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Reject brine management: Denitrification and zero liquid discharge (ZLD)—Current status, challenges and future prospects

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ABSTRACT

Water is at the core of sustainable development. Moreover, it is essential for social, economic, and environmental well-being. However, water resource availability has been significantly threatened in the 21st century. In general, freshwater supplies are being depleted by natural and anthropogenic activities, such as rapid population growth, industrialization, and intensive agriculture. In addition, one of the main causes of water resource shortages is water body contamination. Nitrate pollution is considered one of the most pressing global environmental problems, both in surface and groundwater. The literature reveals that numerous nitrogen removal processes have been developed and proposed. To overcome this problem, desalination is a robust and mature technique for obtaining fresh water from saltwater, and is considered an efficient and reliable process. However, there is growing concern about the adverse environmental impacts generated by brine where concentrated rejection by desalination results in high salinity together with chemical residues. To solve this problem, a zero liquid discharge (ZLD) strategy has been proposed in the specific literature. Furthermore, ZLD can be used to treat and recover valuable resources. This study analyzes and discusses the current status of brine treatment technologies targeting ZLD, highlighting their advantages and disadvantages. Technologies based on membranes and thermal energy were also analyzed, and their performance and operating costs were compared. Finally, the different denitrification processes are listed. This ZLD solution is currently considered an essential and compulsory treatment in reject brine to remove nitrate that, because of high concentrations in the environment, is one of the most widespread global contaminants.

1. Introduction

Water is at the core of sustainable development. It is also critical for socioeconomic development, healthy ecosystems, energy, and food production. In the first decades of the 21st century, the availability and sustainability of water resources faced remarkable threats (Vörösmarty et al., 2018). In general, water resources are affected by climatic evolution and are characterized by a rise in ambient average temperatures. This results in sharp declines in net water reserves, as well as degradation and loss of quality (Cramer et al., 2018). Furthermore, the increase in global population is causing an increase in water demand. New solutions for water security are necessary, and it is estimated that approximately four billion people (60% of world population) live in regions that experience almost permanent water stress. Currently, one in four large cities is already facing water stress, and the demand for water is projected to increase by 55% in 2050 (Liyanaarachchi et al., 2014; Lorite et al., 2018). In addition, water stress is aggravated by pollution; between 80% and 90% of all wastewater in developed countries is discharged directly into surface water bodies, posing serious risks to human health (Bond, 2018; Abascal et al., 2022). Therefore, global freshwater scarcity is a serious humanitarian issue that must be addressed. In 2015, the United Nations General Assembly established Sustainable Development Goals (SDGs). The Sixth Goal relates to water, ’Ensure the availability and sustainable management of water and sanitation for all’. The goals cover all aspects of both the water cycle and sanitation systems, and their achievements are designed to contribute

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to progress on a variety of other SDGs, especially health, education, the economy, and the environment (UNICEF et al., 2018).

Pollution of groundwater and surface water resources is a pressing environmental problem worldwide. Nitrate is the most widespread global pollutant (Li et al., 2021). Nitrogen deposition on land has doubled since 1920 because of anthropogenic activities (Ward et al., 2018). The main source of pollution is intensive agriculture because of excessive application of inorganic nitrogenous fertilizers and manure. Wastewater treatment and oxidation of nitrogenous waste products in human and animal excrement, including septic tanks, is also a critical global problem (Shrimali and Singh, 2001; Abascal et al., 2022). When nitrate concentration exceeds a certain limit, groundwater is unsafe for drinking. In surface waters, high concentrations of such nutrients can cause eutrophication of aquatic ecosystems, proliferation of algae, and death of fish in water bodies (Kapoor and Viraraghavan, 1997). To satisfy water demand, both for human consumption and for economic activities (mainly agriculture and industry), different technologies have been developed with the aim of removing the major pollutants from water. In the last decade, numerous nitrogen removal processes were developed and optimized. In addition, the exploitation of water from new sources, such as aquifers with saline groundwater, is beginning (Panagopoulos et al., 2019; Panagopoulos and Haralambous, 2020b). To access this water, desalination is perceived as a feasible and viable solution to address water scarcity. In desalination, the feed water is divided into two streams, the permeate stream (freshwater) and concentrate stream (reject brine), typically discharged to the sea. Brine, except for its high salinity, may contain dangerous pretreatment chemicals and organics, such as nitrate and heavy metals. Uncontrolled and prolonged discharge exacerbates the problem of nitrate contamination of water bodies and endangers the survival of aquatic ecosystems (Yaqub and Lee, 2019; Cipolletta et al., 2021a; Sahu, 2020).

The literature indicates that reject brine and the treatment of nitrate-contaminated water has been managed separately. Regarding reject brine management, the latest studies are focused on the development of technology to achieve a zero liquid discharge (ZLD) or close-to-ZLD, with the intention of overcoming environmental issues linked to brine disposal. ZLD refers to a treatment process in which desalination plants discharge no liquid waste into surface waters and obtain high-quality freshwater (Ahirrao, 2014). Many reviews focusing on different technologies for achieving ZLD have been reported in the literature (Table 1). Various contributions presented state-of-the-art technologies for treating waste brine generated by desalination plants. Other studies have focused on the use of membrane-based technology (Yadav et al., 2022; Panagopoulos and Haralambous, 2020b; Yaqub and Lee, 2019; Onishi et al., 2018) to pre-concentrate brine, achieve water recovery, and minimize the volume of brine. ZLD can also be achieved through thermal-based technologies (Panagopoulos and Haralambous, 2020b; Yaqub and Lee, 2019; Ullah and Rasul, 2019), where evaporation and crystallization processes occur, resulting in water recovery, reduction in the volume of the brine and production of a solid by-product. Innovative hybrid approaches that combine two or more different methods are the currently emerging technologies. These recent solutions offer a higher treatment efficiency, reaching high water recoveries of 90% to 98% (Cipolletta et al., 2021a; Sahu, 2020). The number of studies focusing on the importance of treating surface water and groundwater to remove nitrates from water bodies has increased considerably because of the ubiquity of nitrate contamination. Hundreds of sites have been cataloged as possible sources of nitrate pollution (Abascal et al., 2022). Biological and physicochemical treatments have been used to treat these contaminants. The physicochemical treatments have been widely described by Rezvani et al. (2019). Some researchers have highlighted the importance of electrochemical processes such as electrodeposition, electrocoagulation, and electrodialysis (Shahedi et al., 2020; Xu et al., 2018), while other researchers have used adsorbents to remove nitrates, such as clays (Lazaratou et al., 2020) and nanomaterials (Tyagi et al., 2018). Biological treatments for nitrate removal have also been described, based on the metabolic pathways of the microorganisms involved in the process, distinguishing between heterotrophic and autotrophic processes (Huno et al., 2018; Ashok and Hait, 2015). One of the most widely used systems for the natural removal of nitrates are constructed wetlands (Zhuang et al., 2019). In the last decades, alternatives to biological nitrate removal have appeared. They are presented as a new technological platform for sustainable in situ nitrate removal. These are the so-called microbial electrochemical technologies (METs) which, through bioelectrobioremediation, promote the development of electroactive bacteria capable of using an extracellular electron donor for nitrate reduction (Sevda et al., 2018; Ghafari et al., 2008a). Additionally, cost-effective and energy-efficient treatments are required to completely treat the reject brine, with the aim of minimizing the discharge of liquids into the environment to valorize the products generated during desalination, and to completely remove contaminants such as nitrate. Many previous reviews, summarized in Table 1, proposed different technologies to separately address each of the goals listed above.

To the best of our knowledge, there is a lack of reviews that include a comprehensive treatment of reject brine produced by seawater desalination plants. This brine has a high content of both dissolved salts and nitrates; therefore, to avoid pollution problems, all contaminants and nutrients must be removed before they are discharged into the environment. To carry out this integral treatment of the brine, ZLD and denitrification technologies must be used. It is important to revise and compare such technologies, so they are correctly used and combine the most appropriate and efficient methods in each case, taking into account the physical and chemical properties of the brine, the volume generated, and available resources. This study aims to analyze the most relevant technologies used to treat saline effluents by discussing an updated review that encompasses both the technologies to achieve ZLD and the main nitrate removal processes. Additionally, different pathways for the transformation of saline waste streams into valuable products are proposed and described in detail.

The rest of the paper is organized as follows: Section 2 describes the environmental impacts of desalination processes; Section 3 describes the state-of-the-art brine treatment and introduces the ZLD approach; the technologies for nitrate removal from brine rejection are discussed in Section 4; the possible recovery of waste is analyzed in Section 5; and Section 6 provides the main conclusions.

2. Environmental impacts of desalination processes

The seawater desalination process involves the separation of saline seawater into two streams, a freshwater stream and a concentrated brine stream (Khawaji et al., 2008). The concentration of total dissolved solids (TDS) obtained in the fresh water produced, called permeate, depends on the corresponding end use. For domestic, agricultural, or industrial use, freshwater must contain less than 500 mg/L TDS (Rosborg and Kozisek, 2019). For other industrial applications such as pharmaceuticals, chemicals, and sanitation, the purity of the water must be even higher, i.e., less than 20 mg/L TDS (Patnaik, 2003; Ohannesian and Streeter, 2001). In addition, a by-product called the concentrate or reject brine is produced. The liquid stream contains the following: all TDS of the feed water with a high TDS concentration. Within these TDS approaches, different salts are used as well as nutrients such as nitrate, pretreatment chemicals, and microbial contaminants. The volume of reject brine produced in a desalination plant commonly depends on both the efficiency of the process and the total concentration of dissolved solids in the feed water (El-Naas, 2011). Owing to the composition of the reject brine and considering the significant quantities currently produced, suitable treatment processes and environmentally friendly disposal methods are necessary. The main environmental impacts described in the literature related to the discharge of brine are as follows: (i) negative effects on the quantity and quality of natural resources, including soil, air, and water; (ii) deterioration of aquatic ecosystems;

Table 1
Literature review of nitrate removal and zero liquid discharge (ZLD) technologies.

Reference	Main objective	Category of strategy	Year
Abascal et al. (2022)	Nitrate removal in groundwater	Biological and physicochemical processes	2022
Eltaweil et al. (2021)	Nitrate removal	Chitosan-based adsorbents	2021
Zhang et al. (2021)	Converting nitrogen into bioenergy	Microalgae-bacteria consortia	2021
Shahedi et al. (2020)	Nitrate removal in industrial wastewater	Electrocoagulation processes	2020
Wu et al. (2018)	Nitrate removal in wastewater	H2-based membrane biofilm reactor	2018
Lazaratou et al. (2020)	Nitrate removal	Clays absorbents	2020
Rezvani et al. (2019)	Nitrate removal from drinking water	Physicochemical and biological technologies	2019
Zhuang et al. (2019)	Nitrate removal in urban wastewater	Wetland systems	2019
Tyagi et al. (2018)	Nitrate removal from aqueous environment	Nanotechnology	2018
Sevda et al. (2018)	Nitrate removal	Bioelectrochemical technologies	2018
Xu et al. (2018)	Nitrate removal in industrial wastewater	Electrochemical systems	2018
Huno et al. (2018)	Nitrate removal from groundwater	Natural and engineered processes	2018
Ghafari et al. (2008a)	Nitrate removal from water and wastewater	Bioelectrochemical technologies	2008
Yadav et al. (2022)	ZLD	Membrane distillation crystallization technology	2022
Cipolletta et al. (2021a)	ZLD	Hybrid treatment technologies	2021
Sahu (2020)	ZLD	Innovative hybrid processes	2021
Panagopoulos and Haralambous (2020b)	ZLD	Pretreatment and treatment technologies	2020
Yaqub and Lee (2019)	ZLD	Thermal and membrane-based processes	2019
Panagopoulos et al. (2019)	ZLD	Desalination brine disposal methods	2019
Ullah and Rasul (2019)	ZLD	Solar thermal technologies	2018
Onishi et al. (2018)	ZLD	Thermal and membrane applications	2018

(iii) intensive use of chemicals and increased nutrients (nitrogen); and, (iv) impact on the aquifer (Höpner and Windelberg, 1997; Missimer and Maliva, 2018; Sadhwani et al., 2005a; Einav et al., 2003).

The reject brine discharge from a desalination plant is, on average, 50% of the influent flow, which is 1.5 times the saline concentration of the seawater. The negative effect of reject brine discharge on the marine environment depends on the hydrogeological characteristics of the area (e.g., depth, currents, tides, waves) (Sadhwani et al., 2005b). The different species of algae and marine flora are most negatively affected by brine discharge with high salt concentration; the most affected seagrass species are Posidonia oceanica and Cymodocea nodosa. When exposed to TDS concentrations above 45 mg/L, both species have a mortality rate higher than 50% of the population (Fernández-Torquemada et al., 2005; Garrote-Moreno et al., 2014). These grass prairies play a fundamental role in the marine ecosystem because they fi the sand deposits on the seabed. They also allow the development of algae association communities, and are a habitat for different fish and invertebrate communities. In addition, recent studies have shown that hypersalinity might affect benthic bacteria with a consequent influence on the diversity and abundance of these communities (Frank et al., 2017, 2019). In addition to a high concentration of salts, the reject brine contains chemical products used in the pretreatment phase of the desalination plant, including descaling reagents. The main chemical products used are sodium hypochlorite (NaOCl), ferric chloride ($FeCl_3$), sulfuric or hydrochloric acid, and sodium bisulfite ($NaHSO_3$) (Lattemann and Höpner, 2008). Some of these agents are directly toxic and lethal to marine fauna, and others can modify the local pH in the discharge area, affecting the growth and development of different species (Sadhwani et al., 2005b).

The discharge of these compounds (nitrates and phosphorus) into water bodies without treatment causes serious problems for both human health and the environment. Although both nutrients are required by microorganisms for their physiological processes, if their concentrations exceed certain limits, they become negative contaminants. The most important human diseases related to increased nitrate concentrations in water are methemoglobinemia and gastric cancer (Addiscott and Benjamin, 2004). Their impact on marine ecosystems is equally significant. As a consequence of the increase in nutrients, the growth of aquatic plants is favored, creating a negative effect on water quality by accelerating the deposition of dead algae, resulting in bad odor and discoloration, a process known as eutrophication. Dead macrophytes and phytoplankton settle on the seabed and, subsequently, there is an increase in microbial growth that uses dissolved oxygen to degrade settled organic matter. The dissolved oxygen concentration can plummet, causing the death of plants and aerobic organisms, and the suffocation of fish and other aquatic organisms (Liikanen and Martikainen, 2003; Tammi et al., 2003; Akpor and Muchie, 2011). This not only has a negative impact on the environment and marine biodiversity but also affects the economy, particularly tourism and fisheries (Singh, 2016). Therefore, it is important to be aware of the problem of contamination of marine waters to take adequate remedial measures.

3. Nitrate removal from brine rejection

Nitrate is the most common contaminant globally (Craswell et al., 2021), becoming a serious environmental problem, both in terrestrial ecosystems and in surface and groundwater ecosystems (Rivett et al., 2008). Recently, the nitrogen species content in groundwater has markedly increased due to anthropogenic activities, such as applying animal manure and intensive fertilizers on agricultural land (Martinez et al., 2009; Luo et al., 2002). This nitrate contamination, both in surface and underground water resources, leads to major environmental problems, such as algae blooms, eutrophication, and the death of fish in marine water bodies (Liikanen and Martikainen, 2003; Tammi et al., 2003; Akpor and Muchie, 2011). Intensive agriculture places increasing demands for more irrigation water. This has caused greater exploitation of fresh groundwater, especially in semi-arid regions with scarce surface water resources, such as the Mediterranean region (Adopted, 2014). However, the quality of water in these aquifers is compromised by high salinity which, according to the Intergovernmental Panel on Climate Change forecasting, is also expected to increase (Hoegh-Guldberg et al., 2018).

When withdrawals are made from aquifers with saline groundwater, the water must be desalinated to obtain the necessary quality to satisfy the demands of irrigated agriculture and tourism (Palomar and Losada, 2010). Desalination generates a highly saline concentrate (reject brine) with high nitrate content. This residue must be treated to eliminate the nitrates, a process similar to that of nitrogen removal in wastewater treatment plants (WWTP). This is an essential step because of the damage it causes to water bodies and aquatic organisms; additionally, the Nitrate Directive (91/676/EEC) limits the amount of nitrogen that can be discharged into the environment (Bosko et al., 2014; Beliavski et al., 2010). Both chemical and biological technologies are used to remove nitrate from contaminated water. These processes have been studied and developed to achieve the maximum efficiency for each type of water body, depending on its physical and chemical characteristics.

3.1. *Chemical denitrification process*

Chemical denitrification is a process through which nitrate is reduced using metals such as iron and aluminum. Other metals, such as copper and palladium, can also be used (Kapoor and Viraraghavan, 1997; Shrimali and Singh, 2001). However, chemical denitrification methods are difficult to operate at low pressure and temperature (Luk and Au-Yeung, 2002). During the chemical denitrification process, metal is oxidized and electrons are transferred from the donor metal to the nitrate, which is then reduced to nitrogen gas. If partial denitrification occurs, ammonium appears as an intermediate metabolite (Hao et al., 2005): $NO_3 \rightarrow NO_2 \rightarrow NO \rightarrow N_2O \rightarrow N_2$; $NO_3 \rightarrow NO_2 \rightarrow NH_4$. This technology has two major drawbacks to scaling up and commercialization, (i) the limitation of the reaction rate due to differences in large particles, and (ii) the impossibility of optimizing the catalysts so they do not produce ammonium (Kumar and Chakraborty, 2006) because the presence of ammonia in water streams can lead to human health problems (Shrimali and Singh, 2001). Based on these facts, it can be concluded that a large-scale chemical denitrifier system has not yet been developed, and alternative, more efficient, and controlled processes (i.e., biological processes) should be tested to eliminate nitrate from contaminated water bodies, such as brine.

3.2. *Biological denitrification process*

For many years, activated sludge technology has been used to remove nitrogen from wastewater. Nitrogen removal comprises two biochemical steps, nitrification and denitrification. The nitrifier process consists of the aerobic oxidation of ammonium to nitrite and nitrite to nitrate by ammonia-oxidizing bacteria (AOB) and nitrite-oxidizing bacteria (NOB). The second step is denitrification, an anoxic process in which nitrite and nitrate are reduced to gaseous nitrogen oxides by facultative anaerobic heterotrophic denitrifying bacteria (DNB) (Gonzalez-Martinez et al., 2018; Wang et al., 2017; Zhu et al., 2019).

In the denitrification process, denitrifying bacteria use organic matter as an electron donor and nitrate as an electron acceptor. Nitrate is reduced to nitrogen gas under anaerobic conditions (Knowles, 1982; Zumft, 1997; Torrentó et al., 2010). Denitrifying microorganisms are ubiquitous in nature, whether in surface water, groundwater, or soil (Pfenning and McMahon, 1997; Vogel et al., 1981; DeSimone and Howes, 1998; Smith and Duff, 1988; Peterson et al., 2013; Mohamed et al., 2003). Despite this, denitrification in natural systems is a slow process. The term solid-phase denitrification has been proposed in recent years because solid substrates are used as a constant source of carbon to provide a surface for the development of microbial biofilm. A solid-phase denitrification process can be achieved by heterotrophic and autotrophic methods if microorganisms obtain energy from organic or inorganic sources, respectively. Both processes are described as follows.

- Solid-phase heterotrophic denitrification. Heterotrophic bacteria obtain their energy from organic carbon previously synthesized by autotrophic organisms, such as woodchips and methanol, or the remains of other heterotrophic microorganisms, such as dead and lysed cells and decaying matter (Wang and Chu, 2016). Most of the organic matter is consumed in aerobic regions, where oxygen is available to act as an electron acceptor. Aerobic heterotrophic microorganisms oxidize organic matter in these regions, leaving a limited amount of carbon for denitrifying microorganisms in the anoxic zones. Therefore, there is a need for an external carbon (C) source for heterotrophic denitrifying organisms (Zhong et al., 2020). The Wood chips, sawdust, and wheat straw are the most widely used external C sources. Of all the organic materials whose denitrification potential is being verified, wood chips are the most used (Schipper and Vojvodić-Vuković, 2001; Schipper et al., 2001) because of their easy availability, high carbon/nitrogen ratio, and low cost (Robertson and Merkley, 2009; Schipper et al., 2010). A maximum removal efficiency of 99.75% was achieved with wood chips at a hydraulic retention time of 2 days (Leverenz et al., 2010). Sawdust and wheat straw serve as carbon sources and support biofilm growth (Fan et al., 2012; Saliling et al., 2007). More than 95% of nitrates were removed from domestic wastewater using sawdust over a period of 5.5 days (Schipper and Vojvodić-Vuković, 2001; Schipper et al., 2001).
- Solid-phase autotrophic denitrification. In this process, denitrifying bacteria use inorganic substances as electron donors to reduce nitrate to elemental nitrogen gas (Zhou et al., 2011). Chemolithotropic denitrifiers have certain advantages as they lower cell production, decrease the risk of biological contamination, require no external carbon sources, and remove problems associated with surplus organics (Sierra-Alvarez et al., 2007). The most important autotrophic denitrification processes are hydrogen-based denitrification, anaerobic ammonium oxidation, and a bio-electrochemical process. In hydrogen-based denitrification, the electron donor species is hydrogen gas. The main disadvantage of this denitrifier mechanism is its high operation and maintenance costs and sophisticated bioreactor setup (Lee et al., 2010; Rezania et al., 2007; Xia et al., 2010). The anaerobic ammonium oxidation (ANAMMOX) process uses nitrite as the electron donor species for the complete and direct oxidation of ammonium to nitrogen gas (den Camp et al., 2007). The main drawbacks of this process are the slow growth rate of ammonium-oxidizing and anammox bacteria (Kuenen, 2008), the cost involved in maintaining anaerobic conditions, and the lengthy start-up time of the reactor. Electroactive bacteria were

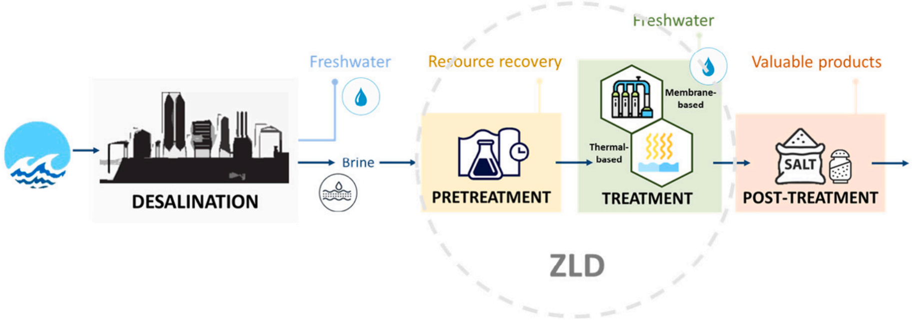

Fig. 1. Schematic diagram of desalination and brine treatment framework toward ZLD.

recently proposed as autotrophic denitrifiers, which have the ability to transfer electrons from carbon matter oxidation directly to a solid extracellular surface, such as a metal oxide or an anode. These extracellular electron acceptors act in a manner similar to the intracellular terminal electron acceptors for cell respiration (Ghafari et al., 2008b). Other electroactive bacteria species, such as cathodic bacteria, can accept electrons directly from a solid extracellular surface and use them to reduce molecules such as nitrate (Pous et al., 2015b; Prado de Nicolás et al., 2022; Pous et al., 2015a).

4. Brine treatment and zero liquid discharge (ZLD) approach

Providing adequate and safe drinking water is a critical global challenge. Freshwater scarcity is a major threat to economic growth, water security, and the health of ecosystems. This challenge is further complicated by climate change and pressure from economic development and industrialization (UNICEF et al., 2018).

In recent years, reverse osmosis (RO) has emerged as an alternative to traditional drinking water sources, especially in arid and semi-arid regions. At the end of 2015, desalination plants produced 86.55 million m^3/day globally. Around 65% of the production came from seawater RO (SWRO) (Ghaffour et al., 2013), which has a water recovery rate lower than 50%, with the remaining 50% being reject brine (Fritzmann et al., 2007), which is a major disadvantage of the RO process (Tsai et al., 2017). Brine discharged from seawater desalination plants has a negative impact on the environment, including soil, groundwater, and aquatic environments (Mohamed et al., 2005). Public health is also negatively affected. Therefore, various methods have been proposed and used to manage this brine (Tsai et al., 2017). Nevertheless, additional research is required to introduce environmentally friendly and economically viable management options for RO brine. The latest approach to the treatment of rejected brine is ZLD. This process aims to eliminate brine discharge while recovering water and salts, thereby reducing the potential negative impact on the planet and resulting in a more sustainable solution (Morillo et al., 2014). ZLD systems comprise a concentration stage (membrane technologies) and successive evaporation and crystallization stages (thermal technologies) (Kress, 2019). In each stage, total evaporation of the liquid fraction and precipitation or crystallization of the solid fraction and/or salts occur, as shown in Fig. 1. If this solid waste was previously treated and the polluting compounds eliminated, it could be revalued and used in industry, organic amendments, or agricultural fertilizers (Ahdab and Lienhard, 2021). However, ZLD is an expensive method and may have an indirect environmental impact because of its high energy requirements. To reduce energy consumption, increase the efficiency of the process, achieve zero discharge of liquids, and to incorporate renewable energy sources. it is necessary to understand each of the available technologies.

4.1. *Membrane-based technologies for brine treatment*

Based on previous contributions, the most widely used membrane technologies for reject brine treatments are set out below. Table 2 compares these technologies in terms of energy demand (kWh/m^3), water recovery ratio (%), and benefits and drawbacks of each.

- High-pressure reverse osmosis (HPRO).
  RO is an effective membrane-based technology for the desalination of seawater and brackish water (Greenlee et al., 2009). In this process, hydraulic pressure forces inflow (water to be desalinated) through a semi-permeable membrane, as shown in Fig. 2. In this way, only water molecules are forced to pass from a stream with a high concentration of salts to another stream where the concentration of salts is minimal. Finally, two flows are obtained, permeate (fresh water) and concentrate (reject brine) (Fritzmann et al., 2007; Qasim et al., 2019). However, the maximum working pressure for RO technology is approximately 80 bar, limiting the treatment of brines with a TDS concentration greater than 70,000 mg/l (Davenport et al., 2018). To overcome this limitation, a new technology that operates above 100 bar, HPRO, has been proposed. Thus, reject brines with TDS greater than 70,000 mg/l can be treated with a freshwater recovery between 40% and 70%. The specific energy consumption (SEC) of treated brine is between 3 and 12 kWh/m^3 (Rautenbach et al., 2000). The main limitation of HPRO is fouling of the membrane, which must receive specific pretreatments. Compared to RO, the capital and operating costs are higher, but represent greater efficiency. The cost of HPRO is significantly lower than that of thermal-based technologies (Davenport et al., 2018; Fritzmann et al., 2007).
- Forward osmosis (FO). This technology has emerged as a potential solution to the problems of scarcity of fresh water and energy. Unlike pressure-driven membrane processes, FO is a naturally occurring osmosis-driven process (Ge et al., 2013). A semi-permeable membrane separates two streams, one with a higher salt concentration. Osmotic pressure differences are used to drive the permeation of water across the membrane (Fig. 2). As in previous technologies, a concentrate and permeate are obtained. FO can overcome certain limitations presented by hydraulic pressure-driven membrane processes. FO features a high fresh water recovery rate, minimal reject brine discharge, low membrane fouling, and low energy requirements when the draw solute regeneration process is not considered (Shaffer et al., 2015). This technology, in addition to being able to alleviate the problem of fresh water scarcity at a low cost, also has applications in the food industry and drug processing (Ge et al., 2013).
- Osmotically assisted reverse osmosis (OARO). OARO is a membrane-based process for desalination of high-salinity brines. This technology has a high recovery rate and is energy-efficient (Peters et al., 2022). OARO combines both the FO and RO working principles (Fig. 2) requiring a hydraulic force to drive water molecules

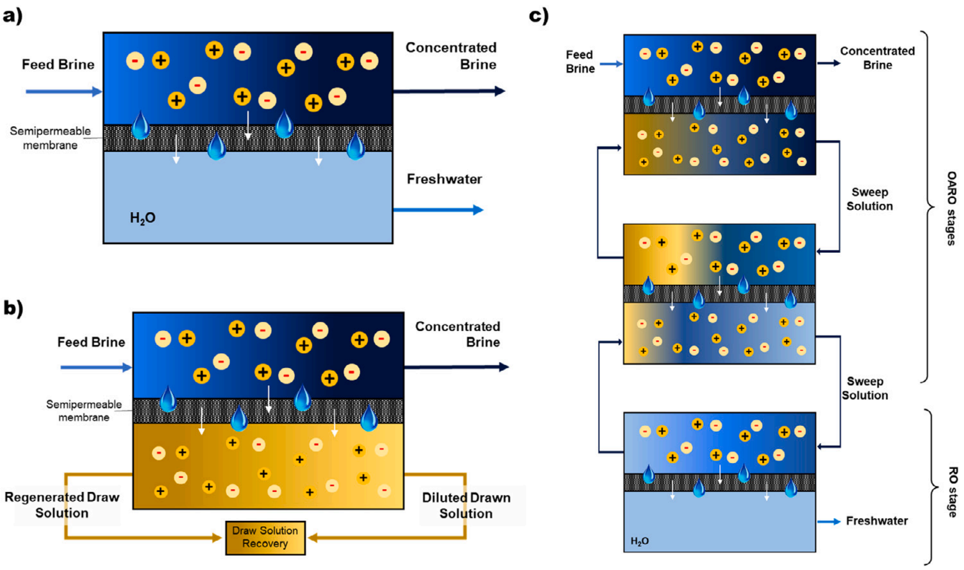

Fig. 2. Schematic diagram of (a) reverse osmosis (RO), (b) forward osmosis (FO), and (c) osmotically assisted reverse osmosis (OARO).

Table 2
Comparison of membrane-based technologies for ZLD.

Technology	Energy demand (kWh/m^3)	Advantages	Disadvantages	Water recovery (%)	Ref.
HPRO	3–12	Less energy intensive technology	Required pre-treatment Membrane fouling	40–70	Mike Mickley
RO	2–6	Less energy intensive technology	Not effective as standalone technology Required pre-treatment Membrane fouling	50	Davenport et al. (2018), Schantz et al. (2018)
FO	0.8–13	No pressure applied Low economic cost in water production	Required pre-treatment Membrane fouling	50–98	Ahmed et al. (2019)
OARO	6–19	No pressure applied Low economic cost in water production	Membrane fouling Multiple stages	50–72	Bartholomew et al. (2018), Peters and Hankins (2019)
ED EDR	7–15	No pressure applied	Required pre-treatment Membrane fouling Not energy optimized	85	Al-Amshawee et al. (2020), Valero et al. (2011), Fubao (1985)
EDM	0.6–5.1	No pressure applied Valuable compounds production	Required pre-treatment Membrane fouling Not energy optimized	90	Cappelle et al. (2017), Camacho et al. (2017b)
MD	39–67	No pressure applied No limit in [TDS] feed	Required pretreatment Membrane fouling	50–90	Abdelkader et al. (2018), Jantaporn et al. (2017)
MCr	50–90	No feed pressure requirements	Required pre-treatment Membrane fouling	90	Abdelkader et al. (2018), Jantaporn et al. (2017)

against osmotic pressure through a semi-permeable membrane, as in RO. However, the TDS concentration of the permeate is not zero, but there is a salinity gradient that reduces the difference in osmotic pressure. This allows the OARO technology to recover fresh water from more concentrated brines, especially when several OARO stages are connected in a series (Bartholomew et al., 2017). The OARO technology is one of the most promising alternatives for achieving ZLD. It features high fresh water recovery rates, minimizes the volume of reject brine, and has a lower SEC than the technologies described above. Furthermore, compared to thermal-based technologies, it is a more economic and efficient solution for minimizing the discharge of reject brine (Peters and Hankins, 2019).

- Electrodialysis (ED) and reversal electrodialysis (EDR). In ED technology, a direct current voltage is applied to achieve electrochemical separation of ions through an ion-exchange membrane. In this process, the positive and negative ions are transferred from the cathode and anode, respectively, to a rejection current, which becomes concentrated over time (Al-Amshawee et al., 2020). In this process, the dissolved solids are selectively eliminated because they migrate depending on their electrical charge and the selectivity of the ion-exchange membrane, as shown in Fig. 3. At the end of the process, three streams are obtained, (i) product water, which, like the permeate, has a low concentration of TDS; (ii) brine or concentrate, a stream that contains the ions removed from the influent; and (iii) electrode feed water, which is the

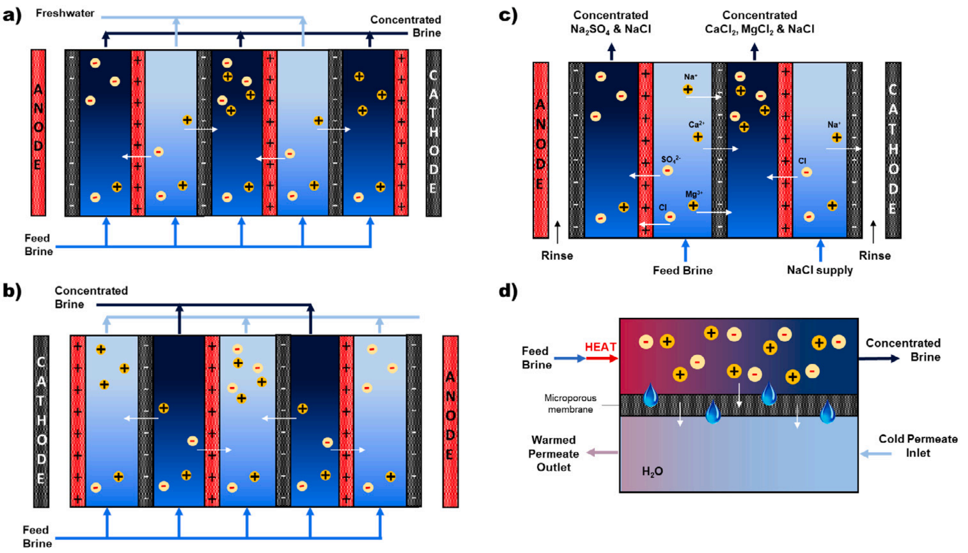

Fig. 3. Schematic diagram of (a) electrodialysis (ED), (b) ED reversal (EDR), (c) ED metathesis (EDM), and (d) membrane distillation (MD).

electrolyte of the anodic and cathodic chambers (Valero et al., 2011). EDR is a variation of the ED process, in which the polarization of the electrodes is used to clean the surface of the membranes (Fig. 3). Automatically, the polarization of the electrode's direct current is two to four times per hour. When the polarization changes, the direction of charge flow through the ion-exchange membranes also changes; in this way, the compartments of diluted water become those of concentrated water and vice versa (Fig. 3). This change in polarization prevents scale formation and precipitation of the membranes (Valero et al., 2011). EDR could be a promising solution for the industrial treatment of brackish water because the water to be desalted does not require pretreatment or the addition of chemicals. However, EDR technology does not reach a TDS concentration of zero in the product water nor does it eliminate pathogens from this stream (Fubao, 1985).

- Electrodialysis metathesis (EDM). EDM is a novel desalination process that is based on metathesis reactions. It is a modification of the ED in which the solubility of poorly soluble salts is increased. An EDM system comprises five chambers separated by four ion-exchange membranes to which a substitution solution is added that provides the necessary ions for exchange in the metathesis reaction (Fig. 3). Each of the chambers contain a different solution, two with influent flow, one substitution solution, and two other newly formed salts of the product that do not precipitate. This exchange of charges occurs because of the selective ion-exchange membranes, which allow the double exchange of ions between the salt solutions and the substitution solution (Camacho et al., 2017a). EDM can treat water with up to three times higher TDS concentration with less energy expenditure than RO. In addition, this technology can be used to recover salts that can be revalued and/or produce liquid by-products that can be used directly in industry (Bond et al., 2011). Similar to ED, the EDM energy requirement depends on ionic concentration. Consequently, EDM energy increases in direct proportion to the TDS (Camacho et al., 2017a).
- Microbial desalination cells (MDC). MDC is a technology developed in the last decade that combines microbial fuel cells (MFC) and electrodialysis for water desalination, also producing wastewater treatment and renewable energy. An MFC comprises two cells, one anodic and one cathodic, separated by an ion-exchange membrane. The electrodes of each cell are electrically connected using an external circuit (Prado et al., 2020a). In the anode cell, wastewater exists under anaerobic conditions; the organic matter is oxidized by the action of the electroactive bacteria, and the electrons produced are transferred to the surface of the carbon electrode by direct extracellular electron transfer. The electrons flow through the external electrical circuit to the cathode, where species such as oxygen, hypochlorite, or ferrocyanide are reduced. The protons migrate from the anode to the cathode through the proton-selective membrane, generating an electrical current. In this way, energy is produced simultaneously and wastewater is treated (Prado et al., 2020b). In an MDC, a third cell is incorporated and separated from the anode and cathode using an anion-exchange membrane (AEM) and a cation-exchange membrane (CEM). This third cell is fed saline water or brine. The potential difference between the anode and cathode, generated by the flow of electrons through the external circuit as a result of the oxidation of organic matter in the anolyte by the action of electroactive bacteria, causes the positive and negative charges of the salts (Na+ and Cl-) to migrate through the selective ion-exchange membranes to the cathode and anode, respectively (Al-Mamun et al., 2018). Operating under this ideal configuration, desalinated water is obtained in the central chamber, whereas residual water is treated in the anodic cell, producing renewable energy that is used to power the process (Sophia et al., 2016). The disadvantage is that this technology has not yet been developed. There are no electrode materials that allow economical profit scaling. MDCs are also not efficient enough to produce the energy necessary for complete desalination; only the most charged ions are eliminated in this process, but other important contaminants such as heavy metals, emerging metals, and nitrate remain, polluting the water. In addition, a more toxic waste than brine is generated, namely, ferricyanide, which must be regenerated or treated properly. Currently, there is no MDC operating at full scale (Saeed et al., 2015; Sophia et al., 2016; Al-Mamun et al., 2018).
- Membrane distillation (MD). This is a novel non-isothermal membrane process; thus, it is presented as a low-cost and energy-saving alternative. The partial pressure difference between the sides of the membrane is the driving force for passing water molecules from the feed stream to the permeate. Thermal energy is used to evaporate volatile molecules from the feed stream that condenses on the cold side of the membrane, as shown in Fig. 3. Owing to its hydrophobic nature, only vapor can pass across the membrane and not the liquid solution containing distilled water (Shirazi and Kargari, 2015). The advantages of MD compared

to other popular desalination processes are, (i) 100% rejection of ions and other non-volatiles, (ii) lower operating temperatures and pressures than conventional pressure-driven membrane separation technologies, (iii) minimal chemical interactions between process solutions and membranes, (iv) less stringent membrane mechanical properties, and (v) reduced vapor spaces compared to conventional distillation processes. However, MD has important limitations because the streams must be aqueous and sufficiently dilute to avoid wetting the hydrophobic microporous membranes. This limits MD to applications such as desalination (currently dominated by RO) (Lawson and Lloyd, 1997).

- Membrane crystallization (MCr). This is an innovative concept related to the implementation of membrane technology in crystallization processes. It is considered an extension of the MD concept based on mass transfer through a microporous hydrophobic membrane. The driving force is normally a temperature gradient between the two sides of the membrane. The hydrophobic nature of the membrane prevents liquid intrusion into the pores. Therefore, only volatile components are transported through the membrane, being condensed at the permeate sites. The mass transfer of volatile solvents allows concentration of the feed solutions to be above their saturation limit. Subsequently, they attain a supersaturated environment, in which crystals may nucleate and grow (Quist-Jensen et al., 2016). The benefits of using MD and MCr are as follows: very low operating temperatures and pressures, high permeate quality independent of water feed characteristics (theoretical 100% rejection of nonvolatile compounds), easy setup, and the possibility of treating highly concentrated streams. Unlike pressure-driven membrane technologies, the impact of TDS concentration on MD and MCr is small. Therefore, these processes are suitable for treatment of the RO reject brines. Furthermore, MCr has important advantages over traditional crystallization processes, well-controlled growth kinetics and nucleation, fast crystallization rates, low induction time, and control of the saturation level and rate (Quist-Jensen et al., 2016).

4.2. *Thermal-based technologies for brine treatment*

Thermal-based technologies use thermal energy (heat) to achieve desired separation during evaporation or distillation. This process mimics the natural water cycle, combined evaporation, and condensation steps (Panagopoulos and Haralambous, 2020a). Generally, thermal-based technologies are competitive systems but are less cost-efficient than membrane-based technologies because of their substantial energy requirements and high capital investment (Cipolletta et al., 2021a). The most commercially available thermal-based technologies are listed below. Table 3 compares these technologies in terms of energy demand (kWh/m^3) and water recovery ratio (%).

- Multistage flash distillation (MSF). Although MSF was developed as a desalination process, recently, it is being used with the ZLD approach to treat brine (Panagopoulos et al., 2019). The MSF technology consists of a series of stages in which the brine is recirculated until it has evaporated. The inlet brine is heated, and the vapor passes through the demisters and condenses to freshwater in the heat transfer tubes. The incoming brine flowing through the tubes is quantitatively heated by latent heat transfer from the vapor. The condensed freshwater is of high quality (Fig. 4). MSF distillers can operate at temperatures around 110oC [136]. The maximum capacity of an MSF system is approximately 75,000 m^3/day in a 20–30 stage configuration. The main advantages of this technology are simple operation, as it requires minimal pretreatment, and low fouling (Cipolletta et al., 2021a). However, MSF has high energy requirements and high capital investment costs. The electrical energy consumption is from 3.5 to 5 kWh-el/m^3, the thermal energy varies between 69.4 and 83.3 kWh-th/m^3 and the land footprint requires an average of 4.5–5 m^2/m^3 h (Panagopoulos and Haralambous, 2020b).
- Multi-effect distillation (MED). There are different types of MED systems, but all have the same distillation process. An MED plant consists of several consecutive hermetic cells, called effects. Each effect contains a bundle of horizontal tubes. Through such effects, consecutive evaporation–condensation processes occur at decreasing pressures and temperatures (Raluy et al., 2006). Seawater enters the first effect where the pressure is higher. It is sprayed onto the tube bundle carrying heating steam, usually from an adjacent power plant, and condenses on opposite sides of the tubes. When the thin-film of seawater comes into contact with the tube bundle, it boils. The seawater vapor produced in the first effect is demanded as an energy source in the next effect, where it subsequently condenses to produce distilled water (Fig. 4). This process continues along various effects, usually between 8 to 16 effects in a typical plant (Pumps, 2010; Kondili, 2010).
- Brine crystallizer (BCr). BCr was developed to produce freshwater and recover salts as valuable resources for other applications. In general, the BCr process comprises forced circulation of steam to concentrate the brine and cause the formation of bulk crystals. Brine crystallizers are composed of vertical cylindrical vessels and a steam source (steam compressor), which transfers heat to the vessels. The feed brine is pumped into a collection vessel. The feed stream is then mixed with the circulating brine and sent to the heat exchanger, where it is boiled with steam from the steam compressor. Subsequently, the heat exchanger tubes are submerged. Consequently, the brine is under pressure and does not evaporate. The recirculating brine enters the vapor body of the crystallizer at an angle, swirling in a vortex. This swirl produces partial evaporation of the brine and the consequent formation of crystals. A centrifuge or filter is used to separate the crystals. The rest of the brine is recirculated, heated in the steam compressor, and then condensed in the heat exchanger (Fig. 4). Finally, the products obtained are fresh water and dry solid salt crystals (Cipolletta et al., 2021a; Panagopoulos et al., 2019). BCr technology is best suited for treating reject brine with TDS up to 300,000 mg/l; thus, it is an efficient technology for the treatment of highly concentrated brine streams. However, BCr technology demands more energy and is more expensive than similar technologies, such as brine concentrator technologies (BC).
- Brine concentration (BC). This is a thermal technology that, via two different processes, reduces the volume of brine, which makes it an appropriate technology for the ZLD approach. The two different processes are (i) brine evaporation–cooling and (ii) concentration. The main design of a BC consists of a vertical tube or falling film evaporator. It can also be used for other evaporators, such as plate-type and horizontal spray films. The BC system consists of a heat exchanger where brine feed is supplied. The temperature of the brine is raised to boiling point, and the non-condensable gases are removed using a deaerator. Brine is inserted into the evaporator sump, which also receives the recirculating stream. The brine mixture is pumped to the top of the concentrator and then flows into a bundle of heat transfer tubes, where the water is evaporated. The steam passes through mist eliminators and reaches the steam compressor, where the temperature is further increased. As steam passes out of the evaporator tubes, heat is transferred to the cold brine. When the steam loses its temperature, it condenses as fresh water and is pumped through the feed heat exchanger, as shown in Fig. 4. The main advantage of the BC system is that 95–99% water recovery can be achieved. In addition, it can be used for brine up to 250,000 mg/l TDS. Furthermore, compared with other thermal-based technologies, the energy consumption is modest (15.9–26 kWh/m^3). However, the expensive materials used for the construction of these systems, such as super duplex stainless steel and titanium, increase their capital costs.

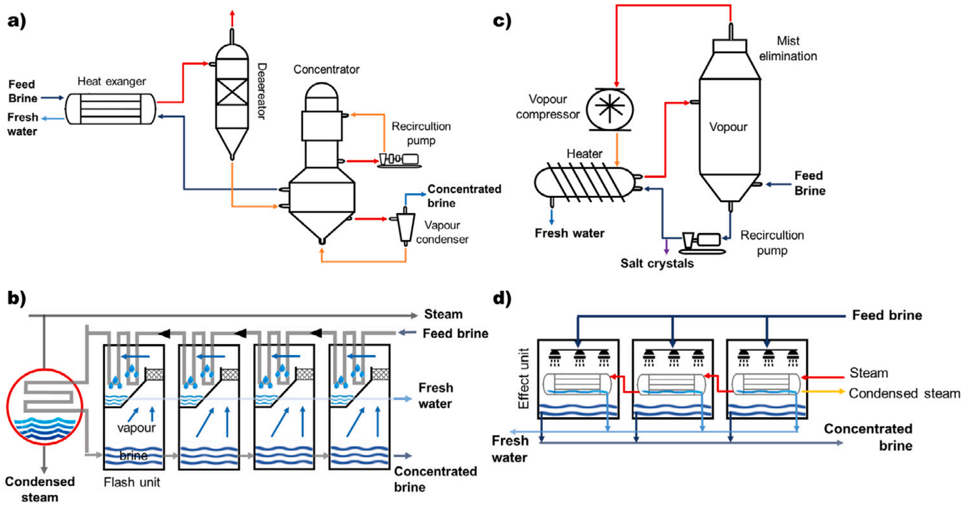

Fig. 4. Schematic diagram of (a) brine concentrator (BC), (b) multi-stage flash distillation (MSF), (c) brine crystallizer (BCr), and (d) multi-effect distillation (MED).

Table 3
Comparison of thermal-based technologies for ZLD.

Technology	Energy demand (kWh/m^3)	Advantages	Disadvantages	Water recovery (%)	Ref.
MSF	12.5–24	Large scale application Minimum pre-treatment No limit in [TDS] feed High-quality water	Scaling issues Moderate capital costs	85–90	Panagopoulos et al. (2019), Ihm et al. (2016), Panagopoulos and Haralambous (2020a)
MED	7.7–21	Minimum pre-treatment No limit in [TDS] feed Low thermal energy demand High-quality water	Scaling issues Moderate capital costs	85–93	Panagopoulos et al. (2019), Ihm et al. (2016), Panagopoulos and Haralambous (2020a)
BCr	52–70	Valuable compounds production	High capital material costs	97–99	Shaffer et al. (2015)
BC	15.86–26	No limit in [TDS] feed High-quality water	High capital material costs	95–99	Shaffer et al. (2015)
EFC	43.8–68.5	Valuable compounds production	High capital costs No large scale	98	Chivavava et al. (2014)
SD	52–64	Simple treatment	Not economically viable on a large No freshwater recovery scale	No recovery	Al Bazedi et al. (2014)
VCE	52–64	No limit in [TDS] feed High efficiency Low energy consumption High-quality water	High investment cost High water production cost	92% brine volume reduction	Cipolletta et al. (2021b), Panagopoulos (2020b)
WAIV	52–64	Moderate land requirement Low energy needs Valuable compounds production	No freshwater recovery No selective salt precipitation	No recovery	Gilron et al. (2003, 2019)

- Eutectic freeze crystallization (EFC). This crystallization technology is based on freeze crystallization instead of using evaporation–precipitation processes to obtain the precipitation of salts. The technology uses the difference in densities between the ice and salt produced to separate them. The theoretical basis is the eutectic point (EP) of each saline solution. In EP, there is a balance between ice, salt, and a specific solution concentration. According to this balance, if a solution is cooled below its EP, the salt concentration reaches saturation, and further cooling causes ice and crystal formation. The salt crystals precipitate to the deep layers of the reactor, whereas the ice crystals rise to the surface. Separation of the two solids is carried out using a separation unit (Fig. 5). In comparison to previous evaporating crystallization technologies, EFC has low energy consumption because the energy required to freeze water is less than that required for steam separation. However, EFC technology is still considered an emerging technology, and additional studies are needed for its full-scale implementation.
- Spray drying (SD). This thermal-based technology represents an alternative to crystallizers for brine treatment and concentration, obtaining value-added products such as mixed solid salts. The SD system comprises a feed brine tank, vertical spray drying chamber, and dried brine separator. The feed brine is sprayed into the vertical chamber using a centrifugal atomizer, while hot air is blown into the same chamber. With the help of the separator (bag filter), the dry brine is separated from the hot air stream, while air exits to the environment (Giwa et al., 2017), as shown in Fig. 5. Although the water evaporation capability of this

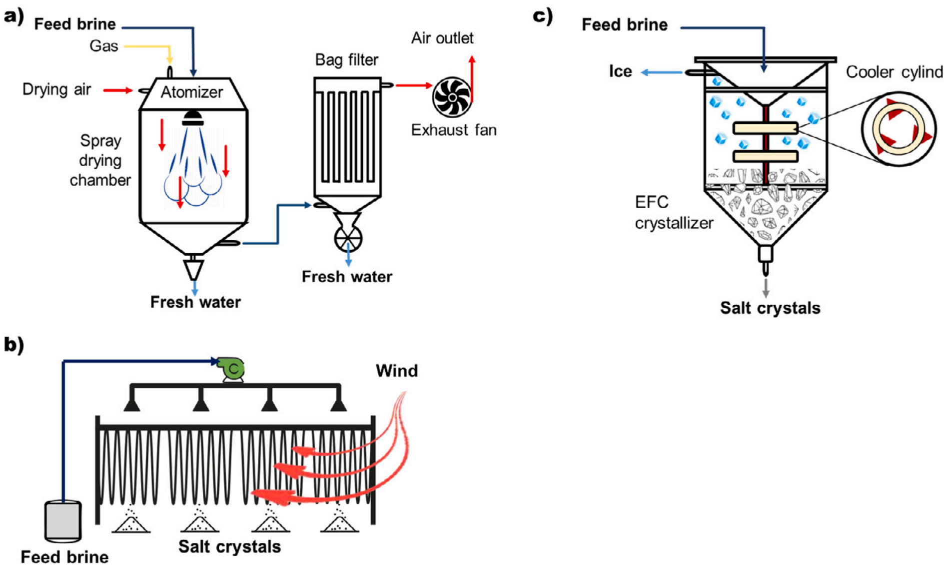

Fig. 5. Schematic diagram of (a) spay dryer, (b) wind-aided intensified evaporation (WAIV), and (c) eutectic freeze crystallization (EFC).

technology is not high, it allows the control of certain parameters in the separation of salts, such as particle shape, particle-size distribution, and bulk density (Panagopoulos et al., 2019).

- Vapor-compression evaporation (VCE) or distillation. This technology was developed to treat small volumes of brine between 250 and 20,000 m^3/d. Evaporation heat can be obtained thermally (TVC) or via mechanical vapor compression (MVC). Compressed steam is used as a heat donor to evaporate brine, which enters the system at low temperature and pressure. The permeate precipitates in the tubes of the evaporator as high-quality fresh water (Cipolletta et al., 2021b). The energy cost of these technologies is mainly associated with electrical requirements and occurs in a low range between 10 and 45 kWh-el/m^3 (Panagopoulos, 2020b). VCE is a technology with high efficiency; however, this is offset by high costs associated with the materials used for its construction and maintenance.
- Cooling tower (CT). The application of CT to brine evaporation is novel and is currently under development. In this solution, brine is heated in a heat-exchanging tank and pumped to the top of the CT. The CT inlet is on the top to guarantee the highest contact between the brine and air. The air stream is induced from the bottom to the top of the CT by an axial fan installed in the upper area of the tower. The liquid fraction of the brine is partially evaporated, and the remaining brine is precipitated. The precipitated brine is then recirculated and concentrated, reducing the rejected volume (Molina-García et al., 2021).
- Wind-aided intensified evaporation (WAIV). This thermal-based technology is used for brine treatment and volume minimization. WAIV technology was conceived with the aim of intensifying conventional evaporation ponds. Instead of solar energy, wind energy is used to increase the evaporation process. This vertical salt pan consists of vertical wetted packing towers made of non-woven geotextiles, woven settings, or volcanic tuff. The feed brine is sprayed on top of the evaporation surfaces and percolates through it using gravity. The wind blows orthogonally, favoring evaporation of the liquid fraction of the brine. The salts precipitate on the evaporation surfaces, thus obtaining a value-added product (see Fig. 5). Few studies have focused on investigating brine treatment with WAIV technology, but the results show that the evaporation performance of the WAIV unit is at least 10 times higher than that of the equivalent size conventional evaporation pond (Gilron et al., 2003, 2019). In addition, among all the technologies developed for the treatment of reject brine, WAIV has the lowest energy consumption because it uses wind energy, and only the brine feed pumps have an external energy requirement. Therefore, WAIV technology is an attractive alternative for brine treatment and should be investigated further.

5. Waste recovery

Fresh water is an increasingly valuable resource, not only because it is essential for the development of life, but also because it is crucial for other economic sectors such as industry. In addition to being an effective and eco-friendly treatment for reject brine, ZLD processes are suitable for the production or recovery of compounds with high added value (Panagopoulos, 2021b,a). Salt recovery from brine is a process with good market potential. Depending on the technologies used in the ZLD process, some salts can be recovered, and their purity percentage can vary significantly. Based on the exhaustive review carried out in this study, the most promising physicochemical processes for achieving ZLD and obtaining a profitable recovery of salts are chemical precipitation and thermal processes of evaporation–precipitation. The salts obtained from these processes will be high-molecular-weight salts ($CaCO_3$, $MgCO_3$, $CaSO_4$, $MgSO_4$, K_2CO_3, K_2SO_4, $BaCO_3$ and $BaSO_4$). In addition, other products of lower molecular weight but higher solubility, such as NaCl, have been recovered (Panagopoulos, 2020a; Gilron et al., 2019; Panagopoulos, 2020b). Industrial salt is a versatile natural element that plays an important role in the food industry and in numerous industrial processes. Common salt, or sodium chloride, is mainly found in seawater, underground in the form of halite rock, and in underground spring water. This resource is obtained by evaporation–crystallization processes of brine. The following are among the most common uses:

- Food industry. Salt is an economical and versatile antimicrobial agent. It also exhibits both preservative and dehydrating properties. In food, salt is also used as a binding agent, softener, and color enhancer. In addition, it is used to preserve cold chains, helping maintain low temperatures and preserving food properties through contributing to optimal conditions (Albarracín et al., 2011).
- Textile industry. In the manufacturing process, brine is used to remove organic contaminants from fibers owing to its ability to fix colors in fabrics. In addition, salt is used in the processing of animal skins in tanneries (Madhav et al., 2018).

- Metallurgical industry. Salt is used in metal processing to remove impurities (Tsakiridis, 2012).
- Mining industry. A common use for salt in the mining industry is for copper leaching (Feldman, 2000).
- Pharmaceutical industry. Salt is used in multiple applications in this industry, such as the production of intravenous saline and hemodialysis solutions.
- Chemical industry. One of the most common uses of salt in the chemical industry is electrolytic extraction of chlorine and sodium. Hydrochloric acid, sodium carbonate, and sodium sulfate are synthesized and later used for the production of plastics such as polyvinyl chloride (PVC). In addition, the chemical industry uses salt in the production of other derived salts, such as calcium hypochlorite ($Ca(ClO)_2$), chlorine dioxide (ClO_2), or sodium chlorate ($NaClO_4 \cdot H_20$). From the processing of salt, sodium hydroxide is also obtained as a base substance for the manufacture of fibers, textiles, soaps, and detergents (Feldman, 2000).
- Road de-icing. Salt is one of the most commonly used products to prevent the formation of ice on roads.
- Livestock industry. Salt is used to prepare natural salt blocks for animals, providing the necessary salts in their diet. It is also used in agriculture as forage silage, conserving nutrients for later consumption.
- Water treatment. Salt is used in saline chlorination or saline electrolysis, an advanced sterilization and disinfection system to treat water in an environmentally sustainable manner. This electrochlorination system converts sodium chloride into chlorine, a bactericidal reagent (Al-Hamaiedeh, 2013).

In addition to marine salts, other salts with high molecular weights and low solubilities are recovered. Those with higher market value are as follows (Panagopoulos, 2020b,a):

- Calcium Carbonate ($CaCO_3$). There are innumerable industrial applications of calcium carbonate. Therefore, the products of this mineral are almost as varied as their applications, with grain size being a determining factor for price. Some industries that use calcium carbonate are animal feed, glass, paper and cardboard, plastics, fertilizers, horticulture, surface coatings, asphalt, chemicals, food, pharmaceuticals, adhesives, and putties.
- Calcium sulfate ($CaSO_4$). Calcium sulfate can be adapted and mixed with a wide variety of products from different sources. It is not combustible, it does not pollute or produce toxic gases, and is odorless. It is used in various fields, such as the food industry and agricultural sector, and its use is expected to become more widespread. In addition, it plays an important role in the cement industry because it is the main component of gypsum used for construction.
- Magnesium Carbonate ($MgCO_3$). This compound has many benefits for human health and well-being. It is widely used in the pharmaceutical, cosmetic and food industries, among others. In pharmaceuticals, it is used as an ingredient in the manufacture of patented drugs, such as antacids and laxatives. Another of its uses is as an insulating and refractory material in different types of industry.
- Magnesium sulfate ($MgSO_4$). More commonly known as Epsom salt, this chemical compound is often hydrated. One of the main applications of the nonhydrated compound is its industrial use as a drying agent. In medicinal preparations, it is an aqueous solution used as a hydrate; in agriculture and water treatment, among others, it is used as a magnesium corrector.
- Potassium Sulfate (K_2SO_4). Potassium sulfate is important to the agricultural sector as potassium fertilizer is commonly used to improve the yield and quality of plants that grow in soils without an adequate supply of this essential nutrient. In addition, it is used for the removal of tartaric acid and has multiple uses in the dyeing and pharmaceutical industries.
- Potassium carbonate (K_2CO_3). Potassium carbonate is used in many industrial processes such as the manufacture of soaps, detergents, and ceramic products.
- Barium Sulfate ($BaSO_4$). Typically known as barite, this is a common heavy mineral, which is widely used in the petroleum and pharmaceutical industries. This element reacts strongly with water and corrodes rapidly in humid air. In fact, the element is so reactive that, in nature, it exists only as a compound. Barium compounds are obtained by mining and converting two barium minerals. The main application of barite is in drilling mud. Among other uses, it is used as a pigment in the production of hydrogen peroxide.
- Barium Carbonate ($BaCO_3$). Known as witherite, it crystallizes within the standards of an orthorhombic system, meaning the crystal faces have three right angles, with edges of different lengths. It is widely used in the ceramic industry as an ingredient in enamels and acts as a fl protection and crystallization agent. It is combined with certain coloring oxides to produce colors that are not easily achievable by other means.

In addition to the common salts, precious and useful metals/elements from brine can be extracted. Lithium, rubidium, scandium, gallium, and vanadium are among these metals. For example, lithium, called the ”new gold”, has been in demand in recent years, being necessary for the production of batteries for vehicles and electronic devices (Park et al., 2020). The value of these metals/minerals in the market determines the economic viability of the extraction/recovery process of these compounds from reject brine.

6. Conclusions

To meet the need for freshwater, desalination processes have recently been presented in the literature as a cost-effective solution. However, negative environmental impacts arising from the management and direct disposal of brine waste from desalination plants have also been reported. Brine management has thus become an important research topic. This review highlights the importance of treating all contaminants in brine, highlighting nitrate, and the ZLD strategy is proposed as a potential solution to end brine dumping while obtaining high value-added products such as salts and metals.

With respect to the removal of nitrate from reject brine, technological innovation is needed to overcome some drawbacks and to become an environmentally friendly, economical, and easily scalable solution. Denitrification transforms nitrate into nitrogen gas through a natural biological process but this nitrate attenuation pathway is a slow process. Currently, heterotrophic/autotrophic solid-phase denitrification processes have proven to be the most efficient and cost-effective approaches. In these processes, the solid organic matter supplemented to the medium usually comprises starch, synthetic polymers, methanol, or ethanol. These systems are also simple in design and in operation. In addition, the latest research reveals new technologies, such as those based on microbial electrochemical methods, which combine all the previous advantages while eliminating the drawbacks related to the contribution of solid organic matter, including residual carbon, cost of organic compounds, dosage, and high biomass production.

Membrane-based and thermal technologies have been proposed and used for brine management and operation in the ZLD framework. Nevertheless, there is no single treatment technology to achieve ZLD; therefore, a combination of technologies is required in most treatment plants. Commercial desalination technologies available on the market (RO, MED, and MSF) have osmotic limitations that prevent them from reaching ZLD, or they have a high economic cost, both in the construction process (expensive materials) and in the operation process (energy demand). Other technologies, such as BC and BCr, have been specifically designed for ZLD. Freshwater recovery is close to 100%; however, their high cost makes these technologies non-competitive.

Technologies such as FO, OARO, MD/MCr, EDM, and WAIV, have recently emerged and show promise in achieving zero discharge for treating high-salt reject brines. However, there are still some problems to resolve, such as fouling of the membranes, energy efficiency in thermal processes, and scaling. Finally, to achieve an efficient and sustainable large-scale application of ZLD desalination, renewable energy sources (RES) are being incorporated to power the processes. By incorporating renewable energies such as solar, wind and geothermal energy, thermal and electrical energy is obtained in a sustainable way and minimizing the emission of greenhouse gases. In this way, sufficient energy would be obtained to feed the desalination processes, achieve zero discharge of reject brine and obtain salts, free of nitrate, as a product with high added value. The ZLD-desalination is presented as an environmentally friendly solution to solve the problems of fresh water scarcity, if in addition the RES are incorporated, this circular economy model would achieve net zero emissions and waste generation.

CRediT authorship contribution statement

Amanda Prado de Nicolás: Methodology, Case example, Data curation, Validation. **Angel Molina-García:** Writing – original draft, Writing – review & editing. **Juan Tomás García-Bermejo:** Methodology, Writing – original draft, Supervision. **Francisco Vera-García:** Conceptualization, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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