

Softening of coal seam gas associated water with aluminium exchanged resins

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ABSTRACT

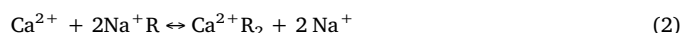
This study concerned aluminium exchanged strong acid cation resin for the removal of alkaline earth ions from simulated coal seam gas (coal bed methane) associated water samples which comprised of significant bicarbonate concentrations. The hypothesis was that use of aluminium exchanged resins for water softening would require only one regenerant stage, thus avoiding health and safety issues associated with strong acids and alkali on a coal seam gas water treatment site. Equilibrium exchange tests revealed that the selectivity of the aluminium exchanged resin was in the order $Ba > Sr > Ca > Mg$, albeit the exchange was unfavourable unless a stoichiometric excess of bicarbonate ions was present in solution. Equilibrium studies of multi-component solutions of alkaline earth ions indicated that at low loadings the alkaline earth ions co-sorbed on the resin surface, but the more preferred ion displaced ions of lesser affinity as monolayer exchange was approached. The presence of sodium ions not only reduced alkaline earth ion loading when in concentrations relevant to coal seam gas associated water application but also appeared to be incorporated into flocs in the form of salts. Column studies revealed significant loading of alkaline earth ions (0.787 eq/kg resin) but regeneration with $AlCl_3$ (aq) did not recover all these species (0.67 eq/kg resin). Partial restriction of the flow was noted during column tests due to floc formation. A subsequent loading/regeneration cycle resulted in further diminishment in alkaline earth ion uptake and an inability to achieve low effluent concentrations of alkaline earth ions.

1. Introduction

Softening of water samples comprising of excessive amounts of alkaline earth ions is a common procedure [1,2]. For example, laundry detergents comprise of zeolites which demonstrate high capacity for calcium and magnesium uptake [3]. Boiler water used in the power generation industry is also softened to inhibit scaling of equipment which would ultimately result in reduced operational efficiency [4]. More recently, unconventional energy sources have been developed such as coal seam gas (CSG) [5–7] which inherently produce significant volumes of brackish water which contains alkaline earth ions in addition to the major dissolved species of sodium chloride and sodium bicarbonate [8,9].

It is often necessary to desalinate the produced or associated water from the CSG industry in order to allow application of the water for beneficial reuse options such as irrigation or stock watering [9,10]. Reverse osmosis has emerged as the leading technology for desalination of coal seam (CS) water [11–13]. However, fouling of equipment and the membrane can occur due to the deposition of scale forming species

[14]. Therefore the use of anti-scalants or a softening stage is recommended [15,16]. Ion exchange softening can be achieved by use of either strong acid cation (SAC), weak acid cation (WAC) or chelating resins [2,17]. The basic concept of the exchange process is described in Eqs. (1) & (2).



For example, Jiang et al. [18] examined the exchange of calcium and magnesium ions with two chelating resins and discovered that regardless of the resin type very high removal efficiencies for both alkaline earth ions could be achieved (> 99.5%). Snoeyink et al. [19] applied strong acid cation resin to also successfully remove barium ions from solutions with significant alkalinity. Resins once loaded with alkaline earth ions were then regenerated in a two stage process as follows [Eqs. (3) & (4)] [20].



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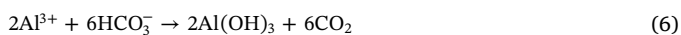
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The regeneration is a two stage process; first the resin is treated with a strong acid to remove the cations that have been exchanged onto the resin. Hydrochloric acid in the range 4–5% concentration is typically preferred as it avoids issues with precipitation of sulphates when sulphuric acid is employed [20]. To convert the resin to the original sodium ion exchanged form a solution of sodium hydroxide is used to remove the hydrogen ions from the resin [20].

Despite the demonstrated success of conventional ion exchange softening approaches, when considering the constraints of the CSG industry there is a desire to further improve aspects such as: use of only one regenerant stage to simplify operation; avoidance of health and safety issues associated with strong acids and alkali on a CSG water treatment site; and, minimisation of waste stream volumes. Thus an innovative strategy is required to address these latter requirements. It is noted that a characteristic of produced water from coal seam gas operations is high bicarbonate alkalinity relative to the amount of alkaline earth ions present [8,21,22]. A recent study by Li et al. [23] reported a new softening method which used aluminium exchanged resin to soften simulated surface water comprising of calcium, sodium, bicarbonate and sulphate ions at pH 7.5. The basis of this latter approach was to make use of the presence of bicarbonate ions in solution as illustrated in Eqs. (5) and (6), where R_2 and R_3 represent 2 and 3 resin exchange sites, respectfully [23].



The premise was to use aluminium loaded resins to remove the alkaline earth ions from solution. Normally, the exchange process in Eq. (5) would not be favourable due to the normal preference of resins for ions of higher charge [24]. However, the secondary reaction of bicarbonate ions with aluminium species conceptually provided a thermodynamic driver for the overall exchange process [23]. The formation of aluminium hydroxide which is commonly used as a coagulant also appears to be potentially advantageous in relation to purifying coal seam water by reducing the presence of silicates [25], turbidity and dissolved organic carbon [26,27]. Once the resin is loaded with alkaline earth ions, regeneration with 3% aluminium chloride solutions has been demonstrated by Li et al. [23] to be effective.

The aim of this study was to determine the feasibility of softening CSG associated water with aluminium exchanged resins and to gain an insight into parameters which influence resin performance. The hypothesis underpinning this investigation was that softening of coal seam gas associated water may be viable if the concentration of bicarbonate ions was sufficient to react with aluminium ions released from the resin to produce aluminium hydroxide flocs. Based upon published literature there were many issues which had not been addressed as yet including: (1) What are the loading characteristics of aluminium species with SAC resin; (2) How does solution bicarbonate concentration impact the uptake of alkaline earth ions; (3) Is there a preference for certain alkaline earth ions by aluminium exchanged resin; (4) What is the fundamental equilibrium behaviour of alkaline earth ions with aluminium loaded resin; (5) Can simulated CSG water be “treated” with aluminium loaded resins in columns; (6) What is the effectiveness of resin regeneration after treating simulated CSG water. Consequently, this study investigated an aluminium exchanged strong acid cation resin for the removal of alkaline earth ions from both simple solutions of individual alkaline earth ions and multicomponent solutions representative of coal seam water. Our approach was to use a combination of equilibrium and column tests to determine resin performance using both simple solutions of alkaline earth ions with bicarbonate ions present and more complex simulated CSG associated water.

2. Materials and methods

2.1. Resin

The resin was Lewatit MonoPlus S108H (S108) which is a strong acid cation (SAC) resin based on a styrene-divinylbenzene copolymer. The stated exchange capacity for this material was 2.0 eq/L and a packing density of 0.790 kg/L. The resin was used as received without further pre-treatment. Moisture content of the resin was measured by freeze drying under vacuum for the aluminium exchanged cation resin. Three resin samples (ca. 1–1.5 g) were weighed into 25 × 50 mm soda glass vials (0.0001 g accuracy) and then capped. The resins were dried using a CHRiST Alpha 1–4 LO plus vacuum freezing apparatus set to 0.044 mbar and –54.0 °C for no less than 24 h. The difference between the ‘wet’ weight of the resin and the ‘dry’ weight was calculated and converted to a% mass loss for each vial. The average% mass loss was found to be 55%.

2.2. Chemicals and CSG water composition

The chemicals used in this study ($\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$, $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$, $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, K_2CO_3 , $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$, $\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$, NaCl , and NaHCO_3) were all of analytical reagent (AR) standard (> 99% pure), with the exception of NaCl which was laboratory grade (LR) standard and NaHCO_3 which was food grade standard. All chemicals used were purchased from Chem-Supply. Solutions were prepared by addition of appropriate quantities of salt to purified water. The simulated CSG associated water sample had the approximate composition illustrated in Table 1 and was based upon extensive analysis of typical CSG associated water compositions found in the Surat Basin in Queensland [28]. At a pH of 8.75 it would be expected that some carbonate species were present in addition to bicarbonate ions, however tabulated is only the amount of bicarbonate as inferred from mass of sodium bicarbonate added to the solution. We note that simulated solutions were used in this study for several pertinent reasons. Firstly, as indicated in Section 1 a key research objective was to understand the impact of changing concentrations of bicarbonate ions in the saline solutions upon resin softening performance. Hence, it was necessary to prepare samples wherein precise ratios of alkaline earth ions and bicarbonate species were present. In terms of the CSG associated water sample, due to the remoteness and cost of sampling, it is challenging to obtain actual CSG associated water from an operating well. However, as shown in previous published investigations the difference in water quality between real and simulated CSG associated water is not substantial with the main parameter not addressed in simulated water being no boron (usually < 5 mg/L) [29].

2.3. Equilibrium isotherms

Equilibrium isotherms were conducted in 250 mL Nalgene containers, filled with approximately 100 mL of solution and a range of resin masses. The constant concentration bottle point method was used

Table 1
Composition of simulated CSG water used in equilibrium and column studies.

Species	Value	Units
Sodium	3190	mg/L
Potassium	77.5	mg/L
Calcium	12.3	mg/L
Magnesium	35	mg/L
Barium	10.6	mg/L
Strontium	14.4	mg/L
Chloride	4441	mg/L
Bicarbonate	1065	mg/L
Solution pH	8.75	

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