



Experimental study of potential wellbore cement carbonation by various phases of carbon dioxide during geologic carbon sequestration



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ABSTRACT

Hydrated Portland cement was reacted with CO₂ in supercritical, gaseous and aqueous phases to understand the potential cement alteration processes along the length of a wellbore, extending from a deep CO₂ storage reservoir to the shallow subsurface during geologic carbon sequestration. The 3-D X-ray microtomography (XMT) images showed that the cement alteration was significantly more extensive with CO₂-saturated synthetic groundwater than dry or wet supercritical CO₂ at high P (10 MPa)-T (50 °C) conditions. Scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS) analysis also exhibited a systematic Ca depletion and C enrichment in cement matrix exposed to CO₂-saturated groundwater. Integrated XMT, XRD and SEM-EDS analyses identified the formation of an extensive carbonated zone filled with CaCO₃(s), as well as a porous degradation front and an outermost silica-rich zone in cement after exposure to CO₂-saturated groundwater. Cement alteration by CO₂-saturated groundwater for 2–8 months overall decreased the porosity from 31% to 22% and the permeability by an order of magnitude. Cement alteration by dry or wet supercritical CO₂ was slow and minor compared to CO₂-saturated groundwater. A thin single carbonation zone was formed in cement after exposure to wet supercritical CO₂ for 8 months or dry supercritical CO₂ for 15 months. An extensive calcite coating was formed on the outside surface of a cement sample after exposure to wet gaseous CO₂ for 1–3 months. The chemical–physical characterization of hydrated Portland cement after exposure to various phases of CO₂ indicates that the extent of cement carbonation can be significantly heterogeneous depending on the CO₂ phase present in the wellbore environment. Both experimental and geochemical modeling results suggest that wellbore cement exposure to supercritical, gaseous and aqueous phases of CO₂ during geologic C sequestration is unlikely to damage the wellbore integrity because cement alteration by all phases of CO₂ is dominated by carbonation reactions. This is consistent with previous field studies of wellbore cement with extensive carbonation after exposure to CO₂ for three decades. However, XMT imaging indicates that preferential cement alteration by supercritical CO₂ or CO₂-saturated groundwater can occur along the cement–steel or cement–rock interfaces. This highlights the importance of further investigation of cement degradation along the interfaces of wellbore materials to ensure permanent geologic carbon storage.

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1. Introduction

Industrialization since the 18th century has increased atmospheric CO₂ concentration from 280 ppm to nearly 400 ppm through combustion of fossil fuels, which is theorized to contribute to global warming (Hansen et al., 1981). A CO₂ capture and storage technology in deep geologic formations including oil and gas reservoirs, deep saline aquifers, coal seams and salt caverns has been proposed to decrease atmospheric CO₂ concentrations. Because of the buoyancy effect resulting from the difference in density between supercritical CO₂ and saline groundwater, injected CO₂ tends to escape upward into overlying formations and potentially to the land surface if a zone of high permeability exists (Gasda

et al., 2004). Potential CO₂ leakage can occur through wellbores, which can adversely affect underground sources of drinking water and consequently human health, as well as the ecosystem (Bruant et al., 2002; Little and Jackson, 2010; Wilkin and Digiulio, 2010).

Portland cement is commonly considered as a sealing material for wellbores for geologic carbon sequestration. During typical well construction, cement slurry is placed in the annulus between steel casing and formation rocks to prevent vertical fluid migration and provide mechanical support (Nelson and Guillot, 2006). Abandoned wells are also typically sealed with cement plugs inside the casing to block the vertical migration of fluids ((Kutchko et al., 2007). Hydrated products formed by mixing Portland cement with water are Ca–silicate–hydrate (C–S–H), a semi-amorphous gel-like material and portlandite [Ca(OH)₂(s)], a crystalline phase (Neville, 2004; Nelson and Guillot, 2006). Wellbore integrity could be damaged by poor completion or abandonment, formation

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damage around the wellbore, geomechanical effects, geochemical degradation of well cements, casing corrosion, and casing failure as a result of thermal or mechanical stresses (Bachu and Bennion, 2009). Potential leakage pathways of stored CO₂ may occur at the interface between casing and cement, cement plug and casing, and cement and host rock, or through the cement pore spaces and fractures (Gasda et al., 2004; Um et al., 2011; Jung et al., 2013). Cement carbonation resulting from solvation of Ca ions from the solid phase by diffused H₂CO₃ and precipitation of CaCO₃-minerals can affect both the chemical and physical properties of cement (Fernandez Bertos et al., 2004). The rate of cement degradation can vary depending on the pH of CO₂-saturated groundwater, which is affected by aquifer types (e.g., sandstone formation or limestone formation) (Duguid and Scherer, 2010) and cement curing conditions (e.g. temperature and pressure) (Kutchko et al., 2008). A number of studies have shown that wellbore cement is susceptible to CO₂ attack, leading to rapid degradation (Scherer et al., 2005; Barlet-Gouédard et al., 2006). Other experimental and field studies have indicated that alteration of hydrated cement by CO₂ under geologic sequestration conditions is limited, and the long-term performance of wellbore cement in a CO₂-enhanced oil recovery (EOR) field or a natural CO₂ production reservoir was good, providing an effective barrier to significant fluid flow for decades (Carey et al., 2007; Kutchko et al., 2008; Crow et al., 2010). However, it is difficult to compare the extent and rate of cement alteration by CO₂ reported from different laboratory experiments because of the wide variation in experimental conditions such as temperature, pressure, and cement curing (Kutchko et al., 2008; Zhang and Bachu, 2011).

During geologic carbon sequestration, cement materials in the injection well will be contacted by dry supercritical CO₂ because a dry-out zone is developed within a few meters of the injection well through displacement by supercritical CO₂ and evaporation of water into the CO₂ stream (Pruess and Muller, 2009; Zhang et al., 2011), while cement materials in the existing wells (e.g. EOR sites) can be exposed to wet supercritical CO₂ or CO₂-saturated formation water (Zhang and Bachu, 2011). In addition, wellbore cement at shallow depth above the groundwater table can be exposed to wet CO₂(g) leaked from the deep reservoir.

This study focused on the potential chemical and physical alteration processes of hydrated Portland cement by various phases of CO₂ that can be present along the wellbore from the deep carbon injection and storage depth (50 °C and 10 MPa) to the shallow subsurface (20 °C and 0.1 MPa) during geologic carbon sequestration. Portland cement columns (Type: I-II or II-V) that had been prepared and cured under the same *P-T* conditions (0.1 MPa and 20 °C; *w/c* = 0.33 or 0.38) were exposed to wet and dry supercritical, aqueous and gaseous phases of CO₂ over time scale of 1–15 months. The changes in physical and chemical properties of the carbonated Portland cement were investigated using a number of advanced characterization methods such as scanning electron microscopy with energy dispersive spectroscopy (SEM-EDS), X-ray diffraction (XRD), and X-ray microtomography (XMT) techniques.

2. Materials and methods

2.1. High pressure and temperature experiment

Cement paste was prepared by mixing Portland cement with water at a water-to-cement ratio of 0.33 (Ash Grove, Type II-V; comparable to Class G cement) or 0.38 (Lafarge North America, Type I-II; comparable to Class A cement) to achieve high strength. The Class A and G Portland cements are among the most commonly used wellbore cements in the oil and gas industry (Lea, 1970;

Nygaard, 2010). Both types of Portland cements primarily consist of CaO (64%) and SiO₂ (21–22%) as well as Al₂O₃ (3–4%), Fe₂O₃ (3–4%) and SO₃ (2–3%). Two different water-to-cement ratios (0.33 and 0.38) were used to explore the effect of water-to-cement ratio on cement carbonation. Cement samples were cast in the form of cylinders by pouring the slurry into 15-mL centrifuge tubes (14 mm diameter and 90 mm length) using Type II-V Portland cement (water-to-cement ratio of 0.33) or a plastic mold (7 mm diameter and 20 mm length or 13 mm diameter and 25 mm length) using Type I-II Portland cement (water-to-cement ratio of 0.38) (Table 1). Steel coupons (cement columns S-1, S-2, S-6, S-7, and DS-1; chemical composition: C = 0.28%, Mn = 1.16%, and Fe = 98.5% by weight) or basalt fragments from the Wallula CO₂ sequestration pilot site in Washington State, USA (cement columns S-5, S-10, and DS-3) were added to the cement paste in order to simulate cement-steel casing and cement-rock interfaces. All cement columns were sealed to prevent the evaporation of moisture and cured for 28 days at ambient pressure (0.1 MPa) and temperature (20 °C), with a relative humidity of 100%. The hardened cement columns were placed in a pressure vessel (Parr Instrument Company; 300 mL volume size with 64 mm internal diameter × 102 mm depth) containing 130 mL of synthetic groundwater (2 mM NaNO₃, 0.5 mM KNO₃, 0.5 mM CaSO₄, and 0.5 mM MgSO₄; TDS = 348 mg/L) to simulate the groundwater composition (TDS = 332 mg/L) of Wallula Basalt Carbon Dioxide Sequestration Pilot Site. The experiment was conducted under a pressure of 10 MPa and a temperature of 50 °C to represent the pressure and temperature condition at the CO₂ injection depth of ~1 km below the surface assuming a geothermal gradient of 30 °C/km and a pressure gradient of 10.5 MPa/km. Under these pressure and temperature conditions, CO₂ is present in a supercritical phase. In vessels 1–4, small-sized cement columns (water-to-cement ratio of 0.38) of 7 mm diameter and 20 mm length or 13 mm diameter and 25 mm length were completely submerged in 130 mL of synthetic groundwater for 1–8 months, while a cement column (WS-1; 13 mm diameter and 25 mm length) was suspended over the groundwater and exposed to water-saturated wet supercritical CO₂ (Table 1). In vessels 5–8, the bottom half of each cement column (14 mm diameter and 90 mm length; water-to-cement ratio of 0.33) was submerged in synthetic groundwater saturated with CO₂ for 5 months, while the top half of the column was exposed to water-saturated wet supercritical CO₂ for 5 months (Table 1). In vessel 9, 3 cement columns (14 mm diameter and 90 mm length; water-to-cement ratio of 0.33) were exposed to dry supercritical CO₂ for 15 months. After the CO₂ reaction, the pressure vessels were slowly depressurized for 24 h, and then cement samples were collected and characterized using SEM-EDS, XRD and XMT (see Section 2.4 for details). Before SEM-EDS analysis, cement columns were cut into slices using a Buehler-ISOMET low-speed saw (Model#11-1280-160). Cement and CaCO₃ precipitate samples were ground into fine powder for XRD analysis.

2.2. Porosity and permeability measurement

Porosity and air permeability of duplicate cement columns (Type I-II; 25 mm diameter × 38 mm length; water-to-cement ratio = 0.38) before and after the reaction with CO₂-saturated groundwater for 2 and 8 months were measured by Core Laboratories using a CMS-300 Automated Permeameter after drying the columns at 115 °C. Porosity was determined using the Boyle's Law method by measuring grain volume at ambient conditions and pore volume at net confining stress (NCS) of 8.3 MPa. Air permeability was measured using the unsteady-state pressure decay method at an NCS of 8.3 MPa. Before and after the reaction of a cement column (13 mm diameter and 25 mm length; water-to-cement ratio = 0.38) with CO₂-saturated groundwater at 50 °C and

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