100] washed the fouled membrane in the VMD process using the acid-alkaline solution since permeate flux reduced 70% due to the scaling. They reported the reduction of conductivity, COD, TOC, and TDS beyond 94.8%, besides removing 81.5% of total oil and 86.8% of petroleum from saline wastewater in a natural gas field. Their findings also suggested that the performance of VMD can be recovered during long-term treatment of saline wastewater by shortening the cleaning intervals.

Coday et al. [173] compared the separation performance and fouling tendency of cellulose triacetate, thin-film composite and modified thinfilm composite membranes in FO of produced water treatment. The modified thin-film composite membrane rejected 95% of monovalent and divalent cations while achieving flux recovery from fouling using a chelating agent, namely ethylenediamine-tetraacetic acid (EDTA). The EDTA effectively removed cation-organic foulants through the chelating mechanism. Nevertheless, EDTA cleaning efficiency is highly dependent on the pH of the solution [156]. Zhao et al. [174] compared the efficiency of three different chemical agents, i.e., NaOH, EDTA and sodium dodecyl sulphate (SDS), in cleaning the hollow fibre FO membrane. Their findings revealed that the SDS cleaning is more efficient than EDTA and NaOH cleaning in achieving stable FO performance for produced water treatment. Although the permeate flux reduced around 22.9-22.8% due to fouling, SDS could be used to clean the membrane effectively within 15 min. In MBR operation, the chemical cleaning can be done either by in-situ or ex-situ cleaning. Unlike in-situ cleaning which is conducted inside the MBR, the ex-situ cleaning is performed by removing the membrane from bioreactor [175]. Although in-situ cleaning is commonly preferred over ex-situ cleaning, ex-situ cleaning is more beneficial when dealing with severe fouling [155,175]. In that case, increasing the chemical dosage in ex-situ cleaning can be performed without disturbing the bioreactor's microbial species. However, the combination of both in-situ and ex-situ is also feasible to maintain the MBR performance [176].

#### 4.1.3. Membrane modification

Membrane surface characteristics in terms of hydrophilicity and smoothness could influence the fouling phenomenon [156,177]. Fouling propensity is more obvious in the case of the membrane with a hydrophobic and rough surface. Thus, surface modification strategies have been proposed to enhance membrane hydrophilicity and smoothness. Chen et al. [178] developed a polyamide thin film composite membrane in FO to treat flow back water generated from shale gas drilling. Although the membrane could reject 95% of NaCl at a 3.5 LMH/bar permeability, severe fouling with flux reduction up to 70% was observed. Thus, the surface modification via poly(ethylene glycol) (PEG) grafting method was implemented to reduce the fouling propensity during FO operation. It was found that the PEG grafting has successfully alleviated the fouling by reducing the membrane roughness and lowering the membrane-foulants adhesion.

Besides the surface grafting technique, surface coating is another method that has been applied to tailor the membrane characteristics. Recently, Galiano et al. [179] adapted the polymerizable bicontinuous microemulsion (PBM) coating on the surface of polyethersulfone (PES) membrane to mitigate the fouling in MBR. The modified membrane's anti-fouling properties were challenged in a pilot plant of MBR, treating the real textile industrial effluent. Their findings revealed that the PBMcoated membrane was less fouled than that of the commercial PES membrane. The PBM-coating also has improved the critical flux, which prolonged the membrane life cycle since less frequent membrane cleaning is required. In another work, Mansour et al. [180] coated the surface of polyethylene membrane using graphene nanoplatelets (GNPs) to control the fouling in pilot-scale DCMD operation for the treatment of RO brine. The GNPs coating on the membrane (0.16 wt% loading) could reduce the membrane fouling due to salts, cleaning agent and antiscalants as much as 78%. Without the coating, the permeate flux reduced nearly 70% in a 77-h operation. Zinadini et al. [181] used

another modification method by blending the hydrophilic graphene oxide (GO) nanosheet with PES dope solution during membrane synthesis to improve the antifouling properties of the UF membrane. The modified membrane was then applied in MBR, which treated the dairy wastewater. The SEM analysis demonstrated that the PES/GO membrane possessed less biofilm formation since the presence of GO induced the electrostatic repulsion between the membrane surface and microorganisms in dairy wastewater.

In the MD process, it has been reported that the omniphobic membrane surface could also help to mitigate the fouling phenomenon. For instance, Woo et al. [182] fabricated omniphobic PVDF membrane for the AGMD process to concentrate brine from produced water. The omniphobic surface was constructed from a highly hierarchical and fluorinated layer. The system achieved a permeate flux of 11.22 LMH without significant flux reduction even when surfactant was added into the feed. The authors suggested that the omniphobic membrane surface not only possessed better anti-wetting property, but its negatively charged surface prevent the adsorption of organic foulants. In another work, Du et al. [183] compared three different membranes, namely hydrophobic PVDF, omniphobic PVDF, and PVDF coated with a hydrophilic layer (PVA coating) in MD operation of shale oil and produced water. Unlike Woo et al.'s findings [182], they found that the composite PVDF with hydrophilic coating indicated better antifouling properties than omniphobic and hydrophobic PVDF membrane in a single treatment. However, the omniphobic PVDF membrane has demonstrated a better reusability than the neat PVDF and composite PVDF membrane after several treatment cycles. Because of the omniphobicity, the feedwater intrusion was delayed, limiting membrane scaling and fouling to a shallow depth near the membrane surface. Based on their finding, it can be suggested that the membrane modification study should not only focus on the antifouling/antiwetting properties, but also need to evaluate other aspects including the reusability for long term operation.

## 4.1.4. Other methods

Innovation in membrane system configuration also has been proposed to reduce the fouling propensity in saline wastewater treatment. For example, Tamersit et al. [17] installed a UF membrane on the anionic membrane in ED during leather-processing wastewater treatment. The UF membrane allowed the permeation of low molecular weight anions such as sulphide, NaCl and amino acids but prevented protein and peptide fouling on the anionic membrane. The modified ED system removed 56% of conductivity, 62.5% of calcium, 72.3% of sulphide, 67% of chlorides and 10.4% of COD. Multiple steps of pretreatment were even introduced. The modification of ED setup was further proposed by Hayes and Severin [184] to mitigate the fouling due to high calcium content in the wastewater. In that study, they investigated the fouling of ED of flowback water generated from shale gas hydro facture containing 4000 mg/L of calcium and 30,000 mg/L of NaCl. The high calcium content caused severe fouling on electrode cell. They suggested clean-in-place by adding HCl into the electrolyte and using a monovalent-selective membrane to reduce up to 70% of calcium flux. In another work, Severin and Hayes [185] used the single cathode chamber boundary membrane, which restricted the permeation of calcium and barium during the operation of ED to treat field water generated from hydraulic fracture. They reversed anode and cathode periodically to remove calcium and barium fouled on membranes successfully but not ferric hydroxide precipitated within the ED stack. The ion flux increased with this clean-in-place strategy up to 45%.

Meanwhile, in MBR system, the air sparging configuration has been implemented to provide aeration that can minimize the concentration polarization as well as membrane fouling [175]. Gao et al. [186] used biogas as the source of sparging gas in the anaerobic MBR to alleviate membrane fouling during the pulping wastewater treatment. The increase in biogas sparging rate was found to mitigate the accumulation and deposition of sludge on/in membrane module and thus enhanced the membrane flux and stability of long-term operation in treating pulping wastewater. Chaiprapat et al. [187] combined the biogas sparging with activated carbon scrubbing and liquid circulation to further improve the fouling control in the anaerobic MBR system, treating the seafood processing wastewater. It was observed that the MBR configuration equipped with biogas circulation resulted in the thinnest cake formation on membrane surface after 18 days operation as compared to that of without biogas sparging (1–11 days operation). The fouling due to the organic and inorganic foulants (phosphate and struvite) was successfully reduced by 90% at 8 h hydraulic retention time. This finding is promising for MBR development since such strategy will require less cleaning frequency and thus, further reduce the maintenance cost.

#### 5. Future challenges and outlook

Despite the encouraging performance of membrane technologies, including integrated membrane process in the saline wastewater treatment, several issues and challenges have been identified in developing membrane technologies that restrict their implementation in real industrial sectors.

#### 5.1. Membrane fouling

Various membrane modification methods, including coating, grafting, incorporation of inorganic particles, have been reported to successfully control or delay the fouling phenomenon [178,181]. Nevertheless, the efficiency of these strategies was commonly evaluated under a short period test. The interaction between foulants is more dominant in determining the membrane performance under a long period, regardless of membrane type [20]. Besides membrane modification, the fouling can also be reduced using a membrane cleaning procedure. However, its influence on overall treatment cost remains questionable. In terms of MBR application in saline wastewater treatment, the installation of pre-treatment process is useful to avoid the salinity shocks of microorganisms, but this will eventually increase the overall process and treatment expense.

# 5.2. Lack of pilot-scale studies

It shall be noted that most of the membrane performance in the integrated saline wastewater treatment process were assessed under labscale experiments. Till today, limited data based on pilot-scale integrated membrane treatment is available, delaying the application of these membrane technologies in the industrial saline wastewater field. Even though recent reports on the pilot study of several saline wastewater treatments have displayed promising performance in terms of water recovery [86,145,188,189], the assessment of the overall cost for the water treatment is unavailable.

#### 5.3. Economic aspect

Since the ZLD approach has been promoted in saline wastewater treatment, the wastewater treatment's capacity and scale will continue to increase. The complexity of the saline wastewater nature requires the integration of several membrane treatments such as pre-treatment and post-treatment step to enhance the water recovery and achieve the ZLD target. In addition, if the integrated membrane process consists of a FO unit, the cost of installation of the DS regeneration unit shall be considered. As an example, it was reported that the FO unit with thermal regeneration step during produced water treatment might consume the energy of 25–150 kWh/m<sup>3</sup> [190], which is higher than that of the RO process (energy used  $\sim$ 4–16 kWh/m<sup>3</sup>) [20,190,191]. Although it has been reported that the performance of the integrated membrane process for treating various saline wastewater sources is promising, the expenditure analysis is rarely provided.

The issues mentioned above need to be addressed in future works to

achieve the market viability of membrane technologies in saline wastewater treatment. In terms of membrane fouling, the efficiency of the fouling mitigation strategies on membrane performance needs to be assessed under a long-term period, considering the complex nature of saline wastewater. Investigation on new materials in developing membrane with good permeability and antifouling feature is still required to explore membrane technologies' potential. The application of halophilic (salt-tolerant) microorganisms in MBR serves as an alternative approach to avoid the installation of a pre-treatment unit which is commonly required to avoid the salinity shock in MBR. Furthermore, the MBR salt tolerance upper limit could be further enhanced up to 150 g/L by adopting the halophilic inoculum [118]. To further explore its potential in MBR development, further investigation focusing on microbial perspective such as the halophilic species' metabolic shift upon salinity change can be conducted. In terms of the economic aspect, the utilization of waste heat can be implemented to reduce the operating cost of saline wastewater treatment. For instance, it was reported that the desalination cost of integrated FO-MD system in dairy wastewater treatment was reduced from \$17.33/m<sup>3</sup> to \$11.25/m<sup>3</sup> after utilizing the waste heat to operate the MD [128]. In addition, the operating cost could also be reduced if the DS which does not require regeneration is adopted in the FO unit. Finally, more studies investigating the membrane performance on the pilot scale is required in future work to further convince the industry to implement the membrane technologies in their saline wastewater treatment plant.

#### 6. Conclusion

In summary, the application and performance of several membrane technologies in treating saline wastewater from various sources have been highlighted in this study. Different membrane processes can be integrated into a wastewater treatment system or integrated with other technologies to boost the water treatment efficiency, considering the challenging properties of saline wastewater. Numerous combinations of membrane technologies in the integrated process have been studied with various targets such as fouling mitigation, process enhancement, resource recovery, draw solution regeneration and zero liquid discharge. To make membrane processes an economically attractive and viable alternative, more areas in membrane technology development must be further explored and improved in future works, especially in the context of economic analysis, performance optimization, large scale process and sustainability.

# Declaration of competing interest

The authors have no affiliation with any organization with a direct or indirect financial interest in the subject matter discussed in the manuscript

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