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Process Design of Coal Seam Gas Associated Water Treatment Plants to Facilitate Beneficial Reuse

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This study used computer simulation to select appropriate process designs to make coal seam gas (CSG) associated water comply with beneficial reuse regulations. The hypothesis was that computational simulation might determine the unit operations required for treatment of CSG associated water over a wide range of salinities. AqMB software was used for a complete analysis of water treatment from initial storage pond to final pH and sodium adsorption ratio (SAR) adjustment. Three samples of CSG associated water ranging from 3650 to 22123 mg/L total dissolved solids content were evaluated. Common to each process design was a settling pond, coarse & fine filtration, softening, anti-scalant addition, and reverse osmosis. When the water hardness was very hard, then a lime softening system may be required. Both microfiltration and ultrafiltration were considered, and in both cases, the level of suspended solids was reduced to close to zero. Sodium exchanged WAC resin removed the majority of alkaline earth ions and inhibited downstream scale formation. The presence of substantial alkalinity was addressed by use of acid exchanged WAC resin in combination with an air stripper. However, process economics indicated this option was not favourable. A two-stage Reverse Osmosis system not only recovered from *ca.* 74 to 79 % water with all CSG associated water types but also the water quality was compliant with beneficial reuse regulations. The computer predictions mirrored actual data from operating industry sites.

Keywords: Coal seam gas; coal bed methane; associated water; reverse osmosis; process design

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1. Introduction

Development and commercialization of coal seam gas (CSG) deposits have primarily occurred within the past 30 years in countries such as USA, Australia, Canada, China, and India [1]. CSG originates from biologically and microbial transformed organic matters that have undergone geological processes [2]. Typically, the gas reserves are located in coal seams which are several hundred meters below ground. The extraction process involves depressurization of the seams [3], which results in the collection of substantial amounts of CSG associated water which is saline in character [4]. Current estimates indicate that for example, in Queensland, the associated water volume from the CSG sector is 60.5 GL per annum with a predicted maximum volume of 70 to 80 GL per annum [5]. The salt content in the associated water is highly dependent upon the location of the CSG wells and typically ranges from 1,000 to 10,000 mg/L in Queensland [6, 7]. Consequently, the associated water is normally not directly applicable for beneficial reuse options such as crop irrigation, livestock watering, dust suppression, aquaculture and drinking water [8].

The most common species in CSG associated water are sodium, potassium, calcium, magnesium, strontium, barium, iron, boron, aluminium, colloidal and dissolved silica, sulphate, chloride, fluoride, bromide, and bicarbonate/carbonate [9]. In all reported CSG associated water samples, the dominant salts are sodium chloride and sodium bicarbonate [6, 10, 11]. Also present can be organic impurities and microbial/algal matter; for instance, a high level of algae was measured in CSG associated water produced in New South Wales, Australia [12]. A recent review has summarized the current methods applicable to the treatment of CSG associated water [4]. Technologies including coagulation, lime softening, microfiltration, ultrafiltration, ion exchange softening, anti-scalant dosing, ion exchange desalination, reverse osmosis (RO), as well as capacitive deionization (CDI) were all discussed. The central desalination methods were ion exchange and RO, both of which required pre-treatment of CSG associated water to promote the optimum operation of the treatment plant. For example, RO membranes are susceptible to fouling due to microorganisms/algae, dissolved organic matter and particulate/colloidal matter as well as scale deposition from alkaline earth ions, silicates, carbonate, sulphates and phosphates [13].

In terms of process designs of water treatment plants to remove the contaminants in CSG associated water, there does not appear to be a standard approach. For example, Le [12]

outlined a treatment strategy for remediating CSG associated water wherein the unit operations employed included a pond for storage of the water, disc filtration, microfiltration, biocide addition, two-stage RO and finally calcium addition to adjust sodium adsorption ratio (SAR) levels to ensure regulation compliance. Alternatively, Pless *et al.* [14] employed an ion exchange system comprising of a cation and anion exchange materials (resin and hydrotalcite) to desalinate CSG associated water from the San Juan basin in the USA. Whereas, Dennis [15] deployed a single cation resin bed as part of a Higgins Loop configuration for treatment of CSG associated water from the Powder River Basin in the USA. Carter [16] described the process layout for the Queensland Gas Company (QGC) CSG associated water treatment facility located in Queensland, Australia. A 100 ML/day system is in operation which comprised of pre-treatment (including lime softening) followed by ultrafiltration, ion exchange softening and RO membrane.

Process modelling using computational tools is an established method in the water treatment industry. In relation to the RO, various software packages have been implemented such as ROSA [17], IMS-Design [18], WINFLOWS [19], Toray DS2 [20] and Projection+ [18]. Nevertheless, the application of computational methods to determine optimal process designs for water treatment units in the CSG industry has received minimal attention. Plumlee *et al.* [21] developed a software-based screening tool for the CSG industry which guided users to which unit operations were recommended when treating CSG associated water for various beneficial reuse options. However, despite the usefulness of this tool, detailed process engineering design was not provided. Recently, Vedelago and Millar [22] used computer simulation to predict the optimal treatment approach for CSG associated water containing high levels of alkalinity. A strategy involving pH adjustment followed by chemical amendment using gypsum was evaluated and it was concluded that, if the bicarbonate concentration was above *ca.* 1000 mg/L, this methodology was not appropriate. The additional sulphate or chloride components made the water not suitable for irrigation purposes, even with highly salt tolerant plants. An alternate approach of using a cation exchange resin was also studied. It was found that, compared to a weak acid cation (WAC) resin, a strong acid cation (SAC) resin type was more effective to mitigate levels of bicarbonate/carbonate, calcium, magnesium and sodium present in the associated water; although regeneration of SAC resin was more expensive. After a strong acid cation resin bed, the subsequent processes of degassing and lime dosing were found to adjust pH and SAR

levels suitable for irrigation. Nevertheless, this study only simulated the effect of pH adjustment and cation exchange resin. Wicks *et al.* [23] also used AqMB software to determine when ion exchange of CSG associated water with various synthetic resins was potentially viable. Venkatesan and Wankat [24] simulated a cation exchange resin to soften brackish water prior to RO filtration and reported that *ca.* 90 % water recovery was achieved without scale formation on the RO membrane.

Notably, there remains a need to design reverse osmosis based desalination plants for the treatment of CSG associated water. The variable quality of the CSG associated water necessitates that the process design requires tailoring to meet the challenges of each water sample. Hence, the use of computational methods to identify the most prospective strategies is recommended. Therefore, this study aimed to develop a means to screen a range of water treatment process designs for CSG associated water and provide a basis for the selection of unit operations. The hypothesis was that computational simulation might accelerate the selection of appropriate unit operations for the treatment of CSG associated water with salinity ranging from low (*ca.* 4000 mg/L) to high values (*ca.* 22,000 mg/L). Specific research questions which were considered included: (1) What are the impacts of the CSG associated water composition upon plant design and performance? (2) What is the recommended pre-treatment strategy to maintain RO membrane integrity? (3) Which process configurations are optimal for CSG associated water treatment of variable quality? The methodology involved use of AqMB (Aqueous Material Balance) software which allowed examination of unit operations such as storage ponds, coagulation, clarification, media filtration, biocide introduction, micro- and ultrafiltration; softening; addition of scaling inhibitors; pH adjustment, nanofiltration and reverse osmosis.

2. Materials and Methods

2.1 CSG Associated Water Composition

Three CSG associated water compositions were investigated, which encompassed a range of salinities which required desalination with reverse osmosis Table 1.

Table 1: Water characteristics of CSG associated water samples

	CSG 1 (Low TDS) [6]	CSG 2 (Medium TDS) [6]	CSG 3 (High TDS) [12]	Units
Total Dissolved solids (TDS)	3892	10030	23130	mg/L
Conductivity	5495	17960	23290	μS/cm
pH	8.4	7.9	8.13	
SAR	162	80	338	
Aluminium	0	0	0.01	mg/L
Barium	0.67	9.38	15.4	mg/L
Bicarbonate	1396	195.8	13110	mg/L
Boron	0.85	0.17	1.3	mg/L
Calcium	3	106	14.6	mg/L
Carbonate	44	7.47	620.5	mg/L
Carbon Dioxide	7.2	2.78	103.5	mg/L
Chloride	1102	5909	2316	mg/L
Fluoride	2.8	0.4	6.5	mg/L
Iron	0.44	3.14	39.5	mg/L
Magnesium	1	33	9.8	mg/L
Potassium	4	20	82.3	mg/L
Silica	21.8	14	25	mg/L
Sodium	1270	3700	6798	mg/L
Strontium	1.05	20.2	3.7	mg/L
Sulphate	0	0	17.8	mg/L
Total Organic Carbon (TOC)	44	138	0	mg/L
Dissolved Organic Carbon (DOC)	37	17	14	mg/L
Total Suspended Solids (TSS)	23	233	130.9	mg/L
Turbidity	8.0	56.1	72.4	NTU

In reality, CSG associated water samples are usually contaminated with algae and other microbiological species as the storage ponds are exposed to the environment for extended periods of time. Indeed, Le reported that a CSG associated water sample collected from northern New South Wales in Australia comprised of >600,000 cells per mL of algae [12]. The removal of algae and other microbial species was not modelled explicitly in this study, but provision was made to add a biocide in the treatment process.

2.2 Software

Aqueous Material Balance (AqMB) water treatment software is provided by Salt Water Solutions [25]. A wide range of physical unit operations are embedded in the software, including separation methods, mixing, dosing, splitting, heat, and mass transfer, and rotating equipment. Accepted equations and theories are used to perform calculations. Economic evaluation is also built into the software with estimates of chemical and energy consumption provided.

2.3 Process Design

Process designs were customized to the particular CSG associated water of interest. The unit operations included: settling pond; coagulation; sand ballasted clarification; microfiltration; ultrafiltration; ion exchange softening; anti-scalant addition; reverse osmosis; pH adjustment and chemical dosing to adjust SAR. Guidance in terms of scale formation involved the application of the Langelier Saturation Index (LSI) or Stiff–Davis Saturation Index (SDSI) models. While LSI reports saturation level of calcium carbonate for water with TDS < 10,000 mg/L, SDSI determines the potential for calcium carbonate formation in water with TDS > 10,000 mg/L [26]. Inspection of the LSI/SDI data revealed that for all three CSG associated water types that calcium carbonate formation was possible as the values were all above zero [Table 2]. Saturation indices also suggested that ferrihydrite was supersaturated in the CSG associated water samples.

Table 2: Scaling potential and saturation indices for CSG associated water samples 1, 2, & 3 after exiting settling pond

	CSG Associated Water 1	CSG Associated Water 2	CSG Associated Water 3
Langelier Saturation Index (LSI)	1.17	0.63	
Stiff–Davis Saturation Index (SDSI)		0.91	1.90
Barium sulphate (Barite)			0.33
Calcium carbonate (Calcite)	0.69	0.17	1.09
Ferrihydrite	1.6	1.6	1.6
Strontium carbonate (strontianite)	0.78		0.95
Talc	0.12		
Barium carbonate (witherite)			0.24

As discussed by Huang and Ma [27], care must be taken when using LSI and SDSI values to predict the scaling potential of species in aqueous solutions. For example, consideration should be made of factors such as concentration polarization, carbon dioxide pressure, and relative activity coefficients of solubility products. AqMB software considers the kinetics of scale formation in addition to saturation indices. Consequently, ferrihydrite precipitation was predicted by AqMB to occur for all three CSG associated water samples at rates of 1.01, 10.9, and 100 mol/h for CSG 1, CSG2 and CSG 3 associated water samples, respectively. In addition, for CSG associated water sample 3 witherite was also suggested to precipitate at a rate of 15.6 mol/h. It is also industry practice to locate a settling pond prior to the desalination stage [4]; hence, a suitable pond was included in all process designs. In addition, as indicated in Section 2.1, the presence of microbiological species was assumed to be present. As such a biocide dosing system was also included in all plant designs.

2.3.1 Low TDS CSG Associated Water (CSG 1)

CSG 1 contained suspended solids which can be generally removed by a clarification unit such as a sand ballasted clarifier [28]. Removal of finer particulates was targeted by the incorporation of both micro- and ultra-filtration units [29]. As there was potential to form calcium carbonate scale [Table 2] an ion-exchange softener was included after the UF stage [30]. The amount of silica dissolved in CSG 1 was significant (21.8 mg/L), which could cause membrane fouling [31]; thus, anti-scalant addition was warranted [32]. The TDS of the CSG associated water was less than 10,000 mg/L, which indicated that brackish water RO (BWRO) units would be suitable for desalination purposes. To make the permeate compliant with regulations, both pH adjustment and micronized gypsum addition to meet SAR requirements were also incorporated in the treatment design [Figure 1] [22].

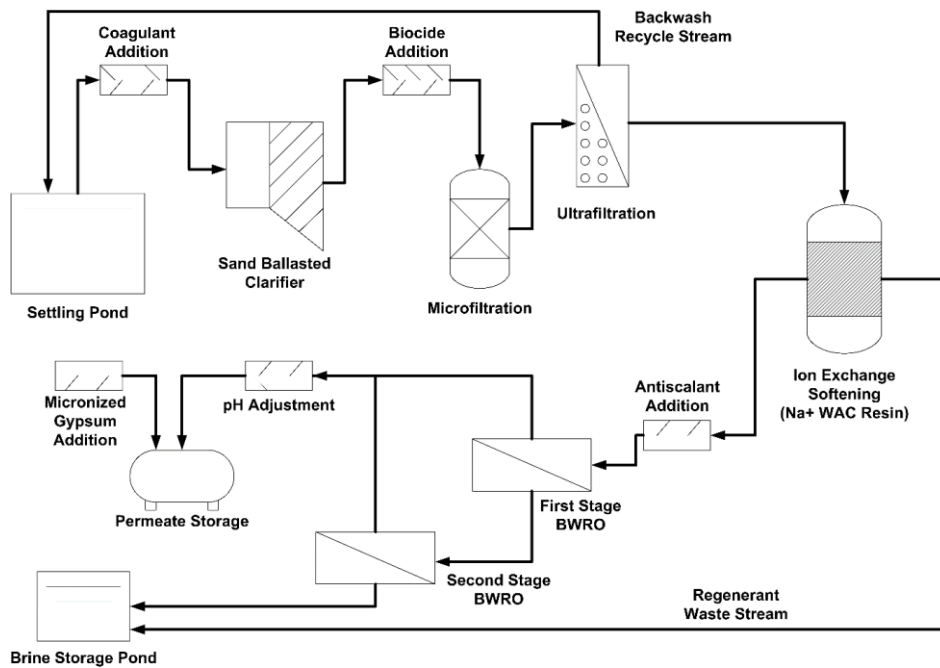


Figure 1: Process design for treatment of CSG associated water sample 1

2.3.2 Medium TDS CSG Associated Water (CSG 2)

A particular feature of CSG 2 water was the relatively high hardness compared to CSG water 1 & 2 (106 mg/L Ca & 33 mg/L Mg). Consequently, initial treatment of CSG 2 water with lime softening was considered [33] instead of a sand ballasted clarifier. Further softening using ion exchange resin was also deemed necessary as the hardness was relatively high. Notably, lime softening does not reduce the amount of alkaline earth ions present to low levels. The higher TDS of CSG 2 compared to CSG 1 meant that seawater RO (SWRO) membranes were required. The remaining operations in the design were similar to those described in Figure 1.

2.3.3 High TDS CSG Associated Water (CSG 3)

CSG 3 was not as hard as CSG 2, but instead, it comprised of a substantial concentration of bicarbonate species (13110 mg/L). Consequently, the introduction of an ion exchange system which could not only soften the CSG associated water but also decarbonize it was a potentially attractive technical solution. The evolved carbon dioxide from the outlined approach would subsequently require removal by a degassing stage. SWRO membranes were again required for the desalination stage.

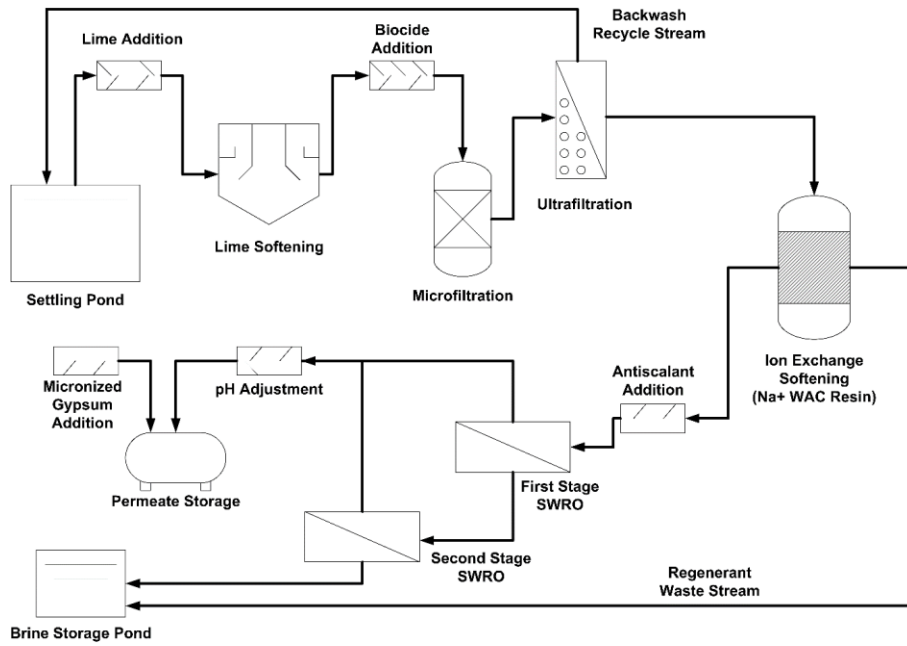


Figure 2: Process design for treatment of CSG associated water sample 2

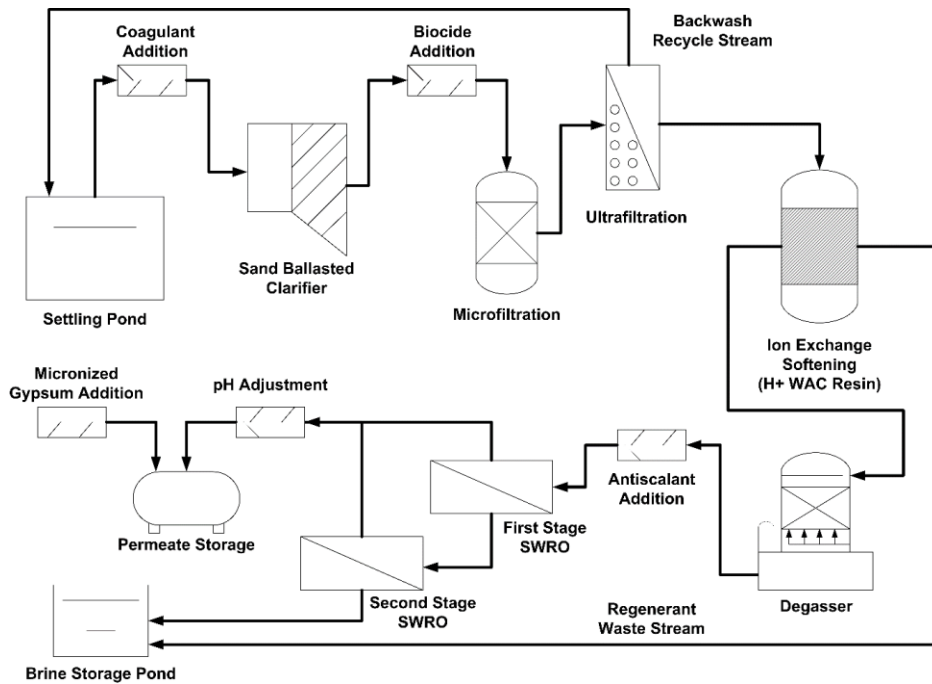


Figure 3: Process design for treatment of CSG associated water sample 3

2.4 Unit Operations

2.4.1 Settling Pond

A settling pond was used in the AqMB simulation as this is standard practice in the CSG industry for collection of water from the various neighbouring wells. The pond parameters were as follows: volume = 47.94 ML; 80 % solids removal; no net evaporation rate; rate of

organics increase = 10 mg/L/annum; residence time = 240 h; evaporation surface area = 11560 m²; wind = 20 km/h; and ambient temperature = 25 °C.

2.4.2 Clarification

Use was made of aluminium chlorohydrate (ACH) coagulant as this material not only does not significantly impact the pH of a CSG associated water sample but also is effective at removing suspended solids [34]. A sand ballasted clarifier was chosen as a unit operation where flocculation occurred as this is employed in the Queensland CSG industry [4]. The clarifier details were as follows: recovery was 98.93 %; overflow solids = 10 mg/L; coagulation tank detention time = 90 s; maturation tank detention time = 240 s; settling tank surface overflow rate = 55 m/h; lamella tube settler overflow rate = 74 m/h.

2.4.3 Lime Softening

When the CSG associated water comprised of high levels of calcium and magnesium ions, a lime softening unit was introduced to the process. In AqMB lime softening was modelled as a thickener to accommodate the significant solids content of the pH adjusted CSG associated water. The model included the following assumptions: minimum underflow solids (0.5 w/v %); overflow solids 10/mg/L; floc/contact zone detention 20 min; and surface loading rate 5 m/h. Lime addition occurred until the pH was equal to 10 in accord with previous literature which suggested this was optimal [33].

2.4.4 Biocide Addition

Sodium hypochlorite was selected as a biocide using a nominal 10 mg/L dose rate. This chemical was added *via* an inline chemical doser unit.

2.4.5 Microfiltration

A microfiltration system was introduced to control suspended solids in the CSG associated water in harmony with the approach of Le [12]. The microfiltration system consisted of a 5 micron membrane cartridge filter of length 1.02 m. The number of cartridges required was 83 and the filtration area per cartridge was 1.5 m. The solids retention capacity of microfiltration filter was 0.15 kg/m².

2.4.6 Ultrafiltration

There are several experimental studies on the performance of ultrafiltration coupling with RO in treating the associated water. For example, Nghiem *et al.* [35] and Qian *et al.* [36] conducted a pilot-scale treatment of CSG associated water using both systems in Australia and China, respectively. Suez ZeeWeed (ZW1500-550) ultrafiltration membranes (51 m² area) were installed in the ultrafiltration system. These membranes were made of polyvinylidene fluoride (PVDF) and the nominal pore size cut-off was 0.02 microns. The total number of UF modules required was 58, and the calculated flux was 62.2 L/m²/h. The recovery rate was 93 % and water backwashing occurred every 21 minutes at 68 L/m²/h. Air scouring also occurred with a duration of 0.5 min.

2.4.7 Ion Exchange Softening

When water hardness was identified as a problem, DOW MAC-3 weak acid cation resin in the sodium exchanged form was used to soften the water. The details are presented in Table 3.

Table 3: Simulation setup for ion exchange softener

Properties	Parameters	
Resin form	Na ⁺ (WAC)	H ⁺ (SAC)
Working capacity (eq/L)	1.8	1.2
Service velocity (m/h)	30	30
Regeneration interval (hr)	48	48
Regeneration type	Reverse-flow	Reverse-flow
Regeneration chemical	NaCl	HCl
Regeneration chemical strength (%)	10	5
Stoichiometry of Regeneration Stage	1.8	2.0
Regeneration/displacement velocity (m/h)	5	5

2.4.8 Anti-scalant Dosing

A silica anti-scalant was simulated using a dosage of 100 mg/L, which was a value promoted as the upper limit when minimizing scale formation in the pipework of a power plant [37]. Anti-scalant was assigned a cost of A\$5000 per tonne [25].

2.4.9 Reverse Osmosis Configuration

The staging ratio for each CSG associated water type was estimated from Equation 1 [38].

Equation 1:
$$R = \left[\frac{1}{1 - Y} \right]^{1/n}$$

Where R is the staging ratio, Y is the desired system recovery expressed as a fraction and n is the number of stages. The desired water recovery was set to *ca.* 75 % in each case [12] which for brackish water should be achieved using a 2 stage RO system [39]. The number of vessels (N_v) required in the first and second stages was estimated by using Equations 2 and 3.

Equation 2:
$$N_v(1) = \frac{N_v}{1+R^{-1}}$$

Equation 3:
$$N_v(2) = \frac{N_v(1)}{R}$$

Due to the relatively low TDS value for CSG 1 coupled with the fact that the permeate did not to be of high purity (as applications such as irrigation and stock watering have less stringent water quality compared to drinking water), DOW FILMTEC™ BW30XFRLE-400/34 elements were chosen in the first & second stage. These membrane modules were: made of polyamide thin-film composite material; characterized by a membrane area of 37 m²; could be used up to a pressure of 41 bar; and had a maximum flux rate of 48.4 L/m²/h [40]. Based upon the operating experience of DOW the design flux rate was assumed to be 22 L/m²/h, which was recommended for a micro-filtered wastewater sample. As the flow rate was 200 m³/h and 75 % recovery was targeted, the minimum number of membrane elements was estimated to be 185. Assuming 6 membrane elements per vessel, then a minimum of 31 pressure vessels were required. A staging ratio of 2 (calculated from Equation 1) indicated that 21 vessels should be in the first stage RO unit and 10 vessels in the second stage RO unit.

AqMB has a condition that if the water to be treated is higher than 10,000 mg/L then seawater RO membranes are required [25]. Therefore, the seawater membrane DOW FILMTEC™ SW30XLE-440 was selected for the first & second stages in the RO unit as it is recommended for high salinity brackish water treatment [41]. These membrane modules were again: made of polyamide thin-film composite material; characterized by a membrane area of 41 m²; could be used up to a pressure of 83 bar; and had a maximum flux rate of 38 L/m²/h [41]. From DOW guidelines, the design flux rate in this instance was assumed to be 22 L/m²/h as micro-filtered wastewater was being analysed. Since the flow rate was 200 m³/h and 75 % recovery was again required, the minimum number of membrane elements was estimated to be 167. Assuming 6 membrane elements per vessel, then a minimum of 28 pressure vessels were

required. A staging ratio of 2 indicated that 19 vessels should be in the first stage RO unit and 9 vessels in the second stage RO unit.

As with CSG 2 water, the TDS was above the 10,000 mg/L limit in AqMB for the use of a brackish water RO membrane. Therefore, it was appropriate to employ the same seawater membrane DOW FILMTEC™ SW30XLE-440 for modelling purposes [41]. Using the same assumptions as for CSG 2, at a staging ratio of 2, 19 vessels were required in the first stage RO unit and 9 vessels in the second stage RO unit. Operational costs for RO membranes were based upon: a unit cost of A\$450; life of 3 years; a lifecycle period of 20 years; 6 membrane changes in a lifetime period; and effective escalation rate (i) of 9.3 %.

2.4.10 pH Adjustment and Addition of Micronized Gypsum to Control Sodium Adsorption Ratio

An in-line dosing unit was simulated, which facilitated the addition of either sulphuric acid or lime as required. Calculations were made to ensure the Sodium Adsorption Ratio was adjusted to < 10 using micronized gypsum addition.

3. Results and Discussion

3.1 Pre-Treatment of CSG Associated Water

3.1.1 CSG Associated Water Sample 1

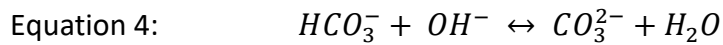
CSG operations utilize a holding pond as a repository for CSG associated waters from the various neighbouring wells prior to being passed to the remediation stage [4]. Computational analysis using AqMB to examine initial changes to the feedwater properties after settling in a pond for 240 hours is displayed in Table 4.

Table 4: CSG associated water 1 composition following pre-treatment stage

	Exit of Settling Pond	Exit of UF Modules	Exit of Ion Exchange Column (Na ⁺ Resin)	Units
TDS	3760	3747	3747	mg/L
Conductivity	5469	5490	5492	μS/cm
pH	9.08	9.09	9.06	
SAR	162.3	162.6	n.a.	
Aluminium	0.02	0.03	0.03	
Barium	0.67	0.67	0	mg/L
Bicarbonate	1132	1131	1142	mg/L
Boron	0.85	0.85	0.85	mg/L
Calcium	3.0	3.0	0.0	mg/L
Carbon Dioxide	1.20	1.20	1.20	mg/L
Carbonate	169.3	170.3	159.1	mg/L
Chloride	1103	1111	1111	mg/L
Fluoride	2.8	2.8	2.8	mg/L
Iron	0.11	0.03	0.03	mg/L
Magnesium	1.0	1.0	0.0	mg/L
Potassium	4.0	3.6	3.6	mg/L
Silica	21.7	19.9	19.9	mg/L
Sodium	1271	1273	1280	mg/L
Strontium	1.05	1.05	0.0	mg/L
Total Organic Carbon	50.0	16.2	16.2	mg/L
Dissolved Organic Carbon	48.5	16.2	16.2	mg/L
Total Suspended Solids	4.6	0.0	0.017	mg/L
LSI	1.17	1.17	-2.94	
Flowrate	214.8	197.6	197.6	m ³ /h
Turbidity	1.5	0	0.008	NTU

In general, the solution pH increased to 9.08 as did carbonate to 169 mg/L. Alternatively, total dissolved solids decreased slightly to 3760 mg/L and in harmony, the solution conductivity diminished to 5469 μS/cm. Sedimentation in the storage pond reduced suspended solids from 23 to 4.6 mg/L as expected for such a unit operation. The rise in

solution pH was ascribed to aeration of the storage pond and consistent with this assumption was the decrease in dissolved carbon dioxide content. In accord with the well-known behaviour of carbonate/bicarbonate equilibria as a function of solution pH, carbonate species instead of bicarbonate ions were promoted by the higher pH conditions [Equation 4].

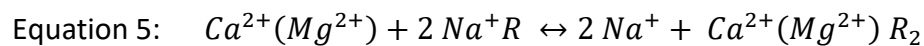


It was also predicted that ferrihydrite (FeOOH) precipitated in the feed at a rate of 1.01 mol/h and from the exit of the pond at 0.29 mol/h. Passage of the pond water through the sand ballasted clarifier, microfiltration, and ultrafiltration system was sufficient to reduce the TSS content to practically zero. Notably, the total flow of the CSG associated water was initially reduced to 184 m³/h due to losses in the clarifier (2.2 m³/h) and the ultrafiltration unit (13.8 m³/h). The need to extensively backwash the ultrafiltration modules was the primary source of the water loss. Consequently, one strategy to address this water loss was to recycle the backwash water to the settling pond. As an outcome, the flow rate from the exit of the ultrafiltration stage was increased to 197.6 m³/h.

It should also be considered that storage ponds inherently contain microorganisms such as algae and fungi [12]. Indeed, researchers have considered growing various algae species in CSG associated water [42, 43] and brine generated from reverse osmosis treatment [44]. For example, Aravinthan and Harrington [44] concluded that RO brine was a highly suitable medium for growth of *Dunaliella Tertiolecta*, which had relevance to biofuel manufacture. Likewise, Hodgskiss *et al.* [45] cultivated a green algae from a CSG associated water storage pond in the Powder River basin (USA) which was classified as a Chlorophyceae organism. Buchanan *et al.* [43] noted that the diversity of the microbial community growing in the CSG associated water was dependent upon the overall salinity and composition. It is also noted that the propensity for algal bloom formation in groundwater is also well known in regions such as Queensland [46]. Therefore, a suitable process design for CSG associated water treatment should encompass a strategy such as dosing appropriate biocides. Le [12] recommended a mixture of biocides was required to minimize microbiological resistance and increase effectiveness. This outlined approach should reduce the susceptibility of downstream membranes to fouling by microorganisms [13]. Hence, an inline dosing facility delivered 10 mg/L sodium hypochlorite solution post the clarification stage. The cost of the

sodium hypochlorite was estimated to be A\$90,388 per annum. It was assumed that the micro- and ultra-filtration units subsequently removed the microorganisms. AqMB did not have the capacity to explicitly monitor the removal of algae from the CSG associated water but the idea proposed was plausible based upon other studies [12]. The inclusion of ultrafiltration in the process design for the treatment of CSG associated water 1 was supported by the studies of both Nghiem *et al.* [35] and Qian *et al.* [36]. The CSG associated water samples were similar to that for CSG 1 water; 1351 mg/L sodium and 1317 mg/L bicarbonate [35]; and sodium 1852 mg/L and bicarbonate 1796 mg/L [36]. The software indicated that 62 UF modules were required for this water treatment system. Assuming a unit cost of A\$1800 and lifetime of 5 years for the UF modules, the present value was A\$385,069 (effective escalation rate = 15.9 %) which equated to an annualized cost of A\$19,253.

Simulation of an ion exchange unit comprising of a Na⁺-WAC resin was considered on the basis that the saturation index for calcite was 0.69, ferrihydrite 1.6, kaolinite 1.56, muscovite 2.996,), paragonite 2.99, and strontianite 0.78. Ion exchange softening with Na⁺-WAC resin was sufficient to remove all the calcium and magnesium ions as expected from Equation 5. In harmony, the concentration of sodium ions in the treated CSG water raised accordingly.



Where R = resin exchange site. Since CSG associated water is invariably alkaline in character [6], the selection of a WAC resin was preferred over a strong acid cation (SAC) resin. This decision was based upon the more efficient regenerability of a WAC resin; wherein a significantly reduced amount of acid is typically required to recover exchange capacity. The amount of resin required was 1400 L and the sodium chloride consumption per regeneration cycle was 559.7 kg. Weak acid cation ion exchange did not reduce the concentration of silicate species present in the CSG associated water. Since the formation of aluminosilicate deposits on reverse osmosis membranes is a known problem [47], use of a silica anti-scalant was employed at an annual cost of A\$865,904 per annum.

3.2.2 CSG Associated Water Sample 2

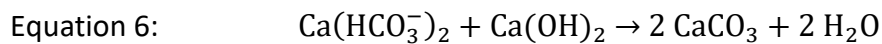
Table 5 shows the pre-treatment results for CSG associated water sample 2. Broadly, the changes in composition and physical properties in the settling pond mirrored those reported in section 3.2.1. Addition of a lime softener primarily reduced the concentration of calcium, and bicarbonate species along with the amount of suspended solids and total organic carbon present.

Table 5: CSG associated water 2 compositions following pre-treatment stage which included softening with lime

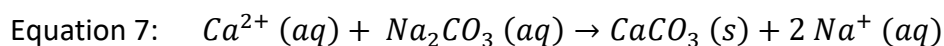
	Exit of Settling Pond	Option 1: Exit of Lime Softener	Option 2: Exit of Soda Ash Clarifier	Exit of UF Modules	Exit of Ion Exchange Column (Na⁺ Resin)	Units
TDS	10020	9887	11060	9885	9909	mg/L
Conductivity	17950	17860	19070	17860	17950	μS/cm
pH	8.23	9.55	9.93	9.61	9.41	
SAR	80.9	87.0	145.8	89.0	1607.8	
Barium	9.38	9.38	2.3	9.38	0.04	mg/L
Bicarbonate	181.3	42.6	264.1	35.2	46.3	mg/L
Boron	0.17	0.17	0.17	0.17	0.17	mg/L
Calcium	104.2	82.8	10.2	76.8	0.29	mg/L
Carbon Dioxide	1.20	0.01	0.03	0.01	0.01	mg/L
Carbonate	8.77	38.1	530.4	36.3	25.3	mg/L
Chloride	5908	5909	5908	5913	5913	mg/L
Fluoride	0.40	0.40	0.40	0.4	0.40	mg/L
Iron	0.65	0.02	0.02	0.001	0.001	mg/L
Magnesium	33.0	33.0	33.0	33.0	0.09	mg/L
Potassium	20.0	20.0	20.0	20.0	20.0	mg/L
Silica	14.0	14.0	14.0	14.0	14.0	mg/L
Sodium	3700	3700	4256	3703	3866	mg/L
Strontium	20.2	20.2	5.1	20.2	0.08	mg/L
Total Organic Carbon	46.7	23.4	19.3	12.1	12.1	mg/L
Dissolved Organic Carbon	23.4	22.6	18.7	12.1	12.1	mg/L
Total Suspended Solids	44.3	10	10	0	0	mg/L
LSI	0.93	1.97	2.45	1.97	-0.71	
SDSI	1.21	2.25	2.69	2.25	-0.42	
Flowrate	214.1	201.3	196.6	187.2	187.2	m ³ /h
Turbidity	10.5	4.6	4.7	0	0	NTU

AqMB indicated that calcium carbonate species precipitated at a rate of 11.36 mol/h in the overflow and 375.4 mol/h in the underflow. Ferrihydrite also formed at a rate of 0.07 mol/h in the overflow and 2.25 mol/h in the underflow. This observation explained both the reduction of calcium and iron ions seen in Table 5 after the lime softening stage. Similar

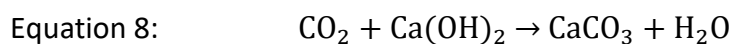
behaviour was described by Mohammadesmaeili *et al.* [48] who added lime to reverse osmosis brine. Calcium carbonate was said to form as illustrated in Equation 6.



Importantly, the degree of calcium ions removed from CSG associated water sample 2 was not substantial (*ca.* 21.6 %); hence, there was uncertainty as to the value of the lime softening stage. One issue may have been inhibition of calcium carbonate formation due to a lack of bicarbonate ions in solution. Blinov *et al.* [49] investigated the softening of oil sands produced water using combinations of lime, soda ash (Na_2CO_3) and sodium hydroxide (NaOH). An outcome of their study was the identification that soda ash could be more effective at softening produced water [Equation 7] which was in agreement with the data in Table 5.



Addition of lime or soda ash resulted in substantial removal of the relatively small quantity of the dissolved carbon dioxide noted due presumably to a combination of pH raising which moved equilibrium towards bicarbonate/carbonate and reaction of lime with carbon dioxide [Equation 8] [50].



No removal of magnesium ions was predicted when lime or soda ash was added to adjust the solution pH to 10. This calculation was consistent with the knowledge that magnesium hydroxide typically does not precipitate as magnesium hydroxide until pH values above 10 [50, 51]. Similarly, strontium ions were also not decreased by lime addition which was in agreement with the study of O'Donnell *et al.* [33] who did not record removal of strontium from drinking water until a pH of 10 if an excess of calcium ions and inorganic carbon was present. The observation of 21 % reduction in strontium presence when soda ash was used to soften the CSG associated water was thus also in agreement with the conclusions of these authors. Due to the relatively low solubility of barium carbonate, this species was additionally majorly precipitated after soda ash application. The amount of resin required was 35,660 L, and the sodium chloride consumption per regeneration cycle was 14,260 kg. These values for

resin and sodium chloride were greatly increased relative to the comparable figures when pre-treating CSG associated water 1. The significantly higher amounts of calcium and magnesium present in CSG associated water 2 were in line with the greater softening costs (annualized cost of A\$46,993 for resin).

3.2.3 CSG Associated Water Sample 3

Table 6 illustrates the results obtained from simulating pre-treatment of CSG water sample 3. The settling pond and filtration systems effectively removed suspended solids and the majority of organic carbon from the CSG associated water. Aeration of the CSG associated water in the settling pond raised the pH, reducing the iron content due to ferrihydrite formation (28.02 mol/h) and diminished alkaline earth ion concentrations (calcite 53.15 mol/h; strontianite 0.995 mol/h; hydrotalcite 0.003 mol/h; witherite 4.29 mol/h). The clarifier and ultrafiltration units subsequently removed these precipitates. Exploration of the saturation indices for the species in the filtrate from the UF unit suggested that calcite (SI = 1.132), ferrihydrite (SI = 1.584), muscovite (SI = 2.925), paragonite (SI = 2.219), and strontianite (1.426) may be problematic. Thus an ion exchange unit was considered as being one means to alleviate the outlined problem.

Two approaches to ion exchange pre-treatment were considered: (1) softening using a weak acid cation resin with sodium ions on the exchange sites; (2) demineralization/decarbonation using a strong acid cation resin coupled with a downstream degassing unit. The second option was investigated on the basis of the high concentration of bicarbonate ions present in CSG associated water 3. Employment of Na⁺-WAC resin was shown to be highly effective at removing dissolved calcium, magnesium, strontium, and barium ions [Table 6] with minimal impact upon remaining water quality indicators. The predicted resin volume was 2390 L and the amount of NaCl regenerant was 955.3 kg (100 % basis) per regeneration cycle.

In contrast, use of the H⁺-SAC resin removed a substantial proportion of the TDS (18,560 to 6177 mg/L) and the solution alkalinity (bicarbonate 4506 to 1588 mg/L; carbonate 4754 to 23.5 mg/L). The reduction in TDS was in accord with a proven performance of cation resins to exchange sodium ions [52], and potassium ions [53] and to decarbonize solutions [54]. Wicks *et al.* [23] demonstrated that demineralization of CSG associated water with an H⁺-SAC

resin created 1328 mg/L of dissolved carbon dioxide due to decomposition of bicarbonate/carbonate species under acidic conditions.

Table 6: CSG associated water 3 compositions following pre-treatment stage

	Exit of Settling Pond	Exit of UF Modules	Exit of Ion Exchange Column (Na ⁺ WAC Resin)	Exit of Degasser (H ⁺ SAC Resin)	Units
TDS	18560	18560	18570	6177	mg/L
Conductivity	21370	21370	21390	9415	μS/cm
pH	9.60	9.59	9.59	7.97	
SAR	346.5	625.2	n.a.	1348.3	
Aluminium	0.01	0.11	0.11	0.11	mg/L
Barium	3.43	0.67	0	0.008	mg/L
Bicarbonate	4435	4506	4531	1588	mg/L
Boron	1.3	1.3	1.3	1.3	mg/L
Calcium	13.9	3.9	0	0.06	mg/L
Carbon Dioxide	1.20	1.24	1.24	1.01	mg/L
Carbonate	4841	4754	4732	23.5	mg/L
Chloride	2324	2330	2330	2327	mg/L
Fluoride	6.5	6.5	6.5	6.5	mg/L
Iron	7.7	0.4	0.4	0.4	mg/L
Magnesium	9.4	3.1	0	0.08	mg/L
Potassium	82.5	82.2	82.2	11.25	mg/L
Silica	25	23.9	23.9	23.8	mg/L
Sodium	6822	6816	6830	2145	mg/L
Strontium	1.94	1.54	0	0.025	mg/L
Sulphate	17.8	17.8	17.8	17.8	mg/L
Total Organic Carbon	1.34	0.64	0.9	0.64	mg/L
Dissolved Organic Carbon	1.24	0.64	0.9	0.64	mg/L
Total Suspended Solids	37.0	0	0.01	0.84	mg/L
LSI				-1.61	
SDSI	3.23	2.72	-0.54		
Flowrate	213.9	193.1	180.1	193.4	m ³ /h
Turbidity	18.5	0	0.005	0.42	NTU

A reduction in this value to 1 mg/L was noted after use of the degassing unit. However, despite the success of the degassing system, it is advisable to evaluate the power consumption as air stripping can be costly [55]. The required power for the feed pump and air blower present in the degasser unit was estimated to be 3.279 kWh/kL (out of a total treatment plant consumption of 4.05 kWh/kL). Hence, degassing may not be favoured due to economic concerns. In addition, acid exchanged cation exchange resins although effective

at reducing the content of cations in CSG associated water; may not be best used when high TDS is present. This idea arises from the fact that regeneration chemicals are typically the major cost when employing ion exchange [56]. As such, ion exchange demineralization is ideally suited to solutions with relatively low TDS content [56]. Inspection of the resin demand (1,951,000 L) and HCl regenerant consumption (285,400 kg per regeneration cycle) confirmed that ion exchange demineralization prior to the central RO stage was not feasible.

3.3 Reverse Osmosis Desalination

3.3.1 CSG Associated Water Sample 1

Application of a two-stage RO process to desalinate CSG associated water 1 was investigated [Table 7]. Notably, the permeate quality satisfied regulatory requirements for beneficial reuse for irrigation purposes in Queensland (conductivity <950 mS/cm; pH 6 to 8.5) [57]. Mallants *et al.* [58] confirmed the minimal impact of using RO treated CSG associated water upon plant growth. Moreover, Majee *et al.* [59] demonstrated that even if the RO system did not treat CSG associated water to the standard shown in Table 7, damage to soil was not significant.

Shin and Bae [60] used a pilot plant RO unit to treat an “average Queensland” CSG associated water originally containing 4,450 mg/L TDS. In their study, the final TDS of the treated water was *ca.* 70 mg/L, which was similar to the value of 124 mg/L calculated in Table 7. The predicted water recovery rate of the RO unit was 74 %, and the energy consumption of the RO system was estimated to be 0.763 kWh/kL. Nghiem *et al.* [35] also used reverse osmosis to desalinate a CSG associated water sample at pilot scale and obtained a water recovery of 76 %. Hence, the simulated and actual data were comparable.

The SAR value of the RO permeate was initially infinite due to the lack of either calcium or magnesium ions in solution. An additional 4.5 mg/L of calcium ions was needed in order to change the SAR value to approximately 5 (to comply with regulations which indicated SAR < 6 for heavy soils and < 12 for light soils [57]). This amount of calcium ions necessitated dosing of 15.3 mg/L of micronized calcium sulphate, which corresponded to 2.26 kg per hour of gypsum.

Table 7: Simulation data for reverse osmosis treatment of CSG associated water sample 1

	Brine from BWRO Stage 1	Brine from BWRO Stage 2	Permeate Storage Tank	Units
TDS	7809	15380	124.2	mg/L
Conductivity	10420	19620	185.8	µS/cm
pH	8.90	8.74	8.5	
SAR	n.a.	n.a.	n.a.	
Aluminium	0.03	0.02	0.03	mg/L
Barium	0	0	0	mg/L
Bicarbonate	2264	4443	47.4	mg/L
Boron	0.76	0.57	0.94	mg/L
Calcium	0	0	0	mg/L
Carbon Dioxide	0	0	0.23	mg/L
Carbonate	303	594.5	0.84	mg/L
Chloride	2203	4343	29.4	mg/L
Fluoride	5.53	10.84	0.11	mg/L
Iron	0.03	0.04	0.025	mg/L
Magnesium	0	0	0	mg/L
Potassium	7.1	13.85	0.14	mg/L
Silica	39.4	77.8	0.47	mg/L
Sodium	2533	4986	38.8	mg/L
Strontium	0	0	0	mg/L
Total Organic Carbon	30.7	58.2	2.10	mg/L
Dissolved Organic Carbon	30.7	58.2	2.09	mg/L
Total Suspended Solids	0.22	0.20	0.25	mg/L
LSI			-6.22	
SDSI				
Flowrate	98.9	49.6	148	m ³ /h
Turbidity	0.09	0.10	0.12	NTU

3.3.2 CSG Associated Water Sample 2

For CSG associated water 2, a two-stage RO system was again used to desalinate the pre-treatment sample [Table 8]. The permeate quality was similar to that displayed in Table 7. However, the impact of the higher TDS CSG associated water composition was apparent when considering the brine concentration. In this case the brine had a TDS value of 48,340 mg/L (*c.f.* 15,380 mg/L for CSG associated water 1). The water recovery rate was 74.25 % which was close to the target of 75 %. The electricity demand for the two RO booster pumps was estimated to be 0.769 kWh/kL. The present value of the membranes employed was A\$415,108, which equated to an annualized cost of A\$20,755.

Table 8: Simulation data for reverse osmosis treatment of CSG associated water sample 2

	Brine from BWRO Stage 1	Brine from BWRO Stage 2	Permeate Storage Tank	Units
TDS	22070	48340	126.6	mg/L
Conductivity	37740	72160	261.1	µS/cm
pH	8.95	8.73	8.5	
SAR	2372.3	3487.0	301.1	
Barium	0.08	0.18	0	mg/L
Bicarbonate	118.8	260.9	0.85	mg/L
Boron	0.33	0.59	0.06	mg/L
Calcium	0.63	1.39	0.002	mg/L
Carbon Dioxide	0	0	0.004	mg/L
Carbonate	37.8	82.2	0.015	mg/L
Chloride	12910	28270	74.5	mg/L
Fluoride	0.87	1.91	0.006	mg/L
Iron	0.003	0.007	0	mg/L
Magnesium	0.20	0.45	0	mg/L
Potassium	43.8	96.3	0.08	mg/L
Silica	30.4	66.0	0.42	mg/L
Sodium	8440	18490	48.9	mg/L
Strontium	0.17	0.37	0	mg/L
Total Organic Carbon	25.3	52.9	1.49	mg/L
Dissolved Organic Carbon	25.3	52.9	1.49	mg/L
Total Suspended Solids	0	0	0	mg/L
LSI			-5.50	
SDSI				
Flowrate	85.4	38.8	148.5	m ³ /h
Turbidity	0	0	0	NTU

To reduce the SAR value of the RO permeate to 5, an additional 7.3 mg/L of calcium ions was required. Consequently, dosing of 24.8 mg/L of micronized calcium sulphate was required which equated to 3.68 kg per hour of gypsum.

3.3.3 CSG Associated Water Sample 3

Finally, the RO treatment of CSG associated water 3 was simulated. The permeate from the 2-stage RO unit was of high quality [Table 9] with the exception of the SAR value which was unacceptably high for irrigation purposes [57]. As the permeate pH was 6.1 there was scope to add some lime to raise the permeate pH to 8.5 and concomitantly decrease SAR due to the presence of calcium ions. Indeed, the SAR value was reduced from 215.6 to 2.16, and the amount of lime required was calculated as 3.74 kg/h. The cost for lime used to adjust the SAR value of the RO permeate was predicted to be A\$0.01 per hour.

Table 9: Simulation data for reverse osmosis treatment of CSG associated water sample 3

	Brine from SWRO Stage 1	Brine from SWRO Stage 2	Combined permeate from RO stages	Permeate Storage Tank	Units
TDS	13350	32950	105.4	127.5	mg/L
Conductivity	19470	44050	136.6	194.2	μS/cm
pH	8.04	7.86	6.1	8.5	
SAR			215.6	2.16	
Aluminium	0.22	0.54	0.006	0.006	mg/L
Barium	0.02	0.04	0	0	mg/L
Bicarbonate	3351	8211	20.98	54.5	mg/L
Boron	2.1	3.1	0.89	0.89	mg/L
Calcium	0.14	0.34	0.0004	12.2	mg/L
Carbon Dioxide	0	0	25.35	0.26	mg/L
Carbonate	88.9	249.3	0.02	1.26	mg/L
Chloride	5035	12430	29.8	29.8	mg/L
Fluoride	14.1	34.7	0.09	0.09	mg/L
Iron	0.84	2.05	0.02	0.02	mg/L
Magnesium	0.18	0.45	0.0005	0	mg/L
Potassium	24.4	60.5	0.05	0.05	mg/L
Silica	51.3	125.5	0.73	0.73	mg/L
Sodium	4640	11460	27.4	27.4	mg/L
Strontium	0.05	0.13	0.0002	0	mg/L
Sulphate	38.7	96.1	0.02	0.02	mg/L
Total Organic Carbon	1.33	3.14	0.08	0.08	mg/L
Dissolved Organic Carbon	1.33	3.14	0.08	0.08	mg/L
Total Suspended Solids	2.21	5.46	0.04	0.04	mg/L
LSI				0.02	
SDSI	-0.67	-0.40			
Flowrate	89.0	35.8		157.5	m ³ /h
Turbidity	1.11	2.73		0.02	NTU

The water recovery rate was 78.75 % which was slightly above the targeted design value of 75 %. The brine concentration of 32,950 mg/L was in harmony with reported brine concentrations in the region of *ca.* 40,000 mg/L when treating CSG associated water with reverse osmosis [61]. In terms of operational costs the electricity demand for the two RO booster pumps was estimated as 0.769 kWh/kL. The present value of the membranes employed was A\$429,934, which equated to an annualized cost of A\$21,497.

3.4 Comparison of Simulation Data with Industry Practice

This study has suggested that the best available technology for the treatment of CSG associated water is: (1) settling pond; (2) clarification; (3) micro/ultrafiltration; (4) softening with Na⁺- resin; (5) reverse osmosis; (6) pH adjustment; and (7) SAR adjustment. Biocide addition is also recommended and the use of anti-scalants should also be considered. Australia Pacific LNG (APLNG) have indicated that at their Condabri site that the treatment operations are: settling pond; disc (200 μ)/microfiltration (0.1 μ); ion exchange softening using Na⁺-resins; two-stage RO; and disinfection using chloramines [61]. With regards to the settling ponds, the detention time was set to between 24 and 72 hours (whereas this study used 10 days residence time). However, APLNG aerated the ponds to accelerate degassing of carbon dioxide and settling of insoluble materials. It is also noted that at the APLNG Talinga site the residence time in the settling pond was 14 days on average as no aeration was employed [62]. This investigation included ultrafiltration as a pre-treatment stage to reverse osmosis as is commonly considered for brackish water desalination systems [63]. However, its use was not mandatory if a preceding microfiltration unit satisfactorily removed suspended solids. The RO unit reported by APLNG was a three-stage system as water recovery rates between 82 and 95 % were targeted. The higher recovery rates was in accord with guidelines from DuPont Water Solutions that 2 RO stages could recover 70 to 80 % water and addition of a 3rd stage RO may promote water recovery to *ca.* 85 to 90 % [38]. Thus it was apparent that AqMB software predictions mimicked actual industry practice and allowed rapid refinement/optimization of operational conditions.

4. Conclusions

Simulation of water treatment strategies specific to coal seam gas associated water has been demonstrated. The hypothesis that computer simulation may promote the selection of appropriate unit operations was proven, as it was found that the treatment train had to be tailored to the CSG associated water composition. This situation illustrated the challenges of the CSG industry wherein RO units and the accompanying pre-treatment operations must be able to cope with a wide range of CSG associated water conditions over the life of the

treatment plant. In all case the RO plants produced water of high quality, but the stability and performance of the treatment train depended upon the entire process design.

Pre-treatment of CSG associated water universally involved a storage pond to accommodate the various incoming streams on a daily basis. Filtration to remove suspended solids and disinfection to control the presence of microorganisms also were common to all treatment designs studied. For CSG associated water which is relatively soft, a coagulation/clarifier system may protect downstream unit operations from clogging with suspended solids. Practical experience will indicate if a clarifier is necessary or if filtration is sufficient. If the CSG associated water was very hard, then a lime softening system may be worthy of consideration. However, for CSG water dominated by chloride ions and not bicarbonate/carbonate species it may be required to dose sodium carbonate to the CSG associated water to promote precipitation of insoluble alkaline carbonates. For high TDS CSG associated water which comprised of substantial amounts of dissolved bicarbonate species, the implementation of both an ion exchange softening using acidic resins and subsequent degassing was simulated. Consequently, it was not recommended for use by the CSG industry on the basis of excessive costs.

The process configuration for treatment of a wide range of CSG associated water compositions was determined to comprise of: (1) settling pond; (2) disinfection; (3) micro/ultrafiltration; (4) water softening using Na^+ -resin; (5) anti-scalant for silicate control; (6) as a minimum a 2 stage RO unit using either brackish or seawater membranes as necessary (and a 3-stage system if water recoveries above 80 % were necessary. Final water quality adjustment to ensure pH and SAR values met regulatory guidelines was also proposed. This treatment methodology was consistent with data from CSG companies.

Overall, AqMB software was beneficial in that it simulated the entire treatment train for CSG associated water to make the effluent suitable for beneficial reuse. The robustness of the

outcomes was demonstrated in relation to industry practice and the approach detailed in this study allowed both water quality and economic considerations to be concomitantly evaluated.

5. Acknowledgements

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