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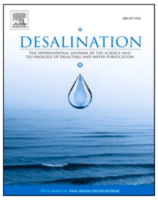

Electrochemical process of chlorination and energy generation as viable alternatives for SWRO brine valorization

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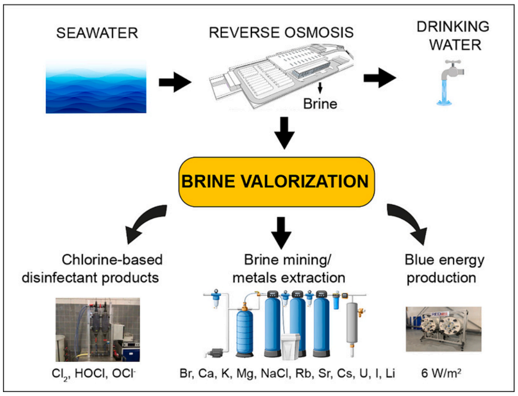
HIGHLIGHTS

- Disinfectant and energy production can be a solution for SWRO brine valorization.
- SWRO brine valorization technologies are still at low technology readiness levels.
- Nanofiltration is a promising pretreatment for brine mining and chlorine production.
- A SWOT analysis of SWRO brine valorization solutions is presented.
- To stimulate brine valorization, specific legislation is needed.

GRAPHICAL ABSTRACT



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ABSTRACT

The number of desalination plants worldwide increased exponentially in the last decade. This is basically a consequence of fast population growth combined with the expansion of water scarcity zones, even though desalination processes are, comparatively, much more energy intensive than other freshwater membrane treatment processes. SeaWater Reverse Osmosis (SWRO) has gained preference among the most used desalination processes due to its compacity, flexibility and energetic efficiency. However, desalination processes produce a concentrated brine that needs to be disposed of and represents an environmental challenge to be tackled. Currently in the literature, the most explored solutions for brine treatment are based on the extraction of valuable

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resources from the concentrate, also called brine valorization. Late discussions have pointed out that other approaches such as reuse for energy production and disinfection products might also be a solution for the same challenge. This review presented the current state of the art of SWRO brine valorization including the following methods: disinfectant (chlorine) production and salinity gradient energy generation; expanding the possibilities for brine treatment and reuse.

Nomenclature

AD	Adsorption
AEMs	Anion Exchange Membranes
BC	Brine Concentrator
BCr	Brine Crystallizer
BM	Brine “mining”
BMED	Bipolar Membrane Electrodialysis
BP	Booster pump
BPED	Bipolar Electrodialysis
BW	Brackish Water
CAPEX	Capital Expenditure
CAPMIX	Capacitive Mixing
CDLE	Capacitive Double Layer Expansion
CEMs	Cation Exchange Membranes
DE	Desorption
DP	Disinfection Production
EC	Electrocoagulation
ED	Electrodialysis
EDBM	Electrodialysis with Bipolar Membranes
EDM	Electro Metathesis
EDTA	Ethylenediaminetetraacetic Acid
EFC	Eutectic Freeze Crystallization
FO	Forward osmosis
F-SMDC	Fractional-submerged membrane distillation crystallization
GHG	Greenhouse Gas
HDH	Humidification-Dehumidification Distillation
HPP	High-pressure pump
IX	Ion Exchange
IXM	Ion-exchange Membrane
LCA	Life-cycle assessment
MBC	Membrane Brine Concentrator
MD	Membrane Distillation
MDC	Membrane Distillation-Crystallization
MED	Multiple Effect Distillation
MLD	Minimal Liquid Discharge (MLD)
MSF	Multi-Stage Flash Distillation
NF	Nanofiltration
NTC	NaCl Thermal Crystallizer
OARO	Osmotically Assisted Reverse Osmosis
OPEX	Operating Expense
PRO	Pressure Retarded Osmosis
RED	Reverse Electrodialysis
RO	Reverse Osmosis
RW	River Water
SD	Solar Desalination
SDGs	Sustainable Development Goals
S-ED	Selective Electrodialysis
SGE	Salinity Gradient Energy
S-MD	Submerged Membrane Distillation
SMSR	Submerged Membrane Sorption Reactor
STY-DVB	Styrene-Divinylbenzene
SW	Seawater
SWRO	Seawater Reverse Osmosis
TD	Thermal desalination
TDS	Total Dissolved Solids
TH	Total Hardness
TRL	Technology Readiness Level
UHPRO	Ultra-high pressure Reverse Osmosis
UN	United Nations
VC	Vapor Compression distillation
WAIV	Wind-Aided Intensified eVaporation
WW	Wastewater
WTP	Water Treatment Plant
WWTP	Wastewater Treatment Plant
ZLD	Zero Liquid Discharge

1. Introduction

Water is a crucial product to allow us reaching a sustainable development, create healthy ecosystems, improve energy and food production, as well as achieve socioeconomic development. Freshwater resource pollution and drought, associated with the increasing population and lifestyles worldwide require the exploration of additional sources, such as brackish water, groundwater and seawater, which have always been considered saline and impaired water sources [2]. To address the global warming crisis, desalinated water production is expected to increase even more in the coming decades [3]. Seawater is considered one of the key water resources for supporting UN Sustainable Development Goal (SDG) 6 (clean water and sanitation) [4]. Moreover, chemical resource recovery from the sea is an attractive research topic and improvements in technologies for the extraction of strategic elements such as lithium and uranium should be developed [5].

Based on plant capacity, seawater accounts for approximately 57 % of the feed water used in desalination plants [6]. Conventional desalination methods are membrane and thermal desalination. Among the membrane desalination methods, the most common is seawater reverse osmosis (SWRO) which is applied in 69 % of desalination facilities worldwide [4]. SWRO is popular because of its low installation costs [7] and low energy consumption (3–4 kWh_e/m^3), when compared with thermal processes. However, although comparatively lower, it has the potential to produce large greenhouse gas (GHG) emissions, due to energy consumption and production of waste streams [8]. Brine production from SWRO is estimated to be around 100 million m^3/day around the world. [3]. This large waste stream is mainly caused by the recovery efficiency of the RO process often being around 40 %, meaning that only this percentage of the feed water becomes permeate, with the rest turning into a concentrate stream (brine) [3].

Reverse osmosis has been the most published topic in the last 40 years compared to other desalination technologies (e.g., multi-effect distillation (MED), multi-stage flash (MSF), electrodialysis (ED)), and emerging technologies) [4]. Fig. 1 shows a representative diagram of a seawater reverse osmosis system. The process usually consists of seven steps: water intake, pre-treatment, pumping system, reverse osmosis, energy recovery system, post-treatment and brine discharge [9,10].

The use of seawater for its valuable compounds, such as salt extraction, has been reported since 2200 BCE by the Chinese [11]. Later in 1914, Dow Chemical Company has produced magnesium from brine in the U.S [12,13]. Even though the topic has not been recent, the scale of brine production has increased dramatically in recent years, with an increase in desalination due to population increases in coastal areas and frequent droughts as a result of climate change [4,14]. Brine production reached 141.5 million m^3/day in 2018 [4], being most of that discharged from SWRO systems.

The disposal of brine can have impacts on the environment, depending on the quantity and composition of the brine, as well as the disposal method used. The brine environmental impact intensity are primarily influenced by temperature, TDS, and density, in a way that, the higher the temperature the lower the impact, the higher the density or the TDS the higher the impact [15]. High recovery rates will increase TDS and consequently, increase the overall environmental impacts [15].

Moreover, the potential environmental effects of rejected brine include eutrophication because of phosphate supplementation, pH changes, discoloration due to iron (III) concentration, high suspended solids, turbidity, accumulation of heavy metals in marine ecosystems and accumulation of negatively buoyant plumes, which can create a range of problems in marine and subsurface habitats [14,16]. Negative buoyant discharges are not specifically addressed in most current environmental laws; therefore, the regulatory framework should revise existing mixing zone criteria to account for site-specific conditions [17].

Another potential effect of brine on the environment is water quality degradation through an increase in salinity levels and a consequent reduction in dissolved oxygen levels, although these effects are restricted to the vicinity of the area. In naturally occurring benthic bacterial populations exposed to water with salinity >5 %, effects such as a reduction in bacterial quantity and inner-cellular metabolic activity were observed [18,19]. Another study revealed that phytoplankton are more susceptible to effluent discharge from desalination plants than zooplankton are, having RO discharges the highest impact; however, thermal desalination plants may have overall greater impacts on planktonic populations than RO plants due to positive buoyancy effluent, non-neutralized chlorine, increased temperature and copper discharge from the corrosion of heat exchangers [20,21]. The elevated temperature of brine discharge from thermal desalination, usually 5 to 15 °C higher than the ambient seawater temperature, has been reported to be harmful for marine life, causing coral bleaching and ultimately coral mortality [22–24]. Moreover, the accumulation of chemicals such as antiscaling and antifouling agents from desalination plants can cause hazardous effects in the environment. Anti-foaming agents, which are required in thermal desalination plants, were reported to cause 84 % of ozone depletion [25]. Chlorine residue from thermal process was reported to be ten times higher than that from RO process [15]. Hence, the literature suggests brine post-treatment, brine dilution and backwash discharge on the continent to minimize the impact on these species.

Brine valorization has been gaining attention as a sustainable solution for managing the increasing volumes of seawater reverse osmosis (SWRO) concentrate. Researchers have associated brine management strategies with many SDGs (6, 7, 8, 9, 12, 13, 14 and 17), but mainly with SDG 9 'Industry, innovation and infrastructure' [3]. Brine treatment schemes allow for material recovery in the form of salts, metals, minerals, nutrients, acids, bases; energy recovery; water recovery through water reuse or even freshwater production [26].

According to the state-of-the-art concept of sustainable waste management, the 9Rs of the circular economy to product chain [27,28] refuse, rethink, reduce, reuse, repair, refurbish, remanufacture, repurpose, recycle and recover can be adapted for use in relation to seawater treatment waste. As SWRO brine is a byproduct of an industrial process for generating clean water, the concept can comprise of 7Rs: rethink, reduce, reuse, recirculate, repurpose, recycle and recover.

Most research on brine management has focused only on reuse, recycling and recovery. However, advances related to the Rs at the beginning of the circular chain, such as reducing the volume of treated/desalinated water consumption are also important. This can be achieved by improving industrial processes that consume water and increase water catch on water basins, among other actions. Flushing toilets and cooling spaces with non-treated seawater are alternatives found in Hong Kong to reduce seawater treatment and, consequently, brine production [29].

Brine are high salinity solutions with TDS concentration above 36,000 mg/L [15]. It comes from many sources including desalination plants, manufacturing processes, salt manufacturing, natural gas storage and mining and chlor-alkali processing. Many resources can be extracted from desalination brine: water, minerals and salts, energy, metals, and chemicals/bioactive compounds [30].

Currently in the literature, the most explored solutions for brine treatment are based on the extraction of valuable resources from the concentrate, also called brine valorization. Reviews have explored the brine valorization from desalination system: brine management practices have been reviewed focusing on minimal liquid discharge (MLD) and zero liquid discharge (ZLD) systems [26,30,31], membrane-based ZLD has been discussed by Tsai et al. 2017 [32], magnesium recovery from seawater brine was technically reviewed [33]. However, due to the large volume of both water and mineral salts present in brine, late discussions have pointed out that combined solutions such as brine mining and chlorine production, or brine mining and salinity energy generation or even chlorine production and salinity energy generation might possible alternatives to be presented in the same plant.

This study aims to provide the readers with an overview of the most recent research developments for the most common desalination brine disposed in the environment, SWRO brine, with a focus on disinfectant (chlorine) production, and salinity gradient energy (SGE) generation, as

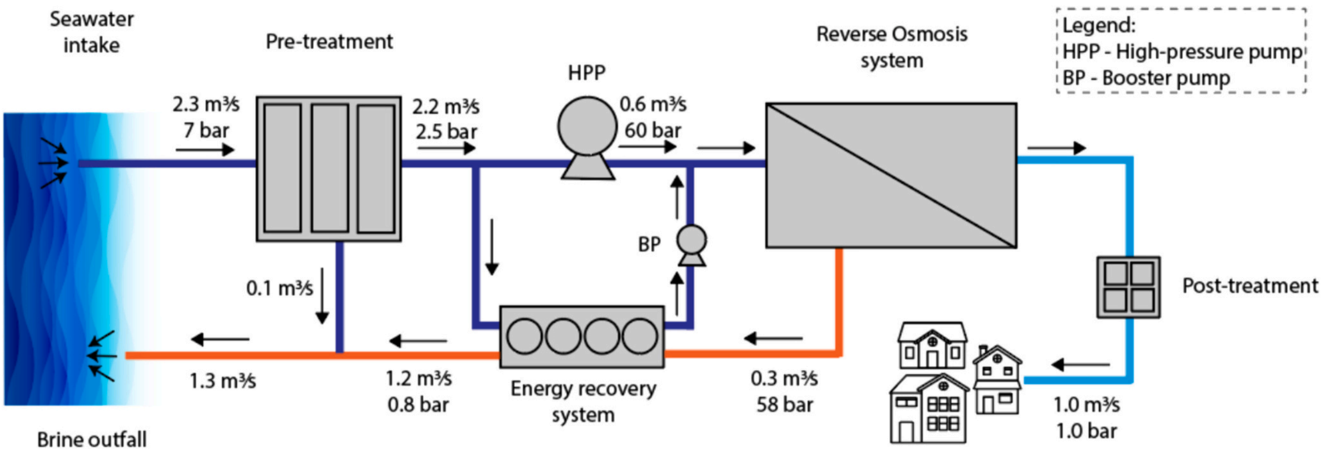

Fig. 1. Generic flow diagram of a seawater reverse osmosis desalination plant producing 1 m^3/s, with a recovery rate of 45 %.

well as to assist decision-makers in producing more cost-effective systems towards a more circular economy. The paper is structured in sections presenting a short bibliometrics on the topic (Section 2), followed by a discussion on SWRO brine composition. The brine valorization approaches are presented in Section 4, and Section 5 presents a comparative analysis of the approaches presented in Section 4, considering environmental impact, technological consolidation, and commercial appeal. Section 6 presents the final consideration and a SWOT summarizing the analysis from Section 5. Lastly, Section 7 draws conclusions according to the findings.

2. Short bibliometrics

A brief bibliometric study on brine valorization was carried out in Scopus and Web of Science databases, in March 2024, to identify the research trends on the topic. In the first moment, the terms "brine AND valorization" and "seawater AND reverse AND osmosis AND brine" were selected. In the second moment, the terms "brine AND mining", "brine AND chlorine AND production", and "brine AND salinity AND gradient" were compared.

While SWRO brine has been a topic of interest since 1969 in Scopus, the first publication on brine valorization was only in 2004 in the Web of Science and Scopus databases. Approximately 70 % of the papers on brine valorization were published in the last five years (2018–2022) (Fig. 2). Among the published documents on this topic, Desalination, Desalination and Water Treatment, Membranes, Separation and Purification Technology and Chemical Engineering Journal were the five most popular journals on both scientific databases. The countries with the most publications on the topic were Spain, Italy, China, the U.S., France and Greece. Another bibliometric study revealed that China, the U.S., India, Iran and South Korea were among the top 5 countries in terms of their scientific production on desalination [34]. In the mentioned study, reverse osmosis and brine were two clusters with publication weights of 27.16 % and 13.07 %, respectively.

For "brine AND valorization" results on Scopus, "electrodialysis" was one of the most common keywords, as well as were "energy utilization", "sodium chloride" and "ion exchange". On the other hand, "recovery" and "resource recovery" were the 16th and 18th most common keywords, respectively. For "seawater AND reverse AND osmosis AND brine", "environmental impact", "recovery", and "sodium chloride" appeared in 13th, 17th and 20th positions, respectively.

Even with the recent growth in brine valorization scientific production, environmental issues and recovering materials from brine are still emerging topics, indicating the need for more study and discussion.

The number of documents reported by Scopus and Web of Science on the terms "brine AND mining", "brine AND chlorine AND production", and "brine AND salinity AND gradient" are found in Fig. 2.b. "Brine AND mining" appears five and three times more in the literature compared to "brine AND chlorine AND production" and "brine AND salinity AND gradient", respectively. Hence, there is a research gap on exploring

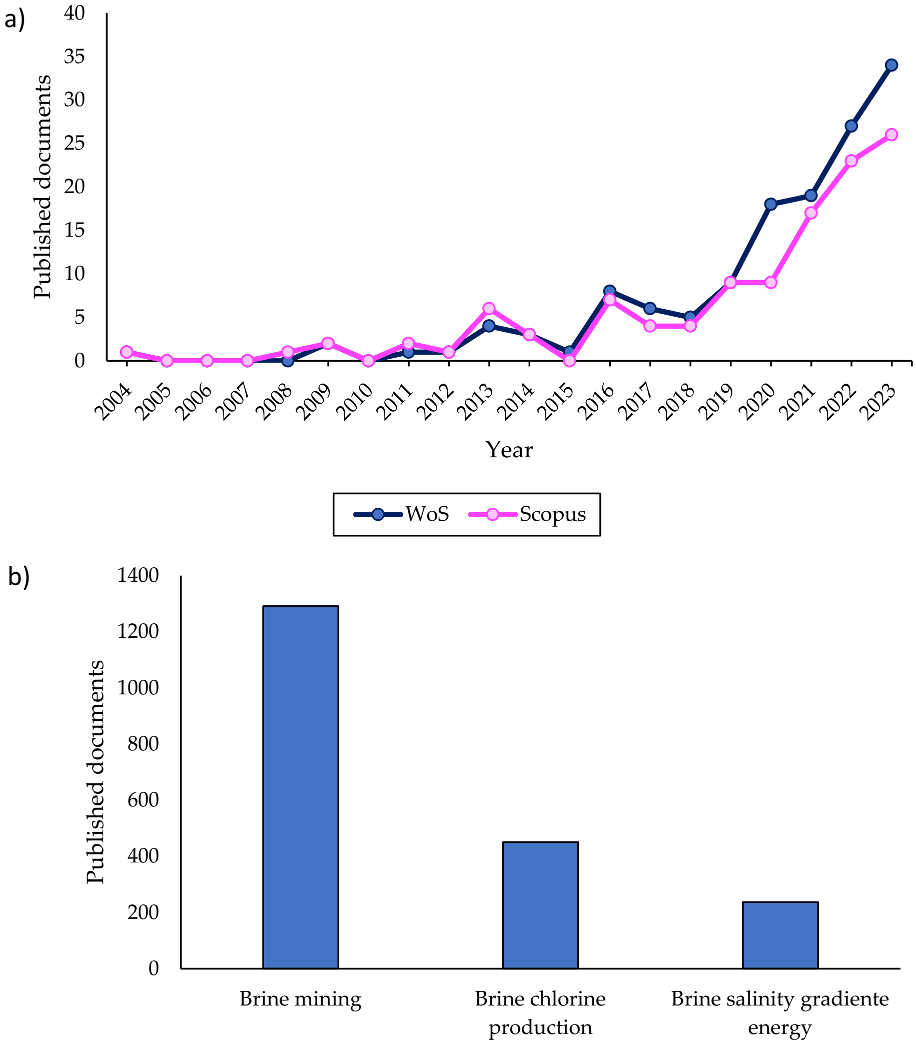

Fig. 2. a) Evolution of the number of documents published on brine valorization, b) Documents published on Scopus on each term.

chlorine production and salinity gradient production as feasible alternatives for brine valorization.

3. Brine composition

Approximately 80 % of the seawater salt is NaCl. Thus, Na^+ and Cl^- are the ions at higher concentrations in SWRO brine. However, their concentrations are slightly different around the globe. Table 1 shows the composition of SWRO brine in different countries. Magnesium, the second most abundant cation in seawater, is present in brine at almost ten times lower concentrations than Na^+, followed by Ca^{2+} and K^+. The concentration of Cl^- ions was 4 to 7 times higher than SO_4^{2-}.

Brine is also composed of SWRO pre-treatment chemicals, such as antiscale additives (e.g., polycarbonic acids, ethylenediaminetetraacetic acid (EDTA), and sodium hexameta phosphate), as well as membrane cleaning chemicals such as oxidants (sodium perborate) and biocides (formaldehyde) [35]. Trace metals such as barium, cesium, indium, iron, lead, lithium, vanadium, zinc, strontium and uranium are also present in SWRO brine [36]. The concentrations of Ba, Zn, Fe, Cu, Pb, V, Li and Sr have been reported to be 0.16 mg/L, 0.845 mg/L, 1.31 mg/L, 1.165 mg/L, 1.505 mg/L, 3.88 mg/L, 43.32 mg/L and 16.93 mg/L, respectively [37].

Brine composition and parameters are dependent upon many factors, such as input quality and quantity, the desalination process, the discharge method, and climatic and seasonal variation, which impact the likelihood of biofouling and membrane scaling [38].

In the studies where total dissolved solids (TDS) were presented (Table 1), the average ratios of Cl^-, Na^+, SO_4^{2-}, Mg^{2+}, Ca^{2+}, K^+, and HCO_3^- to TDS were 53 ± 2 %, 33 ± 4 %, 9 ± 1 %, 3.7 ± 0.5 %, 1.4 ± 0.3 %, 1.4 ± 0.2 % and 0.5 ± 0.1 %, respectively. The percentage range of the major ions in a brine of TDS was aligned with that reported by the SWRO brines from ten SWRO plants in the Canary Islands (Spain) [39]. Fig. 3. shows the boxplot of the most common ions present in brine as described in Table 1.

4. Brine valorization

Freshwater resources will only be sufficient to satisfy the requirements for 60 % of water consumption by 2030, and by 2050 and 2100, they will only be sufficient for 55 % and 40 %, respectively, of consumption, leading to a serious global water crisis [7]. Brine management and valorization are key aspects in making desalination a viable option for mitigation of water scarcity. Water is the most common resource extracted from brine through a variation of methods of brine concentration, which also increases the water efficiency of seawater desalination. [53]. It is also integrated into the zero liquid discharge (ZLD) strategy, which can achieve net zero emissions and waste generation when incorporated into renewable energy [1,30].

Sustainably managing brine requires seeing it as a resource and not a waste [16]. Brine valorization refers to the process of extracting valuable resources from SWRO concentrate, which includes elements in the form of minerals and nutrient; and energy [31]. Hence, this approach offers a promising sustainable solution for managing SWRO concentrates. The brine valorization technologies usually comprise these three stages: pre-treatment to separate the undesirable components from the brine, mostly divalent ions (e.g., chemical coagulation, chemical precipitation, electrocoagulation (EC), ion exchange, nanofiltration, adsorption, etc.) [31,54]; concentration, in which more fresh water is extracted from the brine, concentrating it – this step is dependent on the requirements of the technology applied in the next step (e.g., evaporation, brine concentrator and brine crystallizer) [39,55]; and, finally, conversion, where the concentrated brine is processed into liquid components or solids salts and minerals [39]. A classification of each technology is presented by Rivero-Falcón (2023). The concentration and conversion stages are estimated to correspond to 60 to 70 % of the CAPEX and OPEX of the zero liquid discharge (ZLD) strategy [56].

The selection of a suitable brine valorization method depends on various factors, including the brine composition, volume, and location. It would be useful to develop a tool based on a decision matrix that evaluates the different factors for several technologies, which would allow for comparison of different brine valorization technologies and provide a clear roadmap [39].

For recovering resources from brine, several methods can be used such as stand-alone or hybrid methods, and these processes can be divided into two main categories: membrane-based and thermal-based processes [57].

Membrane-based technologies are the most commonly used methods for brine valorization, but they have limitations, including high capital and operational costs, fouling, and scaling.

Membrane technologies for concentrating brine can be pressure-driven (osmotically assisted reverse osmosis (OARO), nanofiltration (NF), ultra-high pressure reverse osmosis (UHPRO)) or electrically driven (e.g., monovalent selective electrodialysis (MED), electro metathesis (EDM), bipolar electrodialysis (BPED)) [58]. The selection of technology depends on water quality, costs and development stage [59].

Thermal-based methods can also be used to treat SWRO brine, producing water vapor while separating salts but requiring high energy consumption [60]. Some thermal-based methods include multi-stage flash (MSF), multiple-effect distillation (MED), vapor compression distillation (VC), brine concentrator (BC), brine crystallizer (BCr), humidification-dehumidification distillation (HDH), solar desalination (SD), and freezing (FZ) methods [61,62]. These methods are conventionally used for TDS concentrations up to 35 g/L. At higher concentrations, they are less efficient.

Emerging technologies such as forward osmosis (FO), membrane distillation (MD), thermo-ionic processes, eutectic freeze crystallization (EFC) and wind-aided intensified eVaporation (WAIV) were identified as promising solutions for brine concentration with high-TDS brine treatment [59,62].

Membrane-based and thermal-based technologies were reported in literature for treating SWRO brine and transforming it into a valuable product (Table 2): manofiltration (NF), reverse osmosis (RO), Brine crystallizer (BCr or Cr), Brine concentrator (BC), membrane brine concentrator (MBC) multi-stage-flash (MSF), bipolar membrane electrodialysis (BMED), electrodialysis with bipolar membranes (EDBM), electrodialysis (ED), selective electrodialysis (S-ED) ion-exchange membrane (IXM), NaCl thermal crystallizer (NTC), membrane distillation-crystallization (MDC), adsorption (AD) + desorption (DE), submerged membrane sorption reactor (SMSR), submerged membrane distillation (MD)-absorption process, eutectic freeze crystallization (EFC), and CO_2 demineralization. The combination of these methods, also called multistage treatment, has been reported, when the effluent from one stage feeds another stage. A multistage treatment also permits the recovery of more products from the brine.

A wide range of elements recovered from SWRO concentrated has been reported in the literature, most of which are alkali metals, such as Na, Li, K, Cs and Rb. The most common products were sodium chloride and sodium hydroxide. The main application of these elements chemical agents is as substitutes for the chloro-alkali and mining industries. Potential applications of the resources recovered from SWRO brine have been reported in the literature as raw materials for detergent, glass and paper industries [52], water disinfection, construction [63–65], chemical industries [48], and for production of lithium-ion batteries [66,67].

Most common approach, brine mining, will be briefly discussed in the following topic. Disinfectant production and salinity energy generation will be discussed as new approaches to implement cost-effective brine valorization in SWRO plants.

4.1. Current explored brine valorization approach (brine mining)

The extraction of minerals from brine is also known as brine “mining” and has the potential to compete with the traditional mining

Table 1
Major brine composition of SWRO plants (g/L).

	Location												
Parameter	Barcelona (Spain)	Gran Canaria (Spain)	Canary Islands (Spain)	Fortaleza (Brazil)	Sataria (Italy)	Almería (Spain)	Gwangyang (Korea)	Tianjin (China)	Fernando de Noronha (Brazil)	Shuwaikh (Kuwait)	Doha district (Kuwait)	Al Shuqaiq (Saudi Arabia)	Perth (Australia)
pH	–	–	7.9	7.23	–	–	7.6	–	8.0	7.04	7.13	6.8	8.0
Cl^-	38.8 ± 0.4	37.64	39.12	38.0	39.0	31 ± 2	32.6	27.43	40.9	–	–	36.866	–
Na^+	20.8 ± 0.3	20.66	20.51	22.5	21.4	15.3 ± 0.5	–	15.68	22.3	27.802	17.905	20.537	23.100
SO_4^{2-}	5.41 ± 0.2	5.63	5.42	7.8	5.50	5.3 ± 0.4	5.050	3.84	6.07	7.497	4.159	5.131	–
Mg^{2+}	2.64 ± 0.2	2.75	3.04	2.3	2.70	1.9 ± 0.6	6.100	1.87	2.80	2.703	1.673	2.440	2.3905
Ca^{2+}	0.83 ± 0.04	0.81	0.97	0.94	0.88	0.88 ± 0.05	1.760	0.61	0.96	1.040	1.090	0.786	0.7893
K^+	0.75 ± 0.05	0.81	0.92	0.78	0.78	–	–	–	1.93	1.1417	0.997	0.760	0.7902
Br^-	0.13 ± 0.06	–	0.14	0.00	–	–	–	–	–	0.00004	0.00002	0.128	–
Sr^{2+}	0.016 ± 0.003	–	0.022	0.010	–	–	–	–	–	0.0504	0.121	0.015	0.01542
SiO_2	< 1	–	< 0.024	<0.1	–	–	–	–	–	–	–	–	–
Al^{3+}	< 0.5	–	0.012	ND	–	–	–	–	0.0006	–	–	–	–
Fe^{3+}, Fe^{3+}	< 0.2	–	<0.010	ND	–	–	–	–	0.0002	<0.00001	<0.00001	–	–
Ba^{2+}	< 0.2	–	<0.009	< 0.001	–	–	–	–	<0.000002	<0.001	<0.001	0	–
Ni^{2+}	0.07 ± 0.02	–	<0.002	–	–	–	–	–	–	–	–	–	–
Cu^{2+}	0.03 ± 0.01	–	<0.002	ND	–	–	–	–		<0.00001	<0.00001	–	–
Mn^{2+}	0.01 ± 0.01	–	<0.002	ND	–	–	–	–	0.00007	–	–	–	–
Cr^{3+}	0.007 ± 0.003	–	<0.002	ND	–	–	–	–	–	<0.00001	<0.00001	–	–
HCO_3^-	–	0.45	0.30	0.26	0.18	–	–	–	–	–	–	0.260	–
NO_3^-	–	–	0.003	0.02	–	–	–	–	0.0115	–	–	0.004	–
TDS		68.764	70.44	75.682				–	–	78.450	54.900	–	58.80
Ref.	[40]	[41]	[39]	[42]	[43]	[44]	[45]	[46]	[47]	[48]	[48]	[49]	[50]

*ND: not detected: -: data not available.

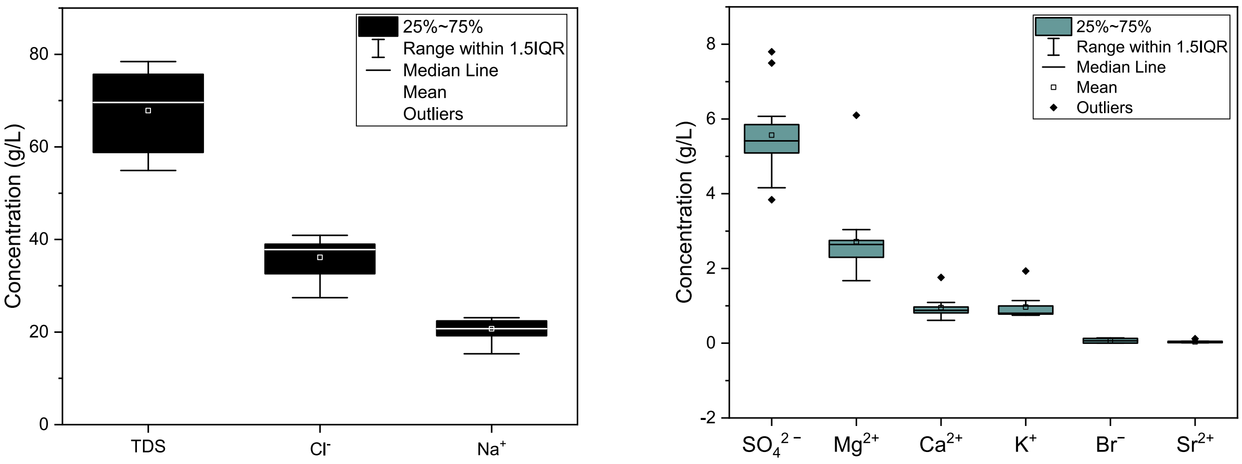

Fig. 3. Boxplot showing a comparison of major ion concentrations in SWRO brine reported in the literature [39–52].

industry. Given that seawater contains almost every element in the periodic table [93], the purpose of brine mining is to recover valuable resources from saltwater [58]. Mineral recovery from desalination brine has the potential to address three issues: mineral shortages, limiting the environmental implications of brine discharge, and lowering desalination costs [94]. Brine mining is often seen as more ecologically friendly than dry-ore mining for various reasons: it is less energy intensive, has salty water acting as a natural solvent for compound extraction, and does not require the excavation of large amounts of land; additionally, the very salty areas in which brine is found are unfriendly to most plant and animal life; hence, the impact of brine mining on biodiversity is minor [58,95].

Brine "mining" of Br, Ca, K, Mg, NaCl, Rb, Sr [96], Cs, U, I and Li [97] has been considered economically feasible. The potential profitability of components extracted from seawater depends on the volume; the more brine produced, the more likely the extraction procedure is to be profitable [98,99]. The feasibility of extracting commodities such as LiCl, NaCl, $CaCO_3$, Rb_2CO_3, $Mg(OH)_2$, Cl_2, NaOH and Br_2 is also dependent upon commodity pricing and final product purity [99].

Although disinfectant production can also be considered brine "mining" it will be discussed in the following section while this section will focus on the potential metals, acids and bases available in brine. Fig. 4 presents a general diagram for mineral recovery from brine.

Evaporation with sequential precipitation, selective sequential precipitation, membrane separation, electrodialysis, membrane distillation and crystallization (MDC), and adsorption/desorption/crystallization are the principal methods used or proposed to harvest minerals from saltwater [97]. In all of these processes, the concentration of the metal to be extracted is first increased to supersaturation to allow crystallization [98]. Except for the last, all of these methods require that the solubility of the salt product be lower than that of the enhanced ionic product of the component ions.

The process of adsorption/desorption/crystallization is not affected by brine content. Rubidium adsorption/desorption from SWRO has been achieved by granular potassium copper hexacyanoferrate (KCuFC) followed by desorption of pure RbCl using NH_4Cl as the eluent [92]. Boron recovery from SWRO brine was simulated through adsorption onto functionalized resin (STY-DVB), followed by desorption using HCl, distilled water and NaOH [69]. The recovery of $Mg(OH)_2$ from SWRO brine was achieved by combining magnetic separation with adsorption on Fe_3O_4 micro-particles, after which acid desorption yielded a high purity (>97 %) Mg(II) solution [76].

Sharkh et al. (2022) reported that brine mining focused on a single product is less viable than applying integrated methods that allow the separation of a variety of commercial products from a process stream [98].

The Sea4value project proposes a multimineral brining process from SWRO brine, which consists of first calcium precipitation, followed by nanofiltration [101]. In brief, the nanofiltration concentrate is concentrated by membrane crystallization, where Mg can be generated, and after membrane crystallization an extraction/purification phase can remove Sc, V, Ga, In and Mo through polymer inclusion membranes, non-dispersive solvent extraction and ionic liquids. The permeate from the NF will undergo through a multi-effect distillation, and via solvent extraction, a polymer inclusion membrane, ion exchange resins or electrodialysis the following metals can be recovered: B, Li and Rb.

The mining processes usually consist of adsorption followed by desorption of metals. Ion exchange resins are mostly used as media. Adsorption has several advantages, including reduced energy consumption, cost, and preparation requirements [36]. More environmentally friendly adsorbents, including those based on natural materials, have been reported to remove lithium [102–105], cesium [106,107], strontium [108–110] and uranium [111]. Hence, these materials should be considered for the recovery of valuable metal elements from brine.

Brine mining is also common in precipitation. Researchers found an optimum operating condition of pH 10 and temperature at 90 °C for the precipitation of magnesium from SWRO brine, achieving a recovery efficiency of 78 % [48]. Under the same conditions, other minerals, such as lithium, boron, sulfate, calcium and strontium, were also recovered. Mg is recovered mainly in batch processes by precipitation/crystallization which allows a recovery rate of 95–100 % [33].

Even though brine mining can be less advantageous than mining effluent, good results have been reported in at the laboratory scale using SWRO brine, as shown in Table 2. The payback for the MgO and NaCl recovery system was calculated in eight years [96], and eleven years for NaOH-Cl_2. Remineralizing RO permeate using minerals recovered from brine has also been investigated [112], however presence of ammonia in the effluent can hinder the process. Lithium is another metal present in brine at concentrations that can be attractive to be produced. Lithium demand is expected to quadruple by 2040 [113] and 90 % of lithium production is not near high consumers of Li-ion battery manufacturers [96]. A total of 834 kg/year and 38.418 kg/year of uranium and lithium recovery potential from SWRO brine from India have been reported [114]. Fouling of the adsorbent by suspended particles or biological growth is the most important aspect influencing the practical use of the technology and the lifetime of the adsorbent [114]. Among the minor components that are most likely to benefit from the upgrade of SWRO extraction technologies, Cs, In, and Rb were identified as the most promising [115].

Membrane distillation-crystallization (MDC) is a promising technology for recovering clean water and valuable resources while treating hypersaline solutions, including SWRO brine [93]. However, MDC

Table 2
Valorization of SWRO brine reported in the literature.

Products	Methods	TRL	Brine location	Ref.
Boron	AD + DE	Lab	–	[68]
Boron	AD + DE	Lab	Simulated	[69]
Ca	Precipitation	Lab	–	[70]
Cs, Rb, Li, U	Adsorption	Lab	Barcelona, Spain	[71]
Cs, Rb, Li, U	AD + DE	Lab	Barcelona, Spain	[72]
Lithium	AD + DE	Lab	Qatar	[66]
Lithium	AD + DE	Lab	Not disclosed	[51]
Lithium and Strontium	AD + DE	Lab	Qatar	[37]
Mg^{2+} and UO^{2+}	CO_2 mineralization + AD + DE	Lab	Simulated	[73]
MgO	NaOH addition	Lab	Singapore	[74]
$Mg(OH)_2$, $Ca(OH)_2$ and NaCl.	Nanofiltration + Mg reactive crystallizer	–	Pantelleria (Italy)	[43]
$Mg(OH)_2$, $CaCO_3$, Cl_2 and H_2	Precipitation + Electrolysis		Singapore	[75]
$Mg(OH)_2$	AD + DE + magnetic separation	Lab	Ashkelon, Israel	[76]
Mixed salts	Brine concentrator + Brine crystallizer	–	Greece	[77]
Mixed salts	Membrane distillation + crystallizer	Pilot	Singapore	[78]
NaCl	Nanofiltration + Multi-effect distillation + (NTC)	–	Pantelleria (Italy)	[43]
NaCl	NF + RO + BC + BCr	–	Greece	[77]
NaCl	NF/RO + MCr	Lab	–	[79]
NaCl	Electrolysis	Lab	Pernambuco, Brazil	[47]
NaCl	IXM-ED	Pilot	Barcelona, Spain	[40]
NaCl	Modified IXM + ED	Lab	Simulated solution	[80]
NaCl	MDC	Bench	Calabria, Italy.	[81]
NaCl	NF-RO-MBC	Pilot	Saudi Arabia	[82]
NaCl	ED	Pilot	Barcelona, Spain	[83]
NaCl- rich brine Na_2SO_4, $Mg(OH)_2$	Selective electrodialysis (S-ED) Selective electrodialysis + crystallizer	Lab	China	[84]
NaOH and HCl	EDBM	Pilot	Lampedusa, Italy	[85]
NaOH and HCl	EDBM	Lab	Almeria, Spain[a]	[86]
NaOH And HCl + Mg^{2+}/Ca^{2+}, $CaCO_3$ and $Mg(OH)_2$	NF + Precipitation + EDBM	Pilot	Barcelona, Spain	[87]
NaOH	Electrolysis	Lab	Simulate based on brine from Gwangyang, Korea.	[45]
NaOH	NF + Ca(II) precipitation	Lab	Simulated based on brine from Fonsalia, Canary Islands	[88]
NaOH	ED-BMED	Lab	–	[89]
Na_2SO_4	MSF + Cr	Lab	Hong Kong	[90]
Na_2SO_4	F-SMDC	Lab	–	[52]
Potassium and Rubidium	SMSR + DE	Lab	Simulated based on brine from Perth, Australia	[50]
Strontium	AD + DE		Qatar	[91]
Rubidium	S-MD + AD + DE	Lab	Simulated	[92]

[a] Data not available.

research has been conducted only at the laboratory scale, and further studies at the pilot scale are necessary to develop the process for real scenario applications [93]. The main issues related to membrane-based methods for lithium extraction from brine were identified by [116]: insufficient development of membrane in process, efficiency and economic benefits and complex of brine content.

The treatment of RO concentrates with MDC resulted in the production of 17 kg/m^3 of NaCl crystals, representing 34 % c.a. of the total content of dissolved solids in the brine [81].

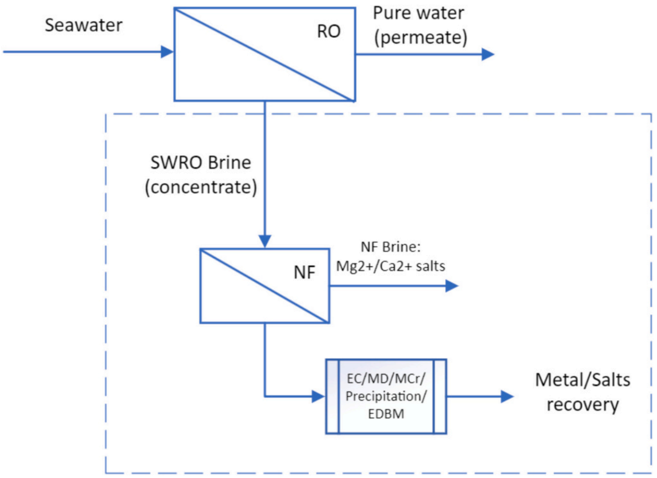

Fig. 4. General diagram of a seawater reverse osmosis brine valorization for mineral recovery (adapted from [87,100]).

Brine may also be used to obtain the chemicals HCl and NaOH, which are both used in desalination operations [117]. Many studies have reported the production of acids and bases from brine, such as NaOH and HCl, with NaOH being the second most common product extracted from brine, after NaCl (Table 2).

In a simulation study of SWRO brine treatment for NaOH production, the author found that placing NF before Ca(II) precipitation had lower CAPEX and OPEX than precipitation before the membrane system [88]. A review on NaOH extraction from SWRO brine identified bipolar membrane electrodialysis as the best treatment for meeting the techno-economic requirements of small-scale caustic production [118].

HCl and NaOH produced from SWRO brine can be combined with the extraction of other chemicals, such as Mg^{2+}/Ca^{2+}, $CaCO_3$ and $Mg(OH)_2$ as demonstrated by Reig. Et al. (2016) [87]. Their process consisted of an NF system to remove the divalent cation that would be applied in wastewater treatment plants for phosphorus recovery, followed by precipitation of the salts and an EDBM step for the recovery of chlor-alkali materials.

4.2. New approaches for brine valorization

4.2.1. Disinfectant (chlorine) production

Chlorine is an important product for a wide variety of industries, such as food and beverages, textiles, paper, water treatment, plastics, metallurgy, and pesticides [119]. Chlorine has risen in price in the recent decades, and its price is expected to increase as the shortage risk is imminent. Seawater and SWRO are rich sources of chloride by-products, including chlorine, and their production can help mitigate future shortages and price increases.

Approximately 97 % of the chlorine in the world is produced electrolytically from sodium chloride [120]. The production of on-site sodium hypochlorite in water treatment plants is a well-known solution for disinfecting supplies. This process is carried out by dissolving NaCl in water and using electrochemical cells to produce NaOCl as a subproduct. The species is pH dependent, with HOCl being the most active biocide and the most dominant occurring between pH 4 and 7. The ideal pH range for obtaining $HOCl^-$, the most biocide species, is between 5 and 7. NaOCl can also be produced by chemical methods using Cl_2 and NaOH as shown in Eq. (1); however, the product lacks the purity and stability required for certain industrial sectors [121].

$$Cl_2 + 2NaOH \rightarrow NaOCl + NaCl + H_2O \tag{1}$$

According to the electro-oxidation reaction, for each hypochlorite molecule (ClO) formed in solution, two electrons are consumed. With this information, it is possible to predict the energy consumption of the system, for different electron currents. Table 3 shows the energy consumption during NaCl production from the literature. A higher current and energy consumption are related to a high NaCl concentration. Nevertheless, NaCl concentrations above 150 g/L could be achieved using low current and energy consumption.

Chlorine products are used in most countries for disinfection in water treatment plants (WTPs) and wastewater treatment plants (WWTPs). Free residual chlorine (Cl_2 + NaOCl + HOCl + Ocl^-) might also be necessary in SWRO pre-treatment to prevent biofouling [9].

Electrochlorination is an electrochemical process in which an electrical current is applied to saline water via electrodes to produce a chlorinated solution via electrolysis. Electrochlorination can be performed with an unpartitioned or a partitioned electrochemical cell. The use of an unpartitioned cell is referred to as an 'open-cell', where the cathode and anode operate in a single chamber and the final product consists of a solution containing sodium hypochlorite (NaClO) and other chlorine species. An example of a partitioned cell is where the cathode and anode are separated by a membrane so that the electrolysis of Na + and Cl- occurs separately. This is the case for electrodialysis (ED), for which the main final product is chlorine gas (Cl_2). Both processes are discussed in more detail below.

4.2.1.1. *Open-cell electrolysis.*

The electrochemical production of chlorine-based products during open cell electrolysis occurs in a flow cell containing a set of electrodes (e.g., cathode and anode), which are connected to a DC power supply as shown in Fig. 5a [124]. A simplified overview of the desired reactions occurring within the open-flow cell during electrochlorination is presented in Fig. 5b.

The flow cell is fed with the NaCl solution, where the water molecules are electrolyzed at the cathode to form hydroxide and hydrogen gas (Eq. (2)):

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \quad (2)$$

The hydrogen gas moves upwards, while the formed hydroxides are repelled from the cathode surface and transferred into the bulk solution by diffusion and migration due to the electrical potential. Simultaneously, the chloride in solution is oxidized at the anode surface, forming chlorine gas according to Eq. (3):

$$2Cl^- \rightarrow Cl_2 + 2e^- \quad (3)$$

Chlorine gas is highly reactive and reacts with hydroxide after diffusion into the bulk liquid to form hypochlorite (Eq. (4)):

$$Cl_2 + 2OH^- \rightarrow Cl^- + ClO^- + H_2O \quad (4)$$

Hypochlorite forms an equilibrium with hypochlorous acid in an acid-base reaction (pKa = 7.55) according to Eq. (5) [124]:

$$ClO^- + H^+ \rightleftharpoons HClO \quad (5)$$

In addition, the Na + ions in solution react with hydroxide to form sodium hydroxide (NaOH) (Eq. (6)). The sodium hydroxide then reacts further with chlorine to form sodium hypochlorite (NaClO), which is the primary disinfectant product of the open-cell electrochlorination process (Eq. (7)). Combining the reactions results in the overall electrochemical reaction within the open cell as shown in Eq. (8) [125]:

$$Na^+ + OH^- \rightarrow NaOH \quad (6)$$

$$2NaOH + Cl_2 \rightarrow NaClO + NaCl + H_2O \quad (7)$$

$$NaCl_{(aq)} + H_2O + 2e^- \rightarrow NaClO_{(aq)} + H_{2(g)} \quad (8)$$

Table 3
Energy consumption x NaCl production reported in the literature.

Energy consumption (kWh/kg)	NaCl production (g/L)	Current density (kA/m^2)	Source	Ref.
0.12	185	0.35	SWRO Brine	[40]
0.19	203	0.50	SWRO Brine	[40]
0.160	174	0.27	Seawater	[122]
0.28	300	0.6	Synthetic RO brine	[123]

As already apparent from Eq. (8), the formation of chlorine species in the open cell is pH-dependent. Fig. 6 shows the pH dependency of chlorine species formation, where a low pH (i.e. ≤3) results in the formation of chlorine gas (Cl_2), while an acid to neutral pH (i.e. 3–7.55) favors the formation of hypochlorous acid (ClOH). A high pH (i.e. ≥7.55) leads to the formation of the conjugate base hypochlorite (ClO^-). The electrochemical reactions occurring during electrochlorination result in a pH gradient within the open cell, where a more acidic environment occurs near the anode surface, while a more basic environment exists near the cathode surface due to the formation of hydroxide ions.

Several advantages and disadvantages of open-cell electrochlorination are listed in Table 4 below. The main advantage of the open cell is the robustness of the system and its relatively simple operation [125]. However, the complex mix of reactions occurring simultaneously within an electrochemical cell results in a mixture of chemical species in the product solution, including parasitic reactions and the possible formation of halogenated by-products [124,126].

4.2.1.2. *Electrodialysis.*

Electrodialysis (ED) is a process that facilitates electrochemical reactions by utilizing the ionic mobility of solutes under an applied potential [125,127]. The application of ED for the production of chlorine products is known as a chloralkyl process. ED differs from open cell electrolysis in that the electrodes within the electrochemical cell are separated into two chambers with an ion-selective membrane, which allows for the occurrence of separate reactions at the cathode and anode (e.g., electrolysis and oxidation, respectively). An overview of the ED process and a simplified representation of the reactions within the chambers of the electrochemical cell are shown in Fig. 7.

The anode chamber is fed with the NaCl solution, while the cathode chamber is fed with water or a less concentrated NaCl solution. The ion-selective membrane allows for the transfer of sodium ions to the cathode chamber while retaining chloride ions in the anode chamber. Like in open cell electrolysis, water is electrolyzed to form hydroxide (OH^-) and hydrogen gas (H_2) (Eq. (2)), after which the hydroxide reacts with the sodium ion (Na^+) to form sodium hydroxide (NaOH) (Eq. (6)). Simultaneously, chloride (Cl^-) is oxidized within the anode chamber to form chlorine gas (Cl_2) (Eq. (3)), which is collected from the electrochemical cell and is the primary disinfectant product produced during ED. The hydrogen gas that forms in the cathode chamber is often released into the atmosphere. The remaining NaCl solution contains a lower concentration and can be recycled in the ED process to improve efficiency. Combining both reactions results in the overall reaction occurring within the ED cell (Eq. (9)):

$$2NaCl_{(aq)} + 2H_2O \rightarrow Cl_{2(g)} + H_{2(g)} + 2NaOH_{(aq)} \quad (9)$$

The produced chlorine and sodium hydroxide can either be used separately, as disinfectants and industrial alkali chemicals, or mixed after the ED process to form sodium hypochlorite as a disinfectant product similar to open cell electrolysis (Eq. (10)).

Another configuration of an ED cell includes a bipolar membrane, which allows for the separate production of an acid stream and an alkali stream. This process is known as bipolar membrane electrodialysis (BMED). Here, the concentrated NaCl solution is fed into the middle chamber, which is enclosed by ion-selective membranes as shown in Fig. 8. Water is fed into the acid and base chambers, which are separated from the cathode and anode by bipolar membranes to facilitate the dissociation of water hydroxide (OH-) and H+ ions. The hydroxide is then transferred to the base chamber, where it reacts with the sodium ions to produce sodium hydroxide similar to regular ED (Eq. (6)). The

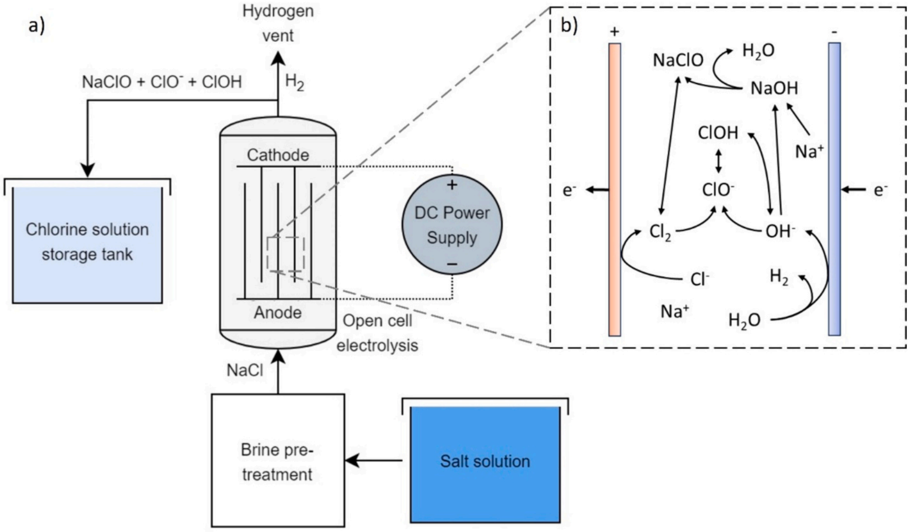

Fig. 5. Simplified representation of the open-cell electrochlorination process, with a) schematic of an open flow cell electrochemical process, and b) overview of (electro)chemical reactions occurring within the flow cell (adapted from [124,125]).

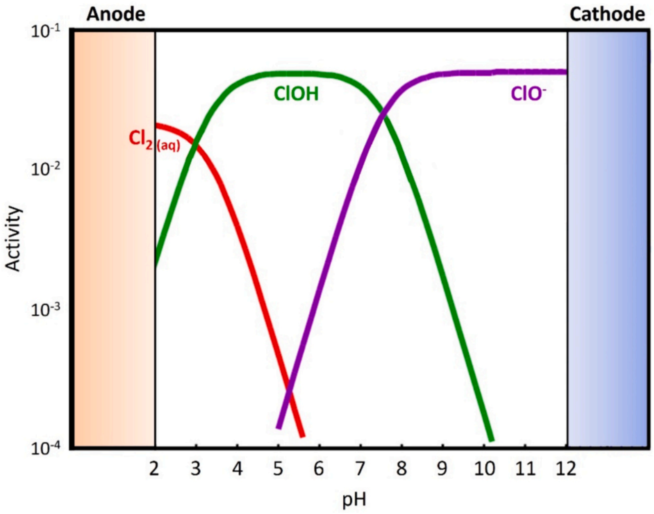

Fig. 6. pH dependency of chlorine species formation within an open cell electrochemical process.

Table 4
Several advantages and disadvantages of open-cell electrochlorination.

Open-cell electrochlorination	
Advantages	Disadvantages
Robust system	Requires specific process conditions
Ease of operation	pH-dependent reactions
Less prone to clogging	Mixture of chlorine species in product solution
Minimal waste stream	Chance of disinfectant by-product formation
	Chance of scaling and corrosion of electrodes

H+ ions are then transferred to the acid chamber to react with chloride to form hydrochloric acid (HCl) (Eq. (10)). Both sodium hydroxide and hydrochloric acid are used as industrial chemicals.

$$Cl^- + H^+ \rightarrow HCl \tag{10}$$

The main advantage of the ED system is the increased selectivity in the chemical reactions occurring within the chambers inside the electrochemical cells, which allows for the separation of the formed products [128]. However, the membrane is also highly sensitive to scaling and possibly biofouling, thus requiring frequent chemical cleaning and replacement, which is the major disadvantage of (BM)ED systems [129]. Table 5 lists several advantages and disadvantages of EDs.

4.2.1.3. Brine pre-treatment. Even though brine is a source of NaCl, the production of chlorine-based disinfectants can be challenging due to the necessity of pre-treatment for the electrochlorination process. While electrochlorination of seawater has already been applied at the industrial scale, the use of RO brine is a relatively novel concept and is still in the experimental phase [125]. The brine composition determines the pre-treatment requirements, which can turn the chlorine production technically or economically unfeasible.

As previously described, seawater contains several other compounds in addition to the NaCl required for electrochlorination, of which divalent ions (Ca^{2+} and Mg^{2+}) and (in)organic matter are the most prominent. These compounds can hinder the functioning and efficiency of the process and even lead to permanent damage of the equipment. The same is true for RO brine, since it consists of the same constituents but at higher concentrations, as well as of several additive chemicals applied during the RO process. One of the main problems in seawater and brine treatment processes, including electrochemical processes and thermal- and membrane-based desalination, is inorganic fouling, also known as scaling. Scaling is the deposition of dissolved salts and is often induced by changes in concentration, pH, temperature or pressure, which can cause incrustations on equipment surfaces, resulting in reduced efficiency or permanent damage [13]. In water treatment processes utilizing seawater, scaling is primarily driven by the divalent ions calcium (Ca^{2+}) and magnesium (Mg^{2+}), 'hardness salts', and sulfate (SO_4^{2-}), carbonate (CO_3^{-2}) and bicarbonate (HCO_3^-) [133–135]. The most commonly formed precipitates are calcium carbonate ($CaCO_3$), calcium sulfate ($CaSO_4$), magnesium hydroxide ($Mg(OH)_2$) and magnesium carbonate ($MgCO_3$) [136–139]. Deposition of these divalent salts is mostly favored under alkaline conditions ($pH > 8.5$). Other ions that can cause scaling include strontium (Sr^{2+}) and barium (Ba^{2+}) when combined with sulfate, as well as iron (Fe^{2+}) and manganese (Mn^{2+}) [134,138,140]. Another inorganic component of particular interest is mineral silica (SiO_2) and its tetrahedral compound silicate (SiO_4), both of which can form crystalline or amorphous structures [141]. In water treatment processes, silica deposition can cause both fouling and scaling on equipment surfaces. Silica is a particularly persistent sealant and is

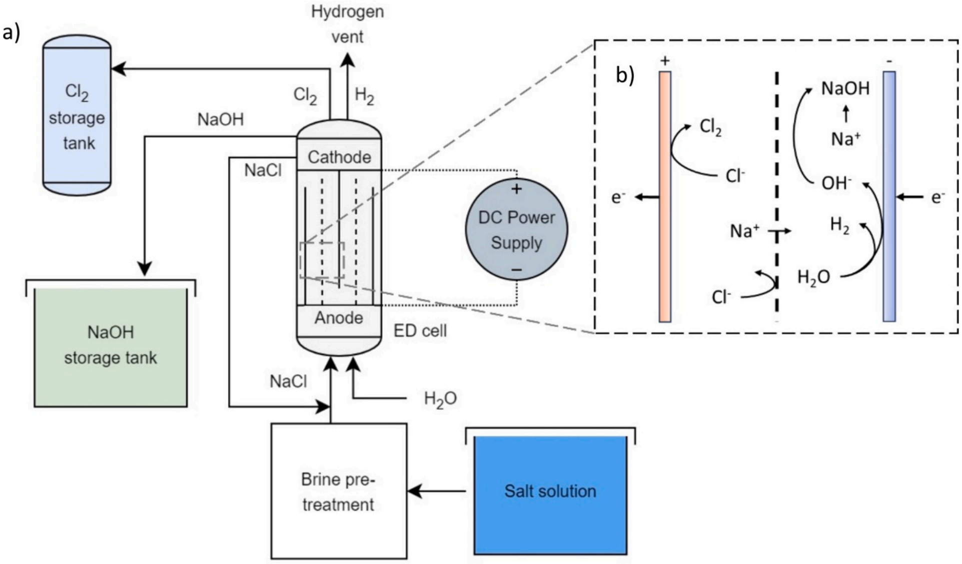

Fig. 7. Simplified representation of the electrodialysis (ED) process, with a) scheme of an ED electrochemical process and b) overview of (electro)chemical reactions occurring within the ED cell.

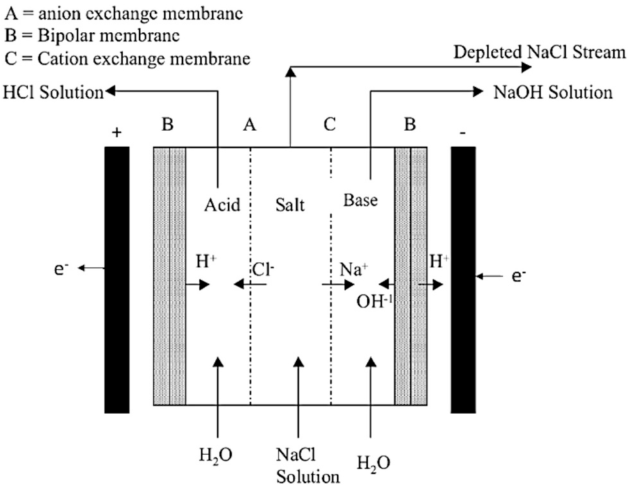

Fig. 8. Schematic overview of the bipolar membrane electrodialysis process (BMED) (adapted from [125]).

Table 5
Several advantages and disadvantages of electrodialysis (ED) [128,130–132].

Electrodialysis (ED)	
Advantages	Disadvantages
Selectivity of chemical reactions	Highly sensitive to scaling and (bio) fouling
Separation of formed products	Higher maintenance
Reduced formation of disinfectant by-products	Requires frequent membrane replacement
	Produces brackish waste stream

difficult to control with regular antiscaling agents due to its complex reactions, amorphous structure and formation of colloidal particles. The presence of divalent cations, magnesium and calcium in particular, catalyzes silica deposition and leads to insoluble metal-silicate scaling and accumulation [142–145]. The presence of organic matter induces fouling in water treatment processes and organic matter can react with chlorine species from electrochlorination to form toxic disinfectant by-products (DBP) [126,146]. Therefore, the removal of these constituents (e.g., divalent ions, (in)organic matter) from brine by pre-treatment is required to increase the efficiency, function and lifetime of the electrochlorination system [125,138]. There are several possible technologies for brine pre-treatment, such as nanofiltration, water softening and ion exchange resins. Sharkh et al. (2022), indicated that nanofiltration is the key pretreatment technology towards brine resources recovery [98]. The following section will focus on nanofiltration.

4.2.1.4. Nanofiltration. Nanofiltration is a membrane-based filtration technology applied for the removal of ions and organic matter from (saline) waters. The working principle of an NF is similar to that of an RO (see Fig. 9a). However, due to the pore size of the NF membrane, the process removes primarily divalent ions, organic matter and other colloidal particles while mostly passing through monovalent ions (e.g., Na^+ and Cl^-). Therefore, this process is considered a water-softening process, in which ions responsible for scaling are primarily removed (e.g., Ca^{2+}, Mg^{2+} and SO_4^{2-}) [127,147].

Membrane filtration technologies are categorized by the size exclusions of the molecules and particles provided by the membranes (see Fig. 9b). Among other membrane technologies, The NF membranes have pore sizes ranging from 1 to 10 nm, with only RO providing smaller pore sizes. Due to the passing of monovalent ions into the permeate during NF, the applied pressure required for the process is lower than that for RO [127]. In contrast to those of RO, other phenomena, in addition to size exclusion, occur at the membrane surface in NF: the separation mechanism is a combination of steric and electrical effects, depending on the size, electrical charge and solute concentration of the particle [101,148]. The main advantage of NF is that it is capable of removing a variety of molecules/particles within a single process while maintaining a low energy demand and high flux. However, like all membrane-based processes, it is also sensitive to scaling and fouling. Thus, a similar (chemical) cleaning regime and pretreatment of the feed of the membrane unit as applied in the RO process are often necessary to prevent membrane damage and maintain efficiency [147–149].

Although NF in seawater is not usually applied as a desalination technology [8], several studies have investigated the use of NF for RO brine treatment. M. Ali (2021) applied a low-pressure commercial NF system for the pre-treatment of RO brine, assessing the rejection of monovalent and divalent ions [149]. They found a TDS rejection of 53 % for SWRO brine and a decrease in total hardness (TH) of 97.2 %,

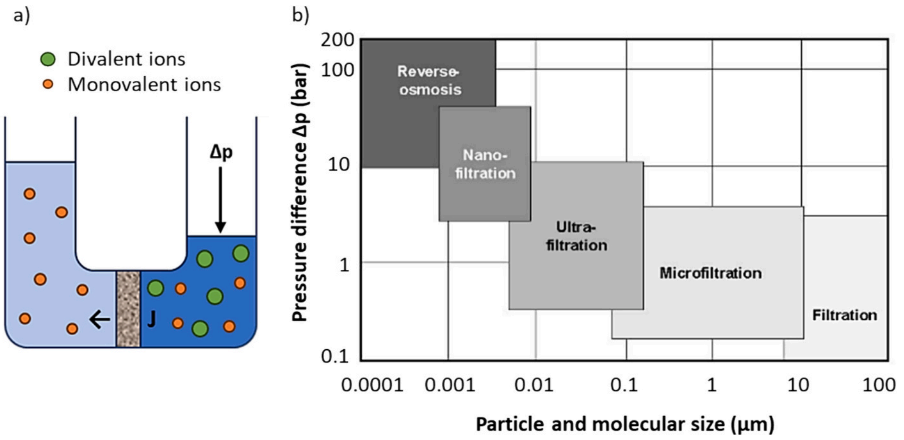

Fig. 9. The nanofiltration process, with a) the working principle and b) size exclusion and applied pressure difference for membrane filtration processes (adapted from [127].

demonstrating the high affinity for divalent ion rejection of NF. Later studies reported rejection rates of 98.35 %, 90.71 %, 54.11 %, 51.42 %, 45.65 % and 54.16 % for Mg^{2+}, Ca^{2+}, Cl^-, SO_4^{2-}, Na^+ and HCO_3^-, respectively, from seawater RO brine [149]. Figueira et al. (2023) studied the application of NF for SWRO brine as a pre-treatment step for multimineral brine extraction, and the necessity of Ca^{2+} removal before NF to avoid scaling [101]. These authors showed an efficient separation of divalent and monovalent ions, with rejections of Ca^{2+}, Mg^{2+} and SO_4^{2-} of up to 77 %, 87 % and > 95 %, respectively.

The need to concentrate brine and eliminate divalent ions, such as calcium (Ca^{2+}), magnesium (Mg^{2+}) and sulfates (SO_4^{2-}) [95], makes nanofiltration a pre-treatment option. Additionally, such systems are already commercially available for a range of industrial applications one of which is for the removal of divalent ions. Reig et al. (2014) reported that SRWO brines were oversaturated with various carbonates ($CaCO_3$, $MgCO_3$ and $CaMg(CO_3)_2$) but could be concentrated up to 4 times without risking the precipitation of NaCl. The presence of antiscalants from the RO process prevented the precipitation of carbonate and sulfate salts despite their oversaturation, benefiting the electrodialysis process. Moreover, the author suggested that the addition of HCl to maintain a constant pH of 5.5 ensured that most of the inorganic carbon was present as bicarbonate to prevent scaling [40].

In a study comparing three different NF membranes (DOW Filmtec NF90-4040, DOW Filmtec NF270-4040, and Toray CSM NE4040-40), the highest rejection of bivalent ions in NF was achieved by the last membranes [150]. In another study, PRO-XS2 from Hydranautics presented the highest selectivity between mono-and multivalent species, compared to Filmtec NF270 and Fortilife XC-N from DuPont [101].

The ion rejection of NF membranes was affected by variations in permeate recovery (from 0 to 70 %) in the following order: Fortilife XC-N > NF270 > PRO-XS2, with the last remaining virtually unaffected [88]. The ion rejection rates by NF membranes reported in the literature are presented in Table 6. As expected, NF presented a high rejection of divalent cations such as Ca^{2+} and Mg^{2+}. Novel NF membranes are being developed to enhance divalent cation rejections targeting applications such ZLD systems [151].

Table 6
Brine ion rejection (%) by NF membrane reported in the literature.

Feed	SWRO brine	BW-Brine	SW-Brine	SW-brine	SW-brine
Permeate recovery (%)	60	-	-	15	35
Pressure	20 bar	-	-	1.2 Mpa	30
Na^+	6	77.39	45.65	<5	20
Ca^{2+}	50	81.8	90.71	37.8	60
Mg^{2+}	71	93.3	98.35	87.8	75
K^+	5	-	-	<5	35
Cl^-	12	75.97	54.11	<5	30
SO_4^{2-}	91	97.82	51.42	100	-
HCO_3^-	45	89.21	45.16	-	-
Ref.	[43]	[149]	[149]	[152]	

* -: data not available.

4.2.2. *Salinity gradient energy*

Salinity gradient energy (SGE), also known as osmotic power or blue energy, it is a clean and sustainable energy source that is produced by combining two water streams with different salt concentrations [153]. SGE can be classified into three different methods depending on the power-generation mechanism: pressure retarded osmosis (PRO), reverse electrodialysis (RED) and capacitive mixing (CAPMIX) [154]. PRO and RED are membrane-based processes, while CAPMIX is electrode-based [30]. This pool of technologies that can be used to extract energy from salinity gradients provides different levels of energy recovery and which is the better suited solution depends strongly on the composition of the feed water.

Like the high-elevation water in pumped hydro, the high salinity of SWRO brine can be considered dense energy storage [155]. Theoretically, the SGE potential between seawater and fresh water is equivalent to 74 % of the world's electricity consumption [153], however, in practice, when considering the needs of other water uses, tidal fluctuations and the difficulties of having a clear separation of the salinity gradient between seawater and fresh water, the estimation is reduced to only 3 % of the world's demand [156].

RED has a similar working principle to ED, with a stack of alternating anion exchange membranes (AEMs) and cation exchange membranes (CEMs) creating alternating compartments of high salinity solution and low salinity solution. In the extremities of the stack, electrode compartments are placed and with the use of electrolyte solutions, redox reactions occur which allow the potential of moving ions to be harvested into electrical energy if an external load is connected (Fig. 10). The by-product of the process is somewhat a mix of the two feed solutions, with the high salinity solution becoming less concentrated than the original feed, while the low salinity solution becomes more concentrated [157]. However, the difference in concentration of the solutions after the process are not major, so it is unlikely that their classification into new categories of salinity change.

PRO, in the other way, has a similar working principle to RO, since electrical power is generated based on the hydrostatic pressure from the movement of water from a semi-permeable membrane between two compartments with different salinity solutions [158]. The semi-permeable membrane allows only the solvent (water) to pass through the membrane, retaining the salts, and due to osmotic power it moves in

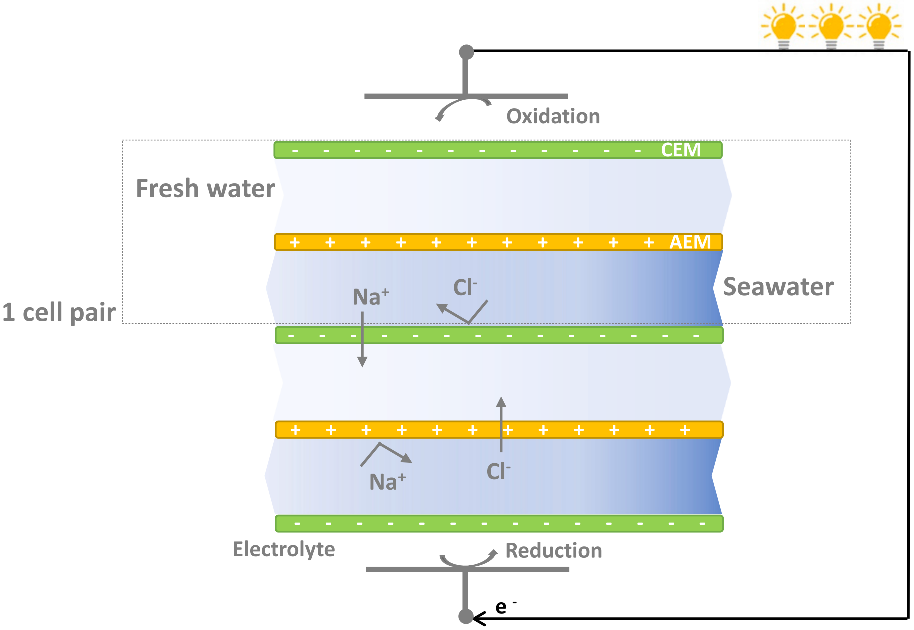

Fig. 10. Schematic representation of reverse electrodialysis principle.

the direction from a low salinity solution to a high concentration solution. The transport of water can be partially retarded by applying hydrostatic pressure to the concentrated solution, which results in a pressurized volume of transported water that can be used to generate electricity by coupling it with a turbine and generator. Concentration polarization is a known phenomenon of this process, that reduces the harvested energy.

A few studies have been performed the use of PRO and RED for energy recovery from brine, which indicates the possibilities and conditions that are needed to make these processes viable.

The use of PRO as a secondary energy recovery device was reported to reduce the energy consumption of SWRO desalination plants by approximately 20 % [159]. A tool for predicting the net energy recovery by PRO has been developed for designing and feasibility analysis of the process [160]. A framework proposed that combines RO-PRO was considered viable when the water price is <$1/m3 and the electricity is high (>$0.15/kWh) [155].

The use of brine streams In RED can be coupled either with seawater or less concentrated solutions, such as freshwater or treated wastewater. The energy recovery increases with the increasing gradient between the two streams; however, the energy efficiency of the process decreases, due to the higher resistance of the movement of ions in the low salinity stream [161]. The use of brine as a concentrated solution is likely to yield more power density than the more studied mix of seawater and fresh water, however, the volumes of water involved in these two cases are different, with the use of brine resulting in a lower capacity for production due to less volume being available for use in the process.

When using brine and brackish water, the presence of divalent ions was responsible for a significant reduction in RED efficiency [162]. Changes from laboratory-scale to real-world conditions can impact the overall efficiency of the technology. A 50 % reduction in performance was observed when RED testing was performed using real brackish water and brine compared to artificial solutions [163]. The authors attributed this to the presence of ions such as Mg^{2+}. Another known factor that can reduce the power output of RED systems is membrane fouling. A dual-media filter of anthracite and sand was proven to be a suitable and cost-effective solution for fresh and seawater pre-treatment in RED applications [157,164]. When using brines, the possibility of scaling also has to be considered.

Capacitive double-layer expansion is a technology that does not require membranes (CDLE) and works by contacting two porous “supercapacitor” electrodes with salt water charged by an external capacitor [165]. The challenges for real-scale CDLE application include the necessity of external power charging and energy leakage [156,166]. Table 7 shows the maximum power obtained in salinity gradient production reported in the literature.

The high-concentration brine types investigated for blue energy production were seawater (SW), either synthetic or real, seawater reverse osmosis (SWRO) brine, brine from multi-stage flash distillation (MSF), forward osmosis (FO) brine and NaCl solutions. The low-concentration brines were seawater, treated wastewater (WW), river water (RW) and NaCl solutions. The maximum power densities reported in the literature were 6.01 and 5.33 W/m^2 for a RED and a CDLE system, respectively.

A potential solution to lower the energy costs of high-consuming SWRO systems and valorize brine is salinity gradient energy generation. However, in each case the choice among seawater, river water, or treated wastewater should be evaluated since it demands a large volume of low-concentrated water.

Table 7
Applied techniques for salinity gradient energy production.

Method	Brine (high and low concentration)	Maximum powers obtained (W/m^2)	Ref.
CDLE	Synthetic SW and RW	5.33	[156]
PRO	SWRO brine and SW	1.97	[167]
PRO	Brine from MSF and treated WW	2.2	[167]
RED	RO brine and RW	1.48	[154]
	FO brine and RW	1.86	
RED	SW and treated WW	0.76	[168]
RED	SWRO brine and treated WW	0.54	[169]
RED	SWRO brine and treated WW	0.46	[170]
RED	SW and RW	1.43	[171]
RED	5 M and 0.5 M NaCl solutions	6.01	[172]
RED	0.55 M and 0.02 M NaCl solutions	1.825	[173]
RED	SW and RW	0.35	[174]

5. Comparative analysis

This section aims to compare the three brine valorization solutions, brine mining, disinfectant productions and salinity gradient energy production, according to the following challenges: a) environmental impact, b) technological consolidation and c) commercial appeal.

a) Environmental impact

It is undiscussable that the production of water through SWRO can improve the quality of people's life and reduce water stress in many countries. However, its impact on the environment is still underestimated. It was only in 2023 that an LCA from SWRO included the effects of brine discharge [175]. The findings revealed that the eutrophication impacts were mainly due to brine rejection. In a comparative study between desalination, wastewater reuse and rainwater harvesting as water sources, the SWRO achieved the highest value in 12 of the 15 environmental impact categories [176].

Final brine disposal must be carefully planned because brine is the most relevant waste contributing to the environmental impact of a desalination plant [177]. Three brine disposal scenarios were compared, direct disposal, dilution with WWTP effluent and dilution with seawater [177]. The results showed that each solution might present advantages and disadvantages depending on the local necessity, but the best option is to dilute the solution with treated WW when there is no reuse of the effluent and when the WWTP is close to the desalination facility. Chronic effects of brine discharge from SWRO facilities have been identified in benthic microbial communities [19]. The effects are mainly changes in bacterial abundance and activity; nevertheless, the impacts were concentrated in a region <1.4 km^2 from the discharge point.

In addition to brine discharge, the high energy consumption is responsible for most of the environmental impacts of SWRO ([175,176,178]. Thus, ways towards energy efficiency are also necessary to reduce the carbon footprint of SWRO plants, combined with brine management methods. SWRO plants can rely upon an energy recovery system from brine, either from pressure or from the salinity gradient. Other alternative energy sources, such as solar and wind power [179], might be also be important for reducing energy consumption and increasing the feasibility of the overall desalination process [180]. An environmental sustainability assessment of SWRO combined with EDBM showed that photovoltaic solar energy can reduce the carbon footprint by up to 83 % [117].

A sustainable brine management framework was proposed by Gil-Trujillo and Alonson (2023) [3]. The SGE and the circular economy (and brine valorization) were among the main indicators, although the others could also be related, such as the use of renewable energies, applied research on the brine effect on marine ecosystems, and minimization of brine discharge in the marine environment.

Generally, use of brine for salinity gradient generation is the approach with less environmental impact The brine mining and disinfectant production approaches can produce a more concentrated brine, a more complex effluent which can be associated to a higher environmental impact. Using salinity gradient energy generation associated to one of these approaches can reduce its impact.

b) Technological consolidation

Brine valorization is a sustainable alternative for SWRO concentrate and can help to reduce the environmental impacts of desalination. However, further research is needed to develop cost-effective and sustainable brine valorization methods that can be implemented on a large scale. Additional data on brine valorization is essential to enhance the existing database on the subject and improve decision-making at both research and policy levels.

Technological innovation in brine valorization is challenging due to the different composition of each brine and location of each plant, which is associated to the legislation to comply with. Authors have suggested a general pretreatment approach to simplify the nature of the brine matrix and hence, create opportunities for investment in novel technologies [181]. Most of the studies reported in the literature presents solutions with low Technological Readiness Levels (TRLs), being carried out with synthetic brine of/and in laboratory and pilot scales. The quality of the recovered minerals frequently does not exceed market standards, making ZLD commercially impractical.

Future work should take into consideration the integration of multiple brine valorization approach, such as disinfectant production or SGE generation, which have already commercial scale, to achieve a feasible solution for SWRO concentrate management.

The likelihood of technological advancement is high. As the SWRO industry expands, brine management and subsequent valorization technology will also advance. This could also result in a general decrease in the price of SWRO-desalinated water and improve the accessibility of the technology to developing nations [58].

c) Commercial appeal

Disinfectant production through NaCl is an already commercially mature technology, its production from SWRO brine is still a challenge due to its composition, which imposes a complexity in the process that needs more investigation to turn it feasible.

SGE generation is already commercially available, but not yet using brine as higher salinity effluent. Brine energy harvesting may not yet be cost-competitive with other renewable resources, but by identifying complementary uses for this technology, the commercial stage can advance faster [182].

Commercial operations are constantly shadowed by scaling and fouling, which can increase cost, energy consumption and chemical usage. Fouling can reduce the efficiency of energy recovery over time and requiring pre-treatment which can increase costs [183]. Emerging developments in membrane antifouling technologies should improve the operation of brine-fed membrane systems in the future [184]. A study on membrane fouling using SWRO as feed showed that $CaCO_3$, $CaSO_4$ and NaCl were the most significant inorganic foulants, but the addition of NaOCl to the pretreated feed solution could mitigate fouling and wetting [185].

Moreover, antiscaling methods should be constantly improved, as they will impact the costs of brine treatment (and consequently the overall costs of SWRO) and reduce environmental impacts, such as the reported increase in salinity resulting from the deposition of polyphosphonate-based antiscalant [186].

Another hindrance in bringing these solutions to the market is the distance of the product to from the final user, which can make it unfeasible due to the high costs of transportation. The SGE is less affected by that as electricity can be easily transmitted compared to chlorine and salts products, for example.

6. Final considerations

Kurihara and Takeuchi (2018) suggested that there are three major challenges to developing sustainable desalination technologies for the 21st century: (1) energy savings, (2) low environmental impact, and (3) low water production costs [187]. Almost a quarter of the 21st century has passed, and brine valorization is one of the keys to addressing these challenges, as it can provide energy production through the use of the salinity gradient, reduce environmental impact by decreasing brine volumes and salt concentration, and ultimately help reduce total process costs by adding commercial value to its subproducts. Consideration of sustainable brine management solution has pointed out that any solutions will have to consider operational complexity, life cycle, energy consumption and chemical usage, and stakeholders [181].

Table 8 summarizes the discussion in the previous section in the form of a SWOT analysis. The strengths, weaknesses, opportunities, and

Table 8

SWOT analysis of SWRO brine valorization (Disinfectant production – DP, Brine “mining” – BM, Salinity Gradient Energy – SGE).

S – Strengths	W - Weakness
• Aligned with SDGs (DP,BM,SGE) • Renewable source (DP,BM,SGE) • More sustainable approach for brine management (DP,BM,SGE) • Reduce carbon footprint of SWRO (DP, BM, SGE) • Integration into Minimal and Zero Liquid Discharge (MLD/ZLD) solutions (DP,BM) • Shifting the locations of metal production closer to those of consumption (BM) • Reduce the energy consumption of the SWRO systems (SGE) • Well-known technology (DP) • Possibility of power generation 24/7 [188] (SGE).	• Most of the research was carried out on lab and pilot scale and operational data on large-scale facilities are limited (DP,BM,SGE) • Generation of a more concentrated brine (DP,BM) • Cost competition with other renewable sources (SGE) • High volumes of non-salinity water source required (SGE)
O – Opportunities • Increase the use of desalinated water as a water source (DP,BM,SGE) • Increase the search for ZLD solutions (DP,BM) • Critical metal supply in the future [189](BM) • Decarbonization policies worldwide (SGE) • Use of renewable energy for water production (SGE) • Research work fronts including. SMART brine, ZERO BRINE, SEA4VALUE, WATER-MINING, Sea4life and SEArcularMINE [190] (DP,BM, SGE)	T – Threats • Market competition with traditional sources (DP,BM,SGE) • Low market costs of disinfectants (DP) • Acceptance by the law of brine discharge in the sea (DP,BM,SGE) • Supply and price variations in energy market (SGE) • No specific legislation applied to enforce brine valorization (DP, BM, SGE).

threats regarding the proposed alternatives for SWRO brine valorization, disinfectant production (DP), brine “mining” (BM), and salinity gradient energy (SGE) are pointed out. This approach helps to identify brine valorization nowadays, as well as future challenges and research opportunities.

Nine strengths and six opportunities were identified, surpassing the four weaknesses and five threats identified. Nevertheless, it is important to highlight that quantifying and comparing these benefits and obstacles can be difficult due to the lack of information.

7. Conclusions

SWRO brine is a valuable reserve of resources but its full potential has yet to be explored. Due to its complexity and lack of economic interest, transforming it into commercial products remains a challenge. This study reviewed the literature on the production of disinfectant, metals/chemicals and energy from SWRO brine, all considered to have a higher chance of profitable production in the future.

It is clear for the authors that the TRL of the solutions presented in the current available literature is still low, which confirms the lack of economic interest in brine valorization solutions. The studies conducted by the authors indicate that the large volumes of brine produced by SWRO plants requests combined solutions. Brine mining, the most explored solution in literature, is promising, however combining it with disinfectants productions through electrochemical systems and energy generation might increase economic viability and realistic use of total brine volume. However, both solutions also require the implementation of pretreatment steps. Nanofiltration is a promising pretreatment for both chlorine production and brine mining, thus more research towards its improvement and application to SWRO brine characteristics should be addressed.

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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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