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A review of strategies for RO brine minimization in inland desalination plants

Javier Rioyo[a,*], Vasantha Aravinthan[a], Jochen Bundschuh[a], Mark Lynch[b]

[a]*University of Southern Queensland, School of Civil Engineering and Surveying, Faculty of Health, Engineering and Sciences, West Street, Toowoomba, Queensland 4350, Australia, Tel. +61 7 4631 2100; emails: javier.rioyorumayor@usq.edu.au (J. Rioyo), vasanthadevi.aravinthan@usq.edu.au (V. Aravinthan), jochen.bundschuh@usq.edu.au (J. Bundschuh)*

[b]*University of Southern Queensland, School of Agricultural, Computational and Environmental Sciences, Faculty of Health, Engineering and Sciences, West Street, Toowoomba, Queensland 4350, Australia, Tel. +61 7 4631 2100; email: mark.lynch@usq.edu.au*

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ABSTRACT

Water scarcity in many inland areas is increasing the demand for new groundwater desalination plants. Co-produced coal seam gas (CSG) water (or coal bed methane as known in the USA), which is mostly brackish, is extracted in huge quantities during CSG production and requires advanced treatment. Reverse osmosis (RO) is the leading technology applied in municipal desalination and for treating CSG water in Australia and in some locations in the USA. Antiscalants are often dosed during RO pretreatment to prevent membrane scaling. Recovery rates are limited by antiscalant efficacy and large volumes of brine are frequently disposed of in evaporation ponds. The search for environmentally friendly methods for RO brine minimization is considered as a key global issue. In this paper, differences between inland and seawater desalination are highlighted. The existing technologies for RO brine minimization and zero liquid discharge (ZLD) for inland desalination are reviewed. The efficacy and application of two scaling reduction technologies for RO brine minimization: (i) acid/antiscalant addition and (ii) 'high pH precipitation treatment' are compared. Finally, more complex ZLD and volume reduction systems, such as the high efficiency RO (HERO™) and the SAL-PROC™, are analyzed as well.

Keywords: Reverse osmosis in inland areas; Brackish groundwater; Coal seam gas water; Brine minimization; Zero liquid discharge

1. Introduction

As freshwater supplies diminish, desalination of brackish groundwater resources is becoming an increasingly viable option for inland communities in countries that have limited access to fresh surface water supplies or desalinated seawater to meet increasing demand [1,2]. Reverse osmosis (RO) is the dominant, widely adopted, affordable technology in municipal desalination in comparison with thermal desalination. However, brine disposal is one of the biggest drawbacks of this technology especially for inland areas that have very limited options [1–3].

Conversely, coal seam gas (CSG) or coalbed methane is an important energy resource in many countries like United States, Australia, China, Canada and India [4–6]. Large amounts of brackish groundwater are co-produced during gas production [7,8]. It is estimated that about 300 GL/year of CSG water could be produced over the next two decades in Australia alone [4,9]. This large-scale extraction of typically brackish groundwater associated with CSG exploitation has created concerns over potential adverse effects on groundwater resources and arable land. In Australia, the simple storage of co-produced CSG water is no longer permitted [6,10], and advanced treatment by RO is considered the best available technology [6,11]. In the USA, deep well reinjection is very common [10] although ion-exchange technology has been also applied for reducing the concentrations of sodium and bicarbonate [12]. In China, CSG water is mainly managed by surface impoundments and evaporation [13].

Overall, RO brine management remains as a significant challenge in co-produced CSG water desalination when the

* Corresponding author.

huge quantities involved and environmental risks of storage are considered [4,8,11,14,15].

Since both inland municipal and CSG water treatment facilities cannot dispose of RO brine into a large body of water, such as the ocean, they operate at high recovery rates (R_w) to minimize the volume of brine to be discharged. Previous studies have reviewed the state of the art technologies for RO brine minimization [16,17]. Among these technologies was the use of two or more RO stages to further increase the R_w [2,18].

Considering that RO is a membrane separation process, the consequences of high R_w on the membranes are increased energy demand due to the increased osmotic pressure required corresponding to the increase in total dissolved solids (TDS), reduced permeate quality, and increased scaling. Increased concentration also leads to increased scaling. Some constituents present in brackish groundwater will precipitate if the concentration product of the salt formed exceeds its solubility product (K_{sp}). Sparingly soluble salts that can scale the RO membrane are $CaCO_3$, $CaSO_4$, $BaSO_4$, $SrSO_4$, CaF_2 and SiO_2 (silica) [19]. Scaled membranes require higher than normal operating pressure, and chemical cleaning is required to minimize the risk of irreversible scaling [20], which leads to reduced membrane lifetime.

Different technologies, such as adsorption, precipitation or ion-exchange, can be applied between consecutive RO stages to prevent scale formation and increase R_w. However, further research is needed to improve the efficiency [2]. Among other technologies, an intermediate 'high pH precipitation treatment' allows the removal of undesirable precipitated species from primary RO brine by physical separation processes.

However, not all the brine can be concentrated by applying multistage RO systems. Once the risk of membrane scaling is overcome, practical restrictions to the osmotic pressure become the limiting factor. RO technologies allow brine concentration to reach 65,000–75,000 mg/L TDS. After this limit is reached, more advanced and expensive concentration technologies are required [21].

High recovery systems can achieve recoveries above 92% depending on the feed water composition [21]. However, even if brine is concentrated and reduced in volume, the final disposal of this reduced volume remains very difficult for inland plants with the focus on avoiding contamination to inland aquifers and other environments. This has led several researchers to investigate the development of zero liquid discharge (ZLD) applications where brine discharge is not possible. In this case, reject brine is no longer considered as a waste but as a resource from which useful dry salts, metals and desalinated water can be recovered so there is no discharge of liquid waste from the treatment facility. The technologies available aiming at ZLD are expensive and the most common approach in municipal desalination involves the following steps: primary RO system, intermediate treatment of RO concentrate to reduce its precipitation potential, secondary RO system, thermal desalination and evaporation ponds [2]. Other conventional processing technologies include thermal crystallizers, spray dryers and landfills [21].

Overall, RO brine minimization, including possible salt recovery, aiming for ZLD is a significant economic challenge for inland desalination facilities. Further research is needed to minimize capital (Capex) and operating (Opex) expenditure in ZLD processes [2,8,21,22]. This paper aims to (a) review the differences between seawater, brackish and CSG water desalination and the challenges faced; (b) critically review the existing technologies for minimizing the brine volume and ZLD in inland desalination; (c) highlight the scaling potential in these technologies; (d) provide insights into strategies integrating an intermediate 'high pH precipitation treatment' with another concentration system for RO brine minimization for groundwater supplies; and (e) analyze more complex ZLD and volume reduction systems like the high efficiency RO (HERO™) and the SAL-PROC™.

2. Differences between seawater, brackish groundwater (municipal desalination plants) and co-produced CSG water desalination

2.1. Comparison of the quality of the source waters

As shown in Figs. 1 and 2, seawater has a similar composition worldwide [23]. Chloride is the predominant ion followed by sodium, sulphate, magnesium, calcium and potassium. The standard TDS concentration in seawater is 35,000 mg/L [23]. In general, desalination costs are influenced by ocean salinity and temperature [24]. Boron removal is complex [25]. Its concentration in seawater is about 4.8 mg/L. Boron content can be reduced below 0.5 mg/L by combining RO and ion-exchange technology [26] or applying multipass RO with pH adjustment [27]. The R_w in seawater desalination is limited by osmotic pressure, energy consumption and allowable salinity/boron concentration in the RO permeate [23]. On the other hand, RO concentrate disposal is not a problem, as brine can be discharged back into the ocean with the pumping system and length of piping key factors in the design process [25]. Seawater desalination plants are often configured with one or more RO passes depending on different factors, such as boron concentration, R_w, energy costs, and product water standards [25]. Scaling is not considered as a limiting factor [23] although seawater membranes can be fouled by organic and particulate material [28]. Seawater desalination plants working with open intakes are also prone to biofouling [23]. About one-third of feed seawater

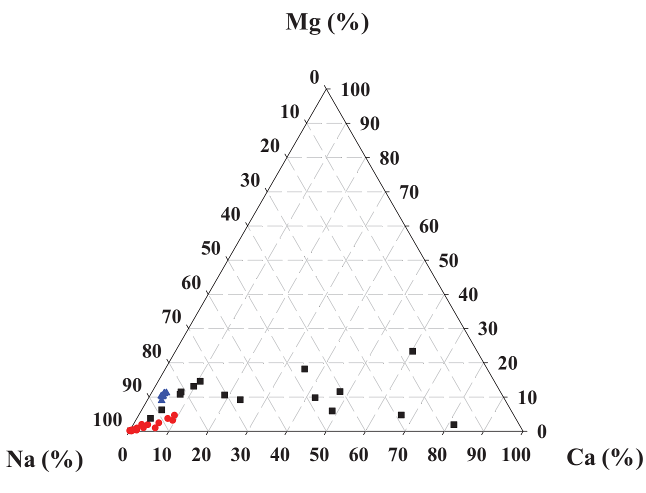

Fig. 1. Relationship (%) between significant cations present in seawater (blue colour), brackish groundwater (black colour) and co-produced CSG water (red colour).

is recovered as permeate while two-thirds is discharged as RO brine with a TDS level of about 52,000 mg/L. The costs of desalinating seawater in Australia and the USA are estimated at US$1.5–2.3/m³ [11] and US$1.54–2.43/m³ [29], respectively. The use of energy recovery devices in seawater desalination is common for reducing overall operating expenses. Efficiencies up to 65% were previously reported [30]. (Note: Quality data to complete Figs. 1 and 2 represent worldwide characteristics and was collected from different books/technical papers [3,9,11,23,31–44] and analytical records supplied by the Western Downs Regional Council (Australia) from different municipal groundwater wells.)

In contrast, brackish groundwater has a lower TDS level ranging from 1,000 to 10,000 mg/L [45]. Its chemical composition varies widely depending on different factors like the host-rock type, the nature of overlying soils and rainfall [1]. It typically has low particulate or colloidal contaminants and low organic carbon content while silica and boron concentrations can vary significantly. Therefore, and in contrast to seawater, precipitation of carbonates, sulphates and silicates, and hence membrane scaling can be problematic. Precipitates of $CaCO_3$, $CaSO_4$, $BaSO_4$, $SrSO_4$ and silicates can become limiting factors for desalination. R_w levels in municipal facilities vary from 75% to 90%, and the ratios of calcium/TDS, carbonate/TDS or sulphate/TDS are higher than in seawater desalination [25]. Further RO brine minimization is required. Brine management costs are also higher in inland desalination compared with seawater desalination [16]. A design including a single stage system with an additional module connected to treat RO brine is considered the best option both economically and environmentally by some researchers as shown in Fig. 3. This configuration increases recovery and minimizes operating costs [46]. Depending on feed water salinity, single stage RO systems are typically sufficient for recovery of around 40%–60% freshwater, while two stage RO systems can increase R_w up to 80% [23]. Due to lower salinity (<10,000 mg/L) and osmotic pressure of the brackish feed water, the first stage in a two stage system commonly operates at high flux but low pressure (up to 4.1 MPa) [47]. In this case, brackish water membranes are selected during the design process [23,46]. Brine generated is then treated in an additional RO stage at higher pressures (up to 6.9 MPa) [47], due to the higher osmotic pressure required when the TDS concentration and salinity are higher. Seawater membranes are often selected for the second RO stage [46] due to their potential to provide higher salt rejection (R_s) while treating high salinity feed water of up to 50,000 mg/L TDS [23].

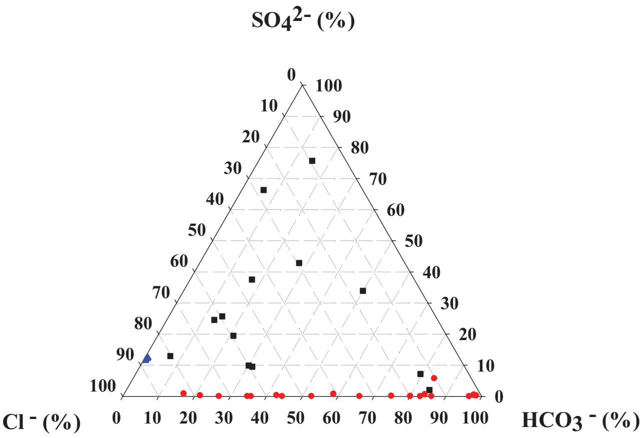

Fig. 2. Relationship (%) between significant anions present in seawater (blue colour), brackish groundwater (black colour) and co-produced CSG water (red colour).

Co-produced CGS water in Australia generally has TDS levels that range from 300 to 10,000 mg/L [48]. In the Rocky Mountains region of USA, the TDS content varies from 150 to 39,260 mg/L [38]. Overall, CSG water quality can vary significantly even between wells in close proximity [6,39]. As shown in Fig. 2, co-produced CSG water primarily contains NaCl and $NaHCO_3$ [7,8,14]. Sulphate concentration is low. CSG water composition is a result of different biological and geological processes that have taken part in the formation of CSG [37].The pH level and sodium adsorption rate can be high. In the Rocky Mountains, values of 9.26 and 452.8 mg/L have been recorded [38]. In Australia values of 9.1 and 567 mg/L have been observed [9]. Co-produced CSG water may also contain hydrocarbons or saturated gases depending on the well source [49]. Variable amounts of aluminium, silica, barium, calcium, magnesium and fluoride can also be present. Boron concentrations up to 4.7 and 3.1 mg/L were recorded in the USA and Australia, respectively [9,38]. In general, modular RO plants for co-produced CSG water treatment are designed in a multistage configuration [11,39] and R_w levels around 75%–80% can be easily achieved [6,14,39,49,50]. In the USA, generated RO brine is frequently managed by deep-well injection [12]. In Australia, RO brine is often placed in evaporation ponds while alternative options for beneficial uses are explored [6,8,14,48,49]. Layers of clay and synthetic membranes are required in pond construction to prevent contamination of groundwater aquifers [10]. The design of

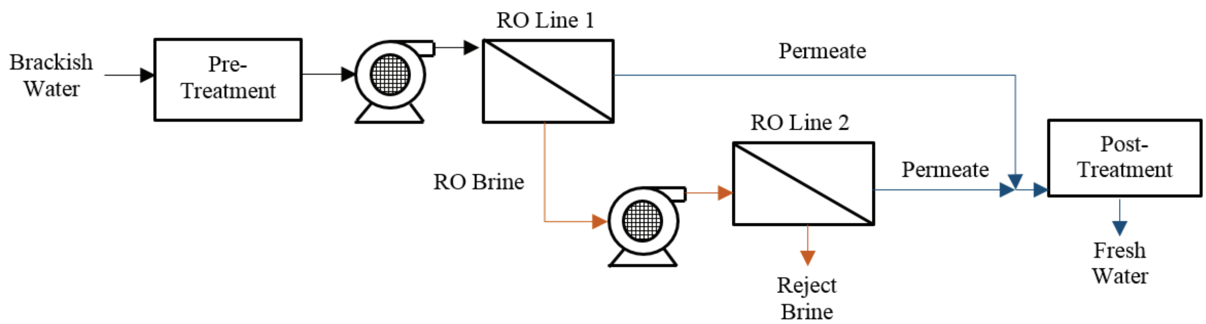

Fig. 3. Multistage RO system with intermediate pumps.

Table 1
Differences between seawater, brackish groundwater (municipal facilities) and co-produced CSG water

	Seawater desalination	Brackish groundwater desalination	Co-produced CSG water desalination
Water quality	Chloride is the predominant ion TDS = 35,000 mg/L [24]	Chemical composition is variable TDS = 1,000 to 10,000 mg/L [45]	Bicarbonate is often the dominant anion species [9,14] TDS = 300 to 10,000 mg/L [9,48]
Rw during RO desalination	35%–50% [25]	75%–90% [25] 65%–85% [21]	75%–80% [6,14,39,49]
RO brine disposal methods	Discharged back into the ocean [25]	Evaporation ponds, surface water discharge, discharge to wastewater treatment, subsurface injection and land application [21]	In Australia RO brine is temporally disposed of in evaporation ponds while other options are studied [6,8,14,48–50]. In the USA, deep well injection is common [8,12]

a CSG brine pond generally includes two separate lining layers and a monitoring system [14]. Table 1 summarizes the general quality characteristics, achievable R_w and existing RO brine disposal methods for seawater, municipal brackish groundwater and co-produced CSG water desalination.

2.2. Pretreatment options in RO desalination

Seawater pumped from the ocean needs to be pretreated to remove suspended solids and other matter in order to avoid membrane fouling. Worldwide, conventional pretreatment in RO plants often includes the addition of coagulant, pH adjustment, media filtration, cartridge filters, disinfection and final antiscalant addition [24,51]. Although $CaCO_3$ precipitation is possible, it is not likely to occur due to the low R_w expected in most seawater desalination plants [25].

Brackish groundwater sources for drinking purposes in municipal systems have less fouling propensity than surface water with consequently less extensive pretreatment needed prior to RO. Depending on the microorganism content, disinfection may be necessary [52]. Total hardness reduction by ion-exchange technology can be limited by water chemistry and chemical regeneration costs [53]. Conventional pretreatment in municipal facilities often combines media filtration with acid or antiscalant addition to prevent scaling. Antiscalants retard the precipitation of sparingly soluble salts and promote supersaturation [19]. Commercially available antiscalants are relatively cheap [54] and work by inhibiting crystal formation or by surface modification of the crystals [20]. However, it is important to mention that antiscalant addition during RO pretreatment can have adverse effects in a later brine treatment that uses precipitation to remove potential scale forming minerals. In fact, $CaCO_3$ precipitation from RO brine can be reduced and filtration performance decreased [55]. Moreover, an excessive dose of antiscalants during RO pretreatment can increase the risk of biofouling [56,57].

Co-produced CSG water can vary in quantity and composition, and so requires a reliable and flexible pretreatment process. Since the wells are not cased, co-produced water with high suspended solids is discharged into the pond from where the water is sourced for RO pretreatment. Algae formed in the feed pond and dissolved organic compounds from the fracking process or the coal seam also need to be removed before the desalination process [11]. Reduction of total suspended solids is essential since a silt density index lower than 3 is required for the desalination process. The use of suitable coagulants and flocculants for solid–liquid separation (clarification process) requires screening work and later sludge management [49]. Particle separation technologies frequently used during CSG water pretreatment are dissolved air flotation and microsand ballasted flocculation [7]. Lime softening or weak acid cation resins can also be used for CSG water softening before RO desalination [4]. Variable levels of fluoride, silica, barium, sulphate and calcium render the treatment process difficult and may require the use of specific antiscalants [49]. Disinfection can be achieved by chlorination/dechlorination [49] or ozone addition [4]. Recently, the use of containerized treatment plants combining microfiltration for suspended solids removal with spiral wound RO membranes has shown to be a cost-effective and reliable solution for CSG water treatment. These mobile treatment plants can be automatically controlled via a programmable logic controller [58].

3. ZLD and RO brine minimization technologies in inland desalination plants

3.1. ZLD concepts, brine minimization technologies, and challenges in municipal desalination

High recovery systems aimed at brine minimization have been defined in municipal desalination as those systems achieving recoveries higher than 92% [21]. ZLD is defined as a high recovery system allowing that no effluent leaves the ground-level plant boundary. In a ZLD approach, all the brine is either recovered by a combination of technologies to produce desalinated water or dry salts. Technologies commonly recommended in ZLD processing systems include: RO, vacuum evaporators, crystallizers, evaporation ponds and spray dryers. Salinity and composition of the brine to be processed in the ZLD system has a substantial influence on capital and operating costs. Sequential and selective removal/recovery of salts from concentrated brine should follow the sequence shown in Table 2, from low to high solubility levels. Although technically feasible, high recovery and ZLD systems are currently not economically viable in municipal desalination [21].

Table 2
Sequence for salt recovery/removal in a ZLD process

Salt solubility level	Salt	
Sparingly	$CaCO_3$	Salt removal/recovery Sequence
	$CaSO_4$	
	$Mg(OH)_2$	
Moderately	$Na_2(CO)_3$	
	$Na_2(SO)_4$	
Soluble	NaCl	
Highly	$CaCl_2$	
	$MgCl_2$	

As previously stated, membrane desalination is considered to be the predominant technology to be used in municipal desalination [2] and ZLD systems. Operating costs are reduced by applying a system including consecutive RO stages for brine minimization. Fig. 3 shows a 'two-stage RO system' with modules connected to reject brine with booster pumps. By implementing 'seawater membranes' in the second stage, R_w increases and additional permeate output can be achieved [52]. The osmotic pressure depends on the concentration of dissolved salts in solution [31]. Due to the lower salinity of the brackish feed water, 'RO line 1' operates at a lower pressure. The brine generated in 'RO line 1' is then fed into 'RO line 2', where the salinity becomes higher. Overall, tandem RO processes for maximum water recovery and RO brine minimization are considered to be promising alternatives in brackish water desalination. However, particular process conditions need to be analyzed carefully on a site-by-site basis [18].

Electrodialysis (ED) or electrodialysis reversal (EDR), forward osmosis (FO) and membrane distillation (MD) are also membrane separation processes. ED is another desalination technology that employs electrical potential difference to move ions through ion-exchange membranes. As shown in Table 3, ED is considered as an alternative to RO but it is often only recommended for treating brackish water with TDS level below 10,000 mg/L [23]. For higher salinities, RO is more competitive since ED cost is proportional to the amount of salts carried through the membrane [59]. Further research is required to avoid scaling in ED units and to improve selectivity and permeability of membranes [17].

FO is another technology for brine concentration with low energy consumption [17]. In contrast to RO, the osmotic pressure is the driving force for mass transport [60]. The main drawbacks of FO technology are the risk of salt precipitation on the membrane, the need to develop more robust membranes and a suitable draw solute to improve the separation process [17,60]. Finally, MD is a promising alternative for treating highly concentrated water. The vapour pressure difference induced by the temperature difference across a hydrophobic membrane acts as the driving force [8,14,50]. Still MD is not a mature technology and most of the research work thus far published has focussed on laboratory scale studies to investigate the influence of operating conditions [61].

Although RO is the leading technology in inland desalination and ZLD systems, not all of the RO brine can be concentrated using additional RO stages [2]. The configuration shown in Fig. 3 increases R_w but is far from achieving ZLD [17]. Once the brine is highly concentrated (TDS > 50,000 mg/L) and thus limited by the osmotic pressure, RO technology is not effective [23]. In this situation another downstream process, such as vacuum evaporation, has to be applied in municipal desalination facilities [18]. In this approach, vacuum evaporation follows RO when sufficient treatment has been performed to remove potential scalants. This approach minimizes costs and reduces the volume of brine that eventually has to be concentrated by the evaporator, as shown in Fig. 4 [2,21]. Finally, concentrated brine can be disposed of in an evaporation pond. Alternatively, it is technically possible to obtain salts in a solid, dry and crystalline form by applying a later crystallization process [62]. Such a ZLD solution is environmentally friendly with consequent nil discharges into the environment. However, further research is required to reduce energy requirements and to develop new systems recovering residual heat or steam [17]. Table 3 summarizes the main features of significant concentration technologies applicable in municipal desalination depending on feed water salinity.

As shown in Table 3, RO is by far the most cost-effective solution in terms of energy consumption, capital and operating costs.

MD, FO and ED technologies have been tested on a pilot plant scale for RO brine minimization in inland desalination, although it is difficult to assess their feasibility on an industrial scale. Martinetti et al. [68] tested/compared vacuum-enhanced direct contact membrane distillation (VEDCMD) and FO for RO brine minimization in two different streams with TDS levels averaging 7,500 and 15,000 mg/L. R_w levels in both technologies were limited by salt precipitation. Water recoveries up to 90% and 81% were achieved, respectively, by FO and VEDCMD. In both cases, antiscalant addition was shown to be effective at maintaining high water flux for an extended time.

Korngold et al. [69] applied ED for the treatment of RO brine saturated with $CaSO_4$ and/or SiO_2. The RO brine was generated in a brackish water desalination facility in Mashabei-Sadeh. The ED treatment was undertaken under reverse polarity in a non-continuous operation. The brine, circulating through the ED system, also passed through a separate $CaSO_4$ precipitator. Mineral precipitation was enhanced by the addition of gypsum seeds. RO brine concentration was successfully increased from 1.5% to 10%. Eventually, further RO brine minimization was limited by SiO_2 precipitation in the ED brine.

Oren et al. [70] also applied ED for RO brine minimization. The feed to the ED unit corresponded to RO brine generated during the desalination of brackish water from the Negev Highlands (Israel). Water recoveries around 97%–98% were achieved by combining RO with ED. Brackish feed water was concentrated from 3,000 to 100,000 mg/L TDS. Chloride levels around 200 mg/L or less were measured in treated water. The ED system was run in a batch mode. Scaling during the ED treatment was prevented by acidification, operating the ED module in a reversal mode (EDR) and by incorporating a side loop crystallizer/settler module. Eventually, concentrated brine from the ED treatment was further concentrated by Wind Aided Intensified eVaporation (WAIV) that brought final brine concentration to over 300,000 mg/L TDS.

Closed circuit desalination is another alternative for brine minimization based on a batch-like operation. Generated RO brine is recirculated to the same RO membrane. A previous study conducted with brackish water achieved 97% water recovery in a single stage operation system and was

Table 3
Recommended operating range, maximum concentration capabilities, energy requirements and production costs for different concentration technologies applicable in brackish water desalination

Technology	System	Operating range	Concentration capacity	Energy requirements	Production costs (US$/m^3)
RO membranes	Membrane separation process	Brackish water RO membranes: TDS, 50–12,000 mg/L [23]	–	Brackish water (0.54 kWh/m^3)[a]	Municipal brackish water desalination (US$0.28 to 0.63/m^3)[c] CSG water (US$1.54/m^3)[d]
		Seawater RO membranes: TDS, 8,000–50,000 mg/L [23]	TDS: 65,000–75,000 mg/L [21]	Seawater (4–6 kWh/m^3)[b]	US$1.5–2.3/m^3 (seawater)[e]
Electrodialysis (ED/EDR)	Membrane separation process	TDS: 300–10,000 mg/L [23]	TDS: up to 100,000 mg/L[f]	Up to 15 kWh/m^3[3,g] 6.6–8.7 kWh/m^3 (TDS = 0.45 g/L) [59] (seawater)	US$0.38–6.38/m^3 (brackish water)[h]
Mechanical vapour recompression (MVR) evaporators	Thermal separation process	Following RO desalination	TDS: above 160,000 mg/L[i]	36 kWh/m^3[3,j] 30–50 kWh/ton of distillate [67]	(US$2.1–4.7/m^3)[K]
Crystallizers	Thermal separation process	Following vacuum evaporation[l]	Solid Most of remaining water is recovered as distillate [21]	Vapour compression crystallizers: 52.8–66 kWh/m^3 of feed water [21]	MVR crystallizer: (US$7.4–10.5/m^3)[k] Mechanical forced circulation crystallizer: (US$14.8–25.4/m^3)[k]

[a]Brackish water RO treatment plant. Feed water TDS = 4,000 mg/L. R_w = 80% [52].
[b]Sydney seawater desalination plant: 4.2 kWh/m^3. Kwinana (Perth) desalination plant: 4–6 kWh/m^3 [24].
[c]Cost of brackish groundwater desalination in Texas [63].
[d]CSG water desalination in Australia. TDS content in CSG feed water = 6,000 mg/L [11].
[e]Cost of seawater desalination in Australia [11].
[f]EDR technology. Brackish water was concentrated from 3,000 to 100,000 mg/L [64].
[g]Typical values of operational parameters for ED units [65].
[h]EDR treatment costs for treating brackish waters. Production costs depends on R_w and brine disposal costs [66].
[i]Concentration capacity depends on feed water salinity. 160,000 mg/L for a feed water salinity of 60,000 mg/L [21].
[j]Falling film vacuum evaporator. 4,000 L/h capacity [62].
[k]Data provided by Condorchem Envitech. Environmental engineering firm specialized in evaporation techniques.
[l]Concentration and precipitation of salt or sludge from liquid brines.

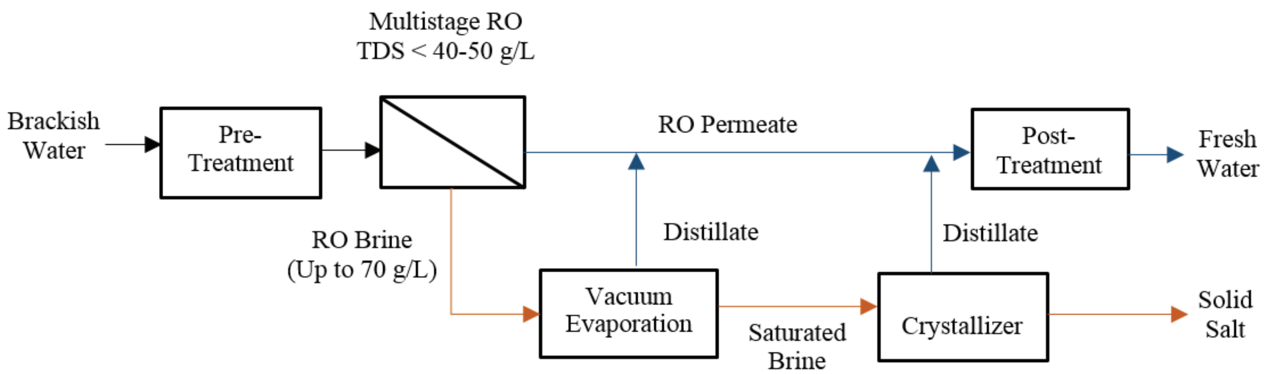

Fig. 4. Traditional ZLD 'lay-out' (municipal desalination).

only limited by salt scaling. This process required less feed pressure and energy when compared with a three stage RO system. In addition, this alternative improved membrane performance and eliminated the use of energy recovery devices [17,71].

WAIV technology was also proposed and compared with traditional evaporation ponds for RO brine minimization. This technology uses wind energy to increase the evaporation rate of brine. This technology, influenced by weather conditions, incorporates a support structure with fabric sheets. During the process, the brine is initially distributed across the sheets. Then, it is concentrated as it flows down the sheets assisted by the evaporation effect of the wind passing across the surfaces. Collected brine at the bottom of the unit can be recycled and further concentrated again in the WAIV plant [72]. Katzir et al. [73] used bench pilot WAIV units to further concentrate ED and RO brines generated during desalination of brackish groundwater. This study also aimed at salt recovery. ED brine was concentrated by WAIV up to 230,000 mg/L TDS. During the experiments, $CaSO_4$ precipitation on the feed basin and the evaporation surfaces was reported. This circumstance led to an enrichment of magnesium relative to sodium in the resulting super-concentrated brine [73]. Overall, one of the main drawbacks of WAIV technology is that it can also pollute groundwater. More experiments at industrial scale are required for process optimization [17].

3.2. ZLD and RO brine minimization technologies in CSG water desalination

The 'CSG Water Management Policy 2012' states the position of the Queensland Government in relation to CSG water management and use. This Policy prioritizes the recovery of useable products from CSG brine wherever feasible [6,8,14,74].

Different studies have been carried out in Australia aiming at RO brine minimization and ZLD in CSG facilities [6,8,14,75]. In this regard, Simon et al. [75] investigated the feasibility of producing NaOH from CSG brine by membrane electrolysis (ME). $NaHCO_3$, Na_2CO_3 and NaCl are the dominant sources of sodium available in CSG brines. In this research, synthetic solutions of these salts were prepared and used as feedstock for the experiments. ME was shown to be more effective for desalting $NaHCO_3$ solutions followed by NaCl and then Na_2CO_3 solutions of equivalent concentration. Moreover, water recovery rates increased as the brine concentration decreased. Finally, it was also reported that the use of 100 g/L $NaHCO_3$ solutions resulted in NaOH production with lower strength (about 12% w/w) than that produced from NaCl solutions with the same concentration. This issue was attributed to the lower electrical conductivity and osmotic pressure of the $NaHCO_3$ solutions.

Duong et al. [8] researched a process for CSG brine (TDS = 14,100 mg/L) minimization including a pilot MD plant equipped with a novel spiral-wound air gap MD module. Water recoveries around 95% were reported by a process combining UF/RO and MD. Membrane scaling was not observed in these experiments. This phenomenon could be attributed to the addition of antiscalant during RO pretreatment and the small temperature gradient applied in the MD step. However, SiO_2 and calcium scales could be present in long-term operation and further research was recommended.

Nghiem et al. [6] investigated a process for the treatment of slightly brackish CSG water (TDS = 2,510 mg/L) generated in the Gloucester Basin (Australia). The process combined UF, RO and multieffect distillation (MED). Water recoveries around 95% were achieved. Generated super-concentrated brine was predominant in $NaHCO_3$ (TDS = 48,000 mg/L). It was reported that antiscalant addition to the RO brine prevented scaling on the evaporative tubes during MED operation. However, mineral deposition on the sight glass of the MED evaporative chamber was observed. The issue was addressed by chemical cleaning with sulphamic acid and NaOH at the end of the experiments.

Duong et al. [14] successfully investigated NaOH production from CSG RO brine by a combination of MD and ME. The feasibility of ME technology for NaOH production using brine generated in seawater or CSG water desalination facilities had been reported in previous studies [75,76]. CSG brine concentration to near saturation was initially required for NaOH production by ME. For this research, synthetic solutions mixing NaCl and $NaHCO_3$ were prepared simulating CSG brine. The MD plant was operated at 90% water recovery and no membrane scaling was observed during the tests. Higher R_w levels resulted in precipitation of NaCl, $NaHCO_3$ and Na_2CO_3 on the membrane and a decrease in distillate quality and water flux. This study concluded that ME combined with MD for NaOH production can achieve energy savings for both processes [14].

Duong et al. [50] also focussed their research on membrane scaling control during RO brine (TDS = 17,100 mg/L) minimization using MD. Generally, prevention of membrane scaling should always be considered as the first option regardless of the efficacy of chemical cleaning. MD experiments showed that water recoveries above 70% resulted in salt precipitation and water flux decline. In this regard, later membrane cleaning was not able to completely remove scale deposits from the membrane. As a consequence, concentration polarization and membrane scaling increased while MD performance decreased. It was also reported that water recoveries around 80% were achievable without membrane scaling by reducing concentration polarization phenomenon by way of limiting feed brine temperature and water flux.

WAIV technology was also tested for CSG RO brine minimization. Initial experiments by a CSG operator were conducted in Roma (Australia) with a demonstration unit. This study concluded that WAIV is able to evaporate 24 times more water than a conventional evaporation pond of equivalent footprint area [72].

Overall, RO brine management remains as a significant technological challenge in CSG desalination and only a limited number of studies on a pilot plant scale have been undertaken in Australia and worldwide [75,77]. Feasible alternatives need to consider RO brine composition and the huge volumes involved. NaOH production from CSG RO brine by ME seems to be a promising option. This alternative has been reported in different technical papers [14,75]. However, the total volume of impurities present in CSG brine could be a limitation for this ZLD approach [76]. Moreover, no matter which technology is initially selected for RO brine minimization/concentration, further research is required to reduce/avoid scaling problems.

3.3. Scaling potential in ZLD and brine minimization technologies

The concentration factor (C_F) in RO desalination can be defined by the following equation where C_c and C_f are the brine and feed water concentrations, respectively, and R_s is the nominal salt rejection [25,56]:

$$C_F = \left(\frac{C_c}{C_f}\right) = \left(\frac{1}{1-R_w}\right) \times \left[1 - R_w \times (1 - R_s)\right] \tag{1}$$

According to Eq. (1), when R_w is increased above 70%, C_F increases dramatically enhancing the precipitation of sparingly soluble salts on the RO membrane. Maximum achievable R_w during brackish water desalination can be limited by antiscalant efficacy and salt precipitation on the RO membrane.

In thermal desalination, scaling of heat transfer surfaces is also of great concern having a substantial influence on the overall performance of the desalination process [78]. The scaling risk also has to be reduced when applying other desalination technologies including FO, MD or ED. Table 4 shows references found in the literature of common sparingly soluble salts that might limit overall R_w and scale different concentration technologies.

3.4. Scale minimization technologies: 'high pH precipitation treatment' vs. acid/antiscalant addition

Two opposite and contradictory solutions can be considered to avoid scaling while increasing recoveries in existing inland desalination plants. The first one, involving acid/antiscalant addition to the RO brine, allows salt supersaturation to a certain extent. Then, R_w can be increased by implementing an additional RO stage. In this case, achievable R_w is limited by antiscalant efficacy.

The second option looks at the integration of an intermediate 'high pH precipitation treatment' for RO brine minimization between consecutive RO stages or between RO and another brine concentration system. In contrast to the use of antiscalants, this solution involves mineral precipitation and removal of sparingly soluble salts from the RO concentrate. Following pH adjustment and possible antiscalant addition, the softened brine can be further concentrated by an additional RO stage or another advanced concentration technology. Higher R_w is achievable by applying this second strategy [56,85] with salt recovery options available.

The integration of an intermediate 'high pH precipitation treatment' during municipal brackish water desalination was broadly investigated by different researchers. Most of the work was undertaken on laboratory scale and, using pilot plants, aimed at increasing R_w during inland desalination.

Ning et al. [88] investigated an intermediate lime softening treatment for the concentrates (TDS = 7,465 mg/L) generated in a primary RO system operated at 85%–90% water recovery. That precipitation treatment successfully removed SiO_2 and $BaSO_4$ from primary RO brine. In addition, an extra 70% brine concentration was achieved in a secondary RO unit.

Williams et al. [89] examined the removal of calcium, barium, strontium, magnesium and silica from primary RO concentrate generated from desalting Colorado River water (TDS = 585 mg/L) to allow further concentration in a secondary RO step. A chemical precipitation treatment including coagulation, sedimentation and filtration was applied before further concentrating the brine in a secondary RO stage. R_w of 98% was possible by following this path.

Table 4
Potential scaling salts affecting different desalination technologies

Technology	Reverse osmosis (RO)	Forward osmosis (FO)[a]	Membrane distillation (MD)[b]	Electrodialysis (ED/EDR)[c]	Thermal desalination systems[d]
Scalant	RO membrane	FO membrane	MD membrane	Ion-exchange membrane	Tubing and process surfaces
$CaCO_3$	Yes [20,23]		Yes [50,79,80] (at relatively high saturation indexes)	Yes [81]	Yes [20,82]
$CaSO_4$	Yes [20,23]	Yes [83,84]	Yes [50,79,80,85]	Yes [69]	Yes [20,82]
$BaSO_4$	Yes [20,23]				
$SrSO_4$	Yes [20,23]				
SiO_2	Yes [20,23]	Yes [84]	Yes [8,50,79,80]	Yes [69]	
$Ca_3(PO_4)_2$	Yes[e]		Not found in MD literature [79]		
CaF_2	Yes [23]				
Magnesium scales			Yes[f]		Yes [20,82]

[a]Following chemical cleaning, FO shows better flux recovery than RO in the event of silica scale. Chemical cleaning is also more effective in FO than in RO in the event of $CaSO_4$ scale [84].
[b]Calcium sulphate scale was found to be a common problem in MD [50]. Overall, MD is more fouling resistant than RO [79,86].
[c]Electrodialysis reversal (EDR) has the advantage of descaling membrane surfaces by utilizing a flow and polarity reversal [4,87].
[d]Thermal brine minimization is often limited by precipitation of sodium sulphate, sodium carbonate and sodium chloride [21].
[e]Calcium phosphate scaling can be common when RO is applied to municipal wastewater [23].
[f]Feed solutions with high levels of Mg^{2+} may cause problems in MD [50,79].

Rahardianto et al. [56] looked into the application of accelerated precipitation softening (APS) for the treatment of primary RO from desalting mildly brackish surface water (TDS = 941 mg/L) to allow further concentration in a secondary RO process. The treatment involved alkaline pH adjustment, calcite crystal seeding, microfiltration and pH adjustment to avoid scaling issues during secondary RO. It was demonstrated that high R_w in brackish desalination was achievable by using this technology.

Qu et al. [85] conducted a similar study combining APS with direct contact membrane distillation for the treatment of primary RO brine, increasing overall R_w from 50% to 98.8%. The process included pH adjustment with NaOH, followed by calcite seeding and final microfiltration.

Gabelich et al. [90] explored an intermediate chemical demineralization treatment for primary RO brine (TDS = 4,995 mg/L) generated from desalting Colorado River water. The process included NaOH and $NaHCO_3$ addition for salt precipitation followed by pH neutralization with H_2SO_4, and achieved an increase of the R_w from 85% to 95%.

Sanciolo et al. [91] studied the application of APS technology for the treatment of primary RO brine generated in inland municipal wastewater treatment plants. The removal of 'calcium scale precursor ions' was tested with three different seed materials: (a) $CaCO_3$, (b) $CaSO_4$, and (c) $Ca_3(PO_4)_2$. The first two were not effective. Best results were achieved after addition of $Ca_3(PO_4)_2$ seed particles at 20 g/L or PO_4^{3-} ion in stoichiometric excess of the Ca concentration. Although effective, this treatment resulted in high chemical and energy costs.

Mohammadesmaeili et al. [92] studied the removal of different potential scalants from reclaimed water RO brine by selective precipitation. Products with resale value were recovered during the precipitation process. Three different softening processes were tested including: (a) the traditional lime-soda ash treatment; (b) a modified process with preacidification to eliminate carbonate, and (c) another one including a gypsum crystallization step in combination with the modified process to be applied with high sulphate brines. Overall, good quality calcite and gypsum were recovered in the precipitation process while high efficiency in foulant removal was achieved.

Bond et al. [2] carried out bench scaling tests with RO concentrates of different characteristics in order to remove different insoluble salts affecting R_w during RO desalination. This intermediate process was considered critical since it has a direct impact on following treatment steps like secondary RO or possible thermal desalination. Different technologies were evaluated either individually or in combination including chemical precipitation, fluidized bed crystallization, adsorption with activated alumina and ion-exchange.

In summary, most previous work was aimed at maximizing recovery of water in municipal desalination by removing the potential scale forming offenders from primary RO by using "high pH treatment" aided by NaOH, $Ca(OH)_2$, $NaH(CO)_3$, Na_2CO_3 and also by inducing the crystal formation by adding seed crystals such as $CaCO_3$ and $CaSO_4$. The nature of the chemicals added essentially depends on the RO brine characteristics, cost and salts recovery options from generated sludge. Overall, NaOH is easier to store and manipulate than other chemicals and could be more effective for treating both low and high alkalinity waters [93].

There is an existing groundwater treatment facility in Southern California incorporating an intermediate softening process. This installation includes a high-rate pellet softening and solids clarifier system to treat primary RO brine. Softened and filtered brine is eventually fed to a secondary RO stage. This plant produces $CaCO_3$ pellets that can be used in different applications [94].

Since the initial RO pretreatment in inland desalination plants is usually aided with antiscalant addition, other researchers have investigated antiscalant scavenge/removal when applying an intermediate 'high pH precipitation treatment' between consecutive RO stages.

Greenlee et al. [55] investigated the impact of antiscalants added during RO pretreatment on a later brine treatment, including salt precipitation and solid/liquid separation processes. It was shown that antiscalant addition during RO pretreatment reduced calcium precipitation from RO brine and negatively affected the following solid/liquid separation process. Overall and for higher antiscalant doses, a greater decrease in calcium precipitation was observed during RO brine treatment. In another report, Greenlee et al. [95] also investigated an intermediate RO brine treatment between consecutive RO stages including the following steps: optional oxidation of antiscalants with ozone and H_2O_2, 'high pH precipitation', and solid–liquid separation. The oxidation step was shown to increase calcium precipitation, while the antiscalants solubilizing capabilities were reduced. The R_w increased from 80% to 90% for the non-ozonated brine and from 80% to 94% for the ozonated brine.

Rahardianto et al. [96] also studied a two-step chemically enhanced seeded precipitation (CESP) process performed between consecutive RO stages for the treatment of primary RO brine. The process combined lime treatment to allow $CaCO_3$ precipitation and antiscalant (polycarboxylic acid) scavenge followed by $CaSO_4$-induced precipitation with gypsum seeding. This process could be less chemically intensive than conventional softening while increasing overall R_w from 63% to 87% by applying a secondary RO stage.

McCool et al. [97] investigated antiscalant removal from an RO concentrate with high gypsum scaling potential by lime treatment prior to seeded gypsum precipitation (CESP process). Adequate antiscalant removal (up to 90%) was achieved after the lime treatment process facilitating later seeded gypsum precipitation. Then, R_w can be further increased by implementing a secondary RO stage [97].

Overall, previous research has shown that the application of an intermediate 'high pH precipitation treatment' was able to decrease the adverse impacts of residual antiscalant added during RO pretreatment in municipal desalination. Lime treatment or direct contact with $CaCO_3$ is potential solutions to mitigate negative effects of antiscalants on later salt precipitation and solid/liquid separation processes. This can be a significant issue when considering feasible strategies for RO brine minimization and salt recovery in municipal desalination. Further research is needed to find economical chemical treatments.

Conversely, high bicarbonate concentration in the CSG RO brine remains as a major constraint for the integration of an intermediate 'high pH precipitation treatment' for scale control and RO brine minimization during CSG water desalination. This issue could be addressed to some extent when an acid is initially added to the RO brine to covert HCO_3^- into CO_2, which may be off-gassed. This alternative reduces the quantity of solids generated in the downstream 'high pH precipitation treatment' and the alkaline reagent demand associated with increasing the pH level, though the neutralization of the initially added acid needs to be factored in.

3.5. Other ZLD and volume reduction systems (commercially available or patent protected)

Other existing 'volume reduction technologies' are the ARROW™, HEEPM™, HERO™, and VSEP™ systems. With the exception of HERO™ the rest of the systems is considered as emerging technologies [21]. The HERO™ system, patented by Mukhopadhyay [98], is a process conceived for treatment of water in membrane separation processes. The integration of the HERO™ system in a two stage RO system has already been considered by different authors [17,21]. The hardness/alkalinity ratio in feed water is most often initially adjusted by alkali addition. Then, hardness is removed quantitatively from the primary RO brine in a weak acid cation exchange resin given the adequate hardness/alkalinity ratio. Following that, CO_2 is removed in a degasification process. Eventually, the pH is increased up to 10.5 or higher enhancing the rejection of various species such as silica in the secondary RO membrane system. This technology minimizes the risk of salt precipitation on the RO membrane and water recovery above 90% is achievable when treating brackish water [17]. The application of HERO™ minimizes Capex relative to the brine concentration system. However, although energy costs are also reduced, chemical and solids disposal costs are increased [21].

SAL-PROC™ is a ZLD system that allows the sequential extraction and recovery of different salts like gypsum, NaCl, $Mg(OH)_2$, $CaCl_2$, $CaCO_3$ and Na_2SO_4 from RO brines [16]. This system is particularly recommended for brackish inland brines and for brines with high concentrations of sulphate, potassium and magnesium [17]. SAL-PROC™ combines multiple evaporations and/or cooling stages, supplemented by mineral and chemical treatments. When RO technology is combined with SAL-PROC™ the system is referred as Reverse Osmosis SAL-PROC (ROSP) [15]. Recovered products are high quality. Ahmed et al. [99] have suggested some potential applications for recovered salts. SAL-PROC™ technology was tested by Arakel et al. [15] for the treatment of brackish water from Lake Tutchewop. This solution allowed the recovery of saleable products and achieved ZLD.

SAL-PROC™ was also tested for the treatment of RO brine generated during CSG water treatment in Queensland (Australia). The volumes of saline water were huge. ROSP technology produced fresh or irrigation quality water, chemicals and minerals products as shown in Fig. 5. Overall, SAL-PROC™ technology allows sustainable management and could facilitate a cost-effective solution for large volumes of saline effluent [15].

There is an existing patent for the manufacture of sodium hydroxide and sodium chloride products from CSG RO brine containing NaCl and at least $NaHCO_3$ or Na_2CO_3. This solution combines different processes including lime addition,

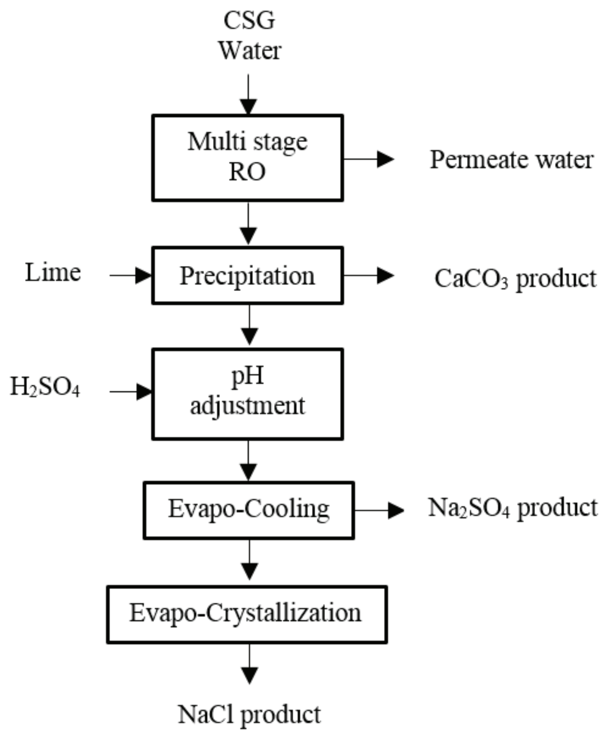

Fig. 5. Application of SAL-PROC™ technology to CSG produced water [15].

chemical precipitation for RO brine purification, concentration by evaporation and cooling [100]. In addition, the 'Optimised Salt Recovery' process has also been presented as an alternative to traditional costly selective salt recovery processes. This process works through modification of the CSG brine chemistry to avoid co-precipitation of major salts, while trace impurities are removed from the RO brine [101].

Finally, the optimized pretreatment and unique separation (OPUS) process developed by Veolia is recommended for desalination of hard water with high concentrations of silica, organics, heavy metals, boron and particulates. It involves a combination of different processes, such as degasification, precipitation softening, media filtration, ion-exchange, cartridge filtration and RO operated at high pH. The OPUS system has been tested in the oil and gas, and mining industries. The application of this system provides high R_w levels, reduced waste volume, low energy consumption and facilitates an effective control of scaling. OPUS II is a new version of this system specifically focussed on the oil and gas industry. OPUS II simplifies the pretreatment by using ceramic membranes that improve oil removal and facilitate a more compact design [102].

4. Advantages and shortcomings of ZLD, RO brine minimization, and scale control technologies: further research opportunities

The advantages and disadvantages, and research needs/opportunities of the most relevant technologies and systems for RO brine minimization, ZLD and scale control in inland desalination plants have been summarized in Table 5.

Table 5
Summary of characteristics of relevant technologies and systems described in this review paper for RO brine minimization, scale control and ZLD in inland desalination

RO brine concentration technologies			
Technology/system	Maturity	Technical	Economic aspects
Evaporation ponds	Industrial scale	Risk of groundwater contamination Discouraged or banned for CSG water management in Queensland (Australia)	Requires large areas of land
WAIV technology	Pilot plant scale	Risk of groundwater contamination Influenced by weather conditions Risk of scaling and fouling Further research required for process optimization	Higher evaporation rates than evaporation ponds
Multistage RO	Industrial scale	Leading technology R_w limited by the risk of scaling and the practical limits to provide the osmotic pressure	Cost-effective solution in terms of energy consumption, Capex and Opex
Evaporators and crystallizers	Industrial scale	R_w in evaporators limited by scaling Further research required to develop new systems recovering residual heat or steam	ZLD approach for RO brine High Capex and Opex
ED	Pilot plant scale	Cost-effective only for treating waters with TDS level below 10,000 mg/L Further research required to avoid scaling and to improve selectivity and permeability of membranes	NaOH production from CSG RO brine can be feasible by ME technology High Capex and Opex
FO	Pilot plant scale	High concentration capacity R_w limited by the risk of scaling Need to develop more robust membranes and a suitable draw solution	Simplicity Moderate energy consumption
MD	Pilot plant scale	Promising alternative Further research required to avoid scaling	High Capex Lower energy requirements than evaporators and crystallizers
Closed circuit desalination	Industrial scale [17]	R_w limited by the risk of scaling	High Capex
Scale control technologies			
Technology/system	Maturity	Technical	Economic aspects
Acid/antiscalant addition	Industrial scale	Limited efficacy An excessive dose can cause biofouling	Simplicity Low Capex High Opex
'High pH precipitation treatment'	Industrial scale	Feasible alternative when antiscalants are not further effective Further research required to find economical chemical treatments Its application for CSG brine minimization can be limited by bicarbonate concentration	Salt recovery options Chemically intensive High Capex Later acid/antiscalant addition can be required
Other ZLD and volume reduction systems			
Technology/system	Maturity	Technical	Economic aspects
HERO™	Industrial scale for non-municipal applications	Interstage system Minimizes risk of salt precipitation	Minimizes Capex relative to the brine concentration system
SAL-PROC™	Patented Tested for the treatment of CSG brines	Combines multiple technologies Reduction of environmental and operational footprints	Allows selective recovery of salts from RO brine High Capex and Opex
OPUS/OPUS II	Tested in the oil and gas and mining industry	Combines multiple technologies. Reduces scaling risk	Reduced brine volume Low energy consumption High Capex and Opex

5. Conclusions

RO is the dominant technology applied in inland municipal desalination and for treating CSG water in Australia. However, RO brine disposal is considered to be an environmental challenge and one of the main handicaps of this technology. Antiscalants are often dosed during RO pretreatment to prevent membrane scaling. Water recoveries are then limited by antiscalant efficacy and large volumes of brine are mainly disposed of in evaporation ponds while alternative options for RO brine minimization and ZLD are researched. Overall, high recovery and ZLD systems could only be applied in specific cases considering the high Capex and Opex involved. In addition, no matter which concentration technology is selected for RO brine minimization, it becomes necessary to cope with the scaling potential problem.

The traditional ZLD approach recommended in municipal desalination combines sequential RO stages with vacuum evaporation/crystallization when sufficient treatment has been performed to remove potential scalants. This solution requires further research to reduce energy requirements and to develop new systems recovering residual heat or steam. Alternatively, MD, FO or ED technologies have also been proposed for RO brine minimization. However, those technologies were tested at a laboratory level or at a pilot plant scale making it difficult to assess their feasibility on an industrial scale.

Conversely, the Queensland Government Policy, in regard to ‘CSG water management and reuse’, has prioritized the recovery of useable products from CSG brine when feasible. Initial studies performed in Australia have revealed the possibility of producing NaOH from CSG brine by ME. In addition, CSG RO brine minimization has also been achieved on a pilot plant scale by the application of processes combining UF/RO and MED or UF/RO and MD. Overall, economic aspects of these promising alternatives require further investigation, including identifying areas where cost reduction might be possible.

The integration of an intermediate ‘high pH precipitation treatment’ in municipal brackish water desalination was broadly investigated on a laboratory scale, and using pilot plants in the USA. The application of this treatment between consecutive RO stages or between RO and another concentration system is chemically intensive and expensive. However, this approach could overcome antiscalant limitations and increase R_w levels, while salt recovery options are enhanced. But options that can be used to increase the water recovery and to facilitate a ZLD solution require rigorous laboratory investigation due to groundwater and RO brine site specific characteristics. Additionally, the application of this precipitation treatment for scale control and brine minimization in the CSG industry can be limited by high bicarbonate concentration in the CSG RO brine.

Finally, more complex systems for ZLD and RO brine minimization like the SAL-PROC™ have been successfully tested for the treatment of brackish water and CSG brine increasing water recoveries while producing saleable products. SAL-PROC™ involves high Capex making its implementation on an industrial scale difficult. However, recovery of saleable products can help to reduce costs, thus making this technology more attractive.

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Conflicts of Interest

The authors declare no conflict of interest.

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