



# Characterizing long-term CO<sub>2</sub>–water–rock reaction pathways to identify tracers of CO<sub>2</sub> migration during geological storage in a low-salinity, siliciclastic reservoir system



Kyle N. Horner<sup>a,b,\*</sup>, Ulrike Schacht<sup>a,c</sup>, Ralf R. Haese<sup>a,d</sup>

<sup>a</sup> CRC for Greenhouse Gas Technologies, Ground Floor, NFF House, 14-16 Brisbane Ave., Barton, ACT 2600, Australia

<sup>b</sup> Geoscience Australia, GPO Box 378, Canberra 2601, Australia

<sup>c</sup> Australian School of Petroleum, University of Adelaide, Adelaide, SA 5005, Australia

<sup>d</sup> Peter Cook Centre for Carbon Capture and Storage Research, School of Earth Sciences, The University of Melbourne, Parkville, Victoria 3010, Australia

## ARTICLE INFO

### Article history:

Accepted 29 September 2014

Available online 13 October 2014

Editor: David R. Hilton

### Keywords:

Carbon dioxide

Sequestration

CCS

Low-salinity

Surat Basin

## ABSTRACT

Given the prevailing use of saline reservoirs for geological CO<sub>2</sub> storage projects, limited data are available on the geochemical evolution of formation water chemistry during geological CO<sub>2</sub> storage in low-salinity formations. The low-salinity (total dissolved solids < 3000 mg/L) middle to lower Jurassic sequence in Australia's Surat Basin has been characterized as a potential reservoir system for geological CO<sub>2</sub> storage, comprising three major siliciclastic formations with distinctly different mineral compositions. Contrasts in the geochemical responses of Jurassic sequence core samples have been identified during short-term CO<sub>2</sub>–water–rock experiments conducted under CO<sub>2</sub> storage conditions (Farquhar et al., in this issue). If persistent, such contrasts may serve as geochemical tracers of CO<sub>2</sub> migration within the Surat Basin. Here we use a combined batch experiment and numerical modeling approach to characterize the long-term response of the Jurassic succession to storage and migration of CO<sub>2</sub> and to assess reaction pathway sensitivity to CO<sub>2</sub> partial pressure. Reservoir system mineralogy was characterized for 66 core samples from Geological Survey of Queensland stratigraphic well Chinchilla 4, and six representative samples were powdered and reacted with synthetic formation water and high-purity CO<sub>2</sub> for up to 27 days at a range of pressures. Formation water alkalinity offers limited buffering at elevated CO<sub>2</sub> pressures and pH rapidly declines resulting in sustained enhancement of mineral dissolution rates. Batch reactor results exhibit regional groundwater-like <sup>87</sup>Sr/<sup>86</sup>Sr values (0.7048–0.7066), less radiogenic than whole-rock results (>0.7085) indicating incongruent dissolution of the reservoir matrix. Carbonate and authigenic clay dissolution are expected to be the primary reaction pathways regulating long-term formation water composition during geological CO<sub>2</sub> storage in the Surat Basin, with lesser contributions from dissolution of the clastic matrix.

Crown Copyright © 2014 Published by Elsevier B.V. All rights reserved.

## 1. Introduction

Carbon capture and storage (CCS) is a relatively new technology for mitigating anthropogenic climate change by separating CO<sub>2</sub> from industrial flue gas, transporting it to and storing it in a subsurface geological reservoir. Depleted oil and gas fields, coal seams and deep aquifers have been identified as suitable reservoirs for geological CO<sub>2</sub> storage, in principle (Bachu and Adams, 2003; Gunter et al., 2004; Benson and Cole, 2008). However, such prospective storage reservoirs may either contain potable water or be located adjacent to other reservoirs that contain potable water. As CO<sub>2</sub>–water–rock interactions can impact formation water composition, some regulators have restricted geological

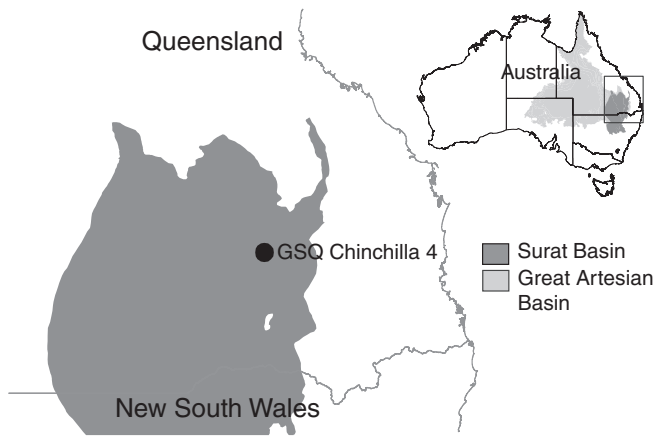
CO<sub>2</sub> storage to reservoirs containing saline water. Where restrictions are imposed, minimum reservoir salinity thresholds for geological CO<sub>2</sub> storage range from 3000 mg/L to 10,000 mg/L of total dissolved solids (TDS) depending on jurisdiction (Bachu et al., 2007; USEPA, 2010; EUCA, 2011).

Minimum reservoir salinity threshold values generally do not apply to geological CO<sub>2</sub> storage projects in Australia (e.g. GGGSA, 2008). Therefore several prospective Australian reservoir systems have lower salinity than is typical of storage reservoirs in other countries (Carbon Storage Taskforce, 2009). One such reservoir system is the Jurassic sequence within the Queensland portion of the Surat Basin (Fig. 1) where formation water salinity is <3000 mg/L (Bradshaw et al., 2011; Grigorescu, 2011a; Hodgkinson and Grigorescu, 2012; Feitz et al., 2014).

CO<sub>2</sub>–water–rock interactions in saline formations have been extensively examined (e.g. Johnson et al., 2004; Allen et al., 2005; Druckenmiller and Maroto-Valer, 2005; Xu et al., 2005; Zerai et al.,

\* Corresponding author at: New South Wales Environment Protection Authority, Sydney, NSW 2000, Australia.

E-mail address: [kyle.horner@epa.nsw.gov.au](mailto:kyle.horner@epa.nsw.gov.au) (K.N. Horner).

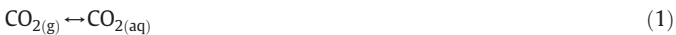


**Fig. 1.** Location of the Surat Basin and stratigraphic bore GSQ Chinchilla 4 within the Queensland portion of the Surat Basin.

2006; Ketzer et al., 2009; Huq et al., 2012; Okuyama et al., 2013). Due to the prevalence of saline waters in deep reservoirs and the jurisdictional moratoria on CCS in fresher aquifers, fewer studies have explicitly examined low-salinity formations under CO<sub>2</sub> storage conditions (e.g. Pashin et al., 2003; Parry et al., 2007; Farquhar et al., 2015—in this issue).

### 1.1. Salinity and CO<sub>2</sub> storage

CO<sub>2</sub> solubility is greatest in fresh water, declining with increasing salinity at any given temperature and pressure (Helgeson, 1969; Portier and Rochelle, 2005; Duan et al., 2006). During geological storage of supercritical carbon dioxide (scCO<sub>2</sub>), solubility and density effects result in structural trapping of a non-aqueous scCO<sub>2</sub> plume at the top of the reservoir. Residual quantities of scCO<sub>2</sub> also become trapped within the reservoir matrix by capillary forces (Lindeberg and Wessel-Berg, 1997; Pruess, 2007; Gaus, 2010). This physically trapped scCO<sub>2</sub> gradually dissolves into formation water as CO<sub>2(aq)</sub> (Eq. (1)).



CO<sub>2</sub> dissolution increases formation water density, particularly at the scCO<sub>2</sub>/water interface, creating a vertical density gradient that drives convection within the reservoir (Lindeberg and Wessel-Berg, 1997). Convective transport of dense water away from the scCO<sub>2</sub>/water interface has a major influence on CO<sub>2(aq)</sub> distribution, with bulk CO<sub>2(aq)</sub> concentrations in a reservoir below the maximum solubility limit (Gasda et al., 2011). As fresh waters have greater CO<sub>2</sub> solubility and lower fluid density than more saline waters, vertical convection should have a significant effect on CO<sub>2(aq)</sub> distribution in low-salinity formations (Ennis-King and Paterson, 2007).

Most dissolved CO<sub>2</sub> remains in solution as CO<sub>2(aq)</sub> but a small proportion hydrates to form carbonic acid, acidifying the formation water (Eq. (2)). The extent of acidification depends upon CO<sub>2(aq)</sub> concentrations and the reservoir's alkalinity which can buffer pH change by limiting carbonic acid dissociation (Eqs. (3) and (4)). Low-salinity reservoirs such as the Surat Basin can have low alkalinity (e.g. Herczeg et al., 1991; Feitz et al., 2014) and therefore generally have higher acidification

potential due to increased CO<sub>2</sub> solubility and decreased pH buffering capacity compared to saline formations. Thus a marked decrease in pH is expected following CO<sub>2</sub> enrichment of low-salinity reservoirs. As dissolution rates of many common rock-forming silicate and carbonate minerals are enhanced under acidic conditions (e.g. Black et al., in this issue) geological CO<sub>2</sub> storage in low-salinity reservoirs may result in significant mineral dissolution.

Studies of CO<sub>2</sub> leakage into shallow, low-salinity aquifers under near-surface temperature, pressure, and/or redox conditions have illustrated the impacts of minor pH changes on formation water chemistry in the near-surface environment (e.g. Carroll et al., 2009; Keating et al., 2009; Smyth et al., 2009; Little and Jackson, 2010; Lu et al., 2010; Nondorf et al., 2011; Peter et al., 2012; Harvey et al., 2013; Humez et al., 2013; Yang et al., 2014). CO<sub>2</sub>–water–rock studies using ultra-pure distilled water as the reaction fluid indicate geochemical impacts will be more extensive during geological CO<sub>2</sub> storage in low-salinity formations (e.g. Ketzer et al., 2009; Huq et al., 2012; Terzi et al., 2014). While such studies provide conceptual insights into geological CO<sub>2</sub> storage in low-salinity reservoirs, further research is required to constrain long-term reaction pathways and identify potential tracers of CO<sub>2</sub> migration under representative sequestration conditions.

This study aims to assess the evolution of long-term CO<sub>2</sub>–water–rock reaction pathways in low salinity, siliciclastic reservoirs under geological CO<sub>2</sub> storage conditions and to identify geochemical tracers of CO<sub>2</sub> migration within the Jurassic aquifers of the Surat Basin. Here, a combined batch experiment and numerical modeling approach is used to evaluate the long-term geochemical response of saturated Surat Basin Jurassic sandstone under a range of CO<sub>2</sub> pressures. Such an approach has previously been applied to characterize the results of CO<sub>2</sub>–water–rock experiments in both low-salinity (e.g. Farquhar et al., 2015—in this issue) and saline solutions (e.g. Fischer et al., 2014).

## 2. Study area

The Great Artesian Basin (GAB) is Australia's largest connected groundwater system, comprising numerous sedimentary sub-basins including the Jurassic Surat Basin (Fig. 1). GAB groundwater is consistently fresh to brackish (Herczeg et al., 1991) with TDS concentrations in the Surat Basin below 3000 mg/L (Grigorescu, 2011a; Feitz et al., 2014).

The Surat Basin was filled throughout the Jurassic and Cretaceous periods following Middle–Late Triassic deformation of the underlying Bowen and Gunnedah Basins and bedrock (Fig. 2; Hoffmann et al., 2009; Korsch and Totterdell, 2009). Surat Basin depth varies extensively, pinching out at its margins while exceeding 2500 m in regional depocenters. Extensive portions of the Jurassic sandstone formations meet the minimum permeability, pressure, and temperature requirements (31 °C, 74 bar) for geological storage of scCO<sub>2</sub>.

Comprehensive characterization of core samples from the Geological Survey of Queensland (GSQ) stratigraphic well GSQ Chinchilla 4 (CHIN-4) by Farquhar et al. (2013, 2015—in this issue) shows significant mineralogical differences between the Jurassic sandstone units which comprise the potential reservoir system in the Surat Basin (Fig. 2). The basal, fluvio-lacustrine Precipice Sandstone is up to 70 m thick and is a quartz-dominated (>90%) sandstone with minor kaolinite and trace feldspar exhibiting a fining-upward trend with variable bedding and a white clay matrix (Grigorescu, 2011b; Farquhar et al., 2013).

The fluvial-marine Early Jurassic Evergreen Formation unit conformably overlies the Precipice Sandstone and is up to 300 m thick. The Evergreen Formation is considered a potential seal unit for geological CO<sub>2</sub> storage, comprising fine to medium-grained fluvial sandstone and siltstone sequences transitioning to marine mudstones with sandstone laminae and with smectitic clays present throughout. The Boxvale Sandstone member of the Evergreen Formation is a fine to coarse-grained, cross-bedded, quartzose sandstone with abundant feldspar (30–50%). The Westgrove Ironstone member consists of siderite-cemented oolitic mudstones (Exon, 1976; Farquhar et al., 2013).

Download English Version:

<https://daneshyari.com/en/article/4698601>

Download Persian Version:

<https://daneshyari.com/article/4698601>

[Daneshyari.com](https://daneshyari.com)