



Ion exchange of sodium chloride and sodium bicarbonate solutions using strong acid cation resins in relation to coal seam water treatment



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ABSTRACT

Coal seam gas exploration has resulted in the production of large volumes of associated water which contains dissolved salts dominated by sodium chloride and sodium bicarbonate. Ion exchange using synthetic resins has been proposed as a method for desalination of coal seam water to make it suitable for various beneficial reuse options. This study investigated the behaviour of solutions of sodium chloride and sodium bicarbonate with respect to exchange with Lanxess S108H strong acid cation (SAC) resin. Equilibrium isotherms were created for solutions of NaCl and NaHCO₃ and an actual sample of coal seam water from the Surat Basin in southern Queensland. The exchange of sodium ions arising from sodium bicarbonate was found to be considerably more favourable than exchange of sodium ions from sodium chloride solutions. This latter behaviour was attributed to the secondary decomposition of bicarbonate species under acidic conditions which resulted in the evolution of carbon dioxide and formation of water. The isotherm profiles could not be satisfactorily fitted by a single isotherm model such as the Langmuir expression. Instead, two Langmuir equations had to be simultaneously applied in order to fit the sections of the isotherm attributable to sodium ion exchange from sodium bicarbonate and sodium chloride. The shape of the isotherm profile was dependent upon the ratio of sodium chloride to sodium bicarbonate in solution and there was a high degree of correlation between simulated and actual coal seam water solutions.

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1. Introduction

Coal Seam Gas (CSG) represents a new and exciting industry which has the potential to supply the world with an abundant source of natural gas for the coming decades [1]. Developments have occurred in the USA in regions such as the Powder River and San Juan basins and in Australia in the Surat and Bowen basins [2]. The extraction of natural gas involves drilling wells into the coal seams which are usually between 200 and 1000 m deep. By reliev-

Abbreviations: C_e and $C_{e,Na}$, equilibrium concentration of sodium ions (mg/L); $C_{e,H}$, equilibrium concentration of protons (mg/L); C_0 , initial concentration of sodium ions (mg/L); CS, coal seam; CSG, coal seam gas; m, mass of resin (g); q_e and $q_{e,Na}$, equilibrium loading of sodium ions on resin (mg/g or mmol/g); $q_{e,H}$, equilibrium loading of protons on resin (mg/g or mmol/g); q_{max} , maximum loading of sodium ions on resin (mg/g or mmol/g); q_t , total ion loading on resin ($q_t = q_{e,Na} + q_{e,H}$); K_{LV} , equilibrium coefficient termed "half value"; $K_{Na/H}^+$, equilibrium coefficient (L/mg); R, resin; SAC, strong acid cation; V, solution volume (L).

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ing the water pressure, the CSG (predominantly methane) flows to the surface, whereupon is piped to a liquefied natural gas (LNG) plant [3]. The coal seam (CS) water which is co-produced with the natural gas contains various levels of dissolved salts, but is dominated by mixtures of sodium chloride and sodium bicarbonate [2,4,5]. The predicted volumes of CS water which will be collected in the forthcoming years in Queensland is significant and as it will be produced in regions where conditions are arid, there exists an opportunity to treat water for various beneficial reuse options. At present, the CSG industry has invested primarily in the application of reverse osmosis for CS water treatment [6,7]. Ion exchange on the other hand has found use in the Powder River Basin in the USA for demineralization of CS water [8,9]. As the Powder River Basin CS water is mainly composed of sodium bicarbonate ions, only a strong acid cation (SAC) resin bed is required for ion exchange as the acidic conditions decompose the bicarbonate species and the resin will exchange the majority of the sodium ions [8]. In the Australian context, the CS water can have a much higher content of sodium chloride [4] and as such it is important to understand how best to employ ion exchange technology in this latter situation.

The choice of materials considered for ion exchange applications includes natural zeolites and synthetic resins. The Vance group [10–12] in Wyoming have studied various natural zeolites to treat CS water from the nearby Powder River Basin CSG development. Zhao et al. [10] showed that calcium exchanged clinoptilolite could ion exchange sodium ions from solution and replace them with calcium ions thus lowering the sodium adsorption ratio (SAR) to values of less than 10. Reduction of the SAR value is important as when CS water with SAR in the range 17–57 was used for irrigation purposes, the soil was noted to not only exhibit increased electrical conductivity but also decreased surface infiltration and Darcy flux rates [13]. However, Huang and Natrajan [14] concluded that the process economics did not justify the use of natural zeolites for treatment of CS water.

The study of synthetic resins for treatment of CS water has received comparatively little study despite the fact that resins are probably the most widely used ion exchange medium in the world. Resins have many advantages compared to natural zeolites such as consistent quality, wide range of types to choose from, known hydraulic properties and the ability to be manipulated to enhance performance. SAC resins have been employed in the Powder River basin to treat CS associated water particularly as part of the Higgs Loop process which is a continuous ion exchange system [8,9]. Recently, Drake [15] reported the outcomes of kinetic studies of sodium bicarbonate solutions with a SAC resin. Notably, Drake [15] did not expand these studies to include detailed equilibrium tests. Our research group has recently conducted an investigation of sodium ion exchange from a range of sodium chloride solutions with different normalities on a SAC resin [16] and like other publications it found that it was important to select an appropriate bottle-point method to generate equilibrium isotherms [17,18]. The “constant concentration bottle point method” suggested maximum sodium ion exchange levels of 61.7–67.5 Na g/kg resin. Column trials confirmed the outcomes of the equilibrium tests with maximum sodium loading of ca. 62.9 g Na/kg resin recorded.

As outlined above, CS water does not simply comprise of sodium chloride species in solution, but contains significant concentrations of sodium bicarbonate. Consequently, the main objective of this study was to evaluate the fundamental differences in equilibrium exchange behaviour of sodium ions in either chloride, bicarbonate or mixed chloride/bicarbonate solutions with a SAC resin. The results of these latter tests were then compared with equilibrium exchange data of sodium ions from actual CS water sourced from an operating gas field. The aim was to understand the nature of the sodium exchange process and to gain an appreciation of how to treat CS water using ion exchange technology.

2. Material and methods

2.1. Resins

A SAC resin supplied by Lanxess termed “S108H” was used, which was a gel type resin in the hydrogen form based upon styrene-divinylbenzene co-polymer with a stated ion exchange capacity of 1.8–2.0 eq/L. The resin was used as supplied without any further pre-treatment.

2.2. Chemicals

Aqueous solutions were prepared using triple distilled water to which appropriate amounts of salt were added. Analytical reagent grade sodium chloride, sodium bicarbonate and concentrated hydrochloric acid (35 wt/v%) were supplied by Rowe Scientific. Surat Basin CS water was provided by a CSG operator from southern Queensland with the composition shown in Table 1. Also provided

is some data from Nghiem et al. [19] which allows comparison between the CS water sample used in this study with another which was sourced from the Gloucester Basin in New South Wales, Australia.

2.3. ICP analysis

Samples were analysed using an Agilent ICP-MS 7500CE instrument. Samples were diluted to a concentration between 10 and 50 mg/L using a Hamilton auto-dilutor with 10 and 1 mL syringes. A certified standard from Australian Chemical Reagents (ACR) containing 1000 ppm of sodium was diluted to form a multi-level calibration curve. An external reference was used to monitor instrument drift and accuracy of the results obtained. Results were obtained using an integration time of 0.15 s with 10 replications.

2.4. Equilibrium studies

Appropriate amounts of “wet” resin were weighed on a balance and then placed in a series of 250 mL Nalgene flasks. 200 mL of solution was then added to each bottle whereupon the system was sealed. Based upon kinetic studies the samples were equilibrated in a temperature controlled incubator (Innova 42R, New Brunswick Scientific) for a period of 24 h which was more than sufficient for equilibrium to be attained. The solution temperature was 30 °C and the samples were shaken at 200 rpm. The resultant solution was separated from the solid resin particles and concentrations of the sodium ions measured as well as solution pH and conductivity. Eq. (1) was then employed to estimate the equilibrium concentration of sodium ions (C_e) on the resin (q_e (mg/g)):

$$q_e = \frac{V}{m} (C_0 - C_e) \quad (1)$$

Experiments were conducted in duplicate.

2.4.1. Langmuir Vageler equation

Application of the Langmuir Vageler model [20,21] has been shown to be highly useful when investigating equilibrium ion exchange behaviour [17,18] [Eq. (2)].

$$q_e = \frac{(VC_0/m) q_{max}}{((VC_0/m) + K_{LV})} \quad (2)$$

The Langmuir Vageler model can be used to demonstrate that the experimental conditions chosen were appropriate for generation of a complete isotherm profile wherein maximum loading on the resin was achieved.

2.4.2. Competitive Langmuir model

The Competitive Langmuir model is applied when investigating ion exchange processes as unlike the Langmuir equation it includes both the exchanging ions involved [22]. For the exchange process represented in Eq. (3), from the law of mass action [Eq. (4)] can be derived.



$$q_{e,Na} = \frac{k q_t C_{e,Na}}{C_0 + (k - 1) C_{e,Na}} \quad (4)$$

2.4.3. Error functions

Non-linear least squares (NLLS) fitting procedures were applied to equilibrium isotherms [23,24]. The methodology of Ho et al. [25] was applied to determine a suitable error function as required by the Solver add-on in Microsoft Excel. Consequently, five functions were used: sum of errors squared (ERRSQ); hybrid fractional error function (HYBRID); Marquardt’s percent standard deviation

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