

Coupled effect of CO₂ attack and tensile stress on well cement under CO₂ storage conditions



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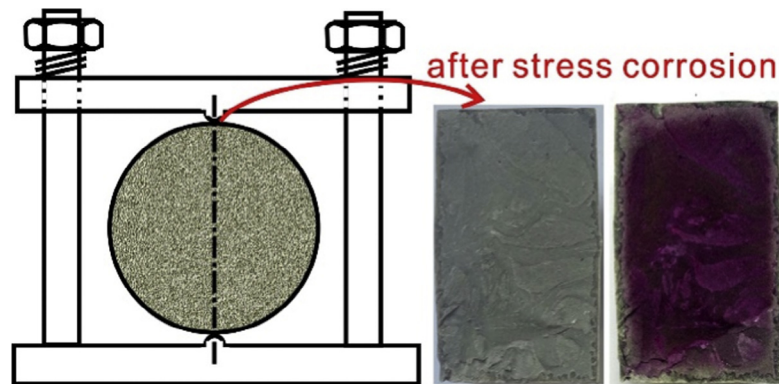
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HIGHLIGHTS

- Coupled effect of CO₂ attack and tensile stress on well cement under CCS condition is studied.
- A higher tensile stress will facilitate the generation and propagation of micro-cracks.
- The tensile stress can accelerate the failure of well cement.
- The carbonation rate of cement was faster in CO₂-saturated brine.

GRAPHICAL ABSTRACT



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ABSTRACT

In carbon capture and storage (CCS) wells, the well cement is attacked by CO₂-rich fluids, coupled with tensile stress. In this study, well cement samples were designed to be exposed to humid CO₂ gas and CO₂ saturated brine and simultaneously subjected to external tensile stresses with load levels of 25%, 50%, and 75% of the initial tensile strength. The experimental results showed that a higher external tensile stress (50% and 75%) facilitated the generation and propagation of micro-cracks in the tension zone of loaded samples. Hence, the aggressive CO₂-rich medium found direct paths to penetrate further into the core, significantly accelerating the rate of carbonation and failure of the well cement. As a result of the faster ion exchange and transfer, the carbonation rate was faster and the onset of failure in samples immersed in CO₂-saturated brine occurred earlier than in the supercritical CO₂ scenario. Findings from this study provide new, important information for understanding the integrity of well cement sheath under actual CCS well conditions, thereby promoting superior cement system design and safer operation.

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1. Introduction

Carbon capture and storage (CCS) technology, an effective solution for reducing anthropogenic emissions of CO₂, has received

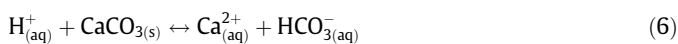
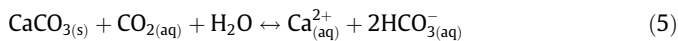
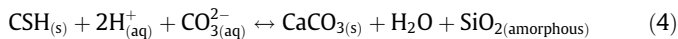
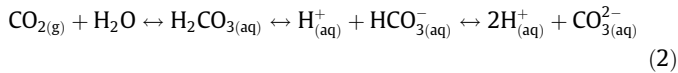
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increased attention in the past decade [1–3]. By the end of 2010, over 200 CCS projects were active or planned worldwide, and the first Chinese full-chain CCS demonstration project was implemented in 2010 by injecting 1.0×10^5 tons of supercritical CO₂ per annum down into brine-saturated low-permeability sandstone and carbonate aquifers at depths of more than 1620 m in the Ordos Basin [4–6]. Long-term seal integrity is one of the most important

factors for the long-term success and safety of CO₂ sequestration technology [7–9]. Portland cement (e.g. API Class G and H oilwell cement) is commonly used for CCS well cementing to isolate gas and fluid flow between different formations and to prevent leakage along the wellbore and out of the storage reservoir [10,11].

Generally, there are four major compounds in oilwell cement: tricalcium silicate (Ca₃SiO₅ or C₃S), dicalcium silicate (Ca₂SiO₄ or C₂S), tricalcium aluminate (Ca₃Al₂O₆ or C₃A), and tetracalcium aluminoferrite (Ca₄Al₂Fe₂O₁₀ or C₄AF). Moreover, 3–5% gypsum is added to clinker to prevent flash setting. C₃S, C₂S, C₃A, and C₄AF are hydrated when the cement is mixed with water. Hydration of C₃S and C₂S produces calcium silicate hydrate (CSH) and portlandite (calcium hydroxide or CH), which both are the main components in hydrated cement. The CSH is a semi-amorphous gel-like material that comprises ~70 wt% of the hydrated cement and is the primary binding material. The calcium hydroxide is crystalline and comprises roughly 15–20% of the hydrated cement. Moreover, the aluminum containing compounds (C₃A and C₄AF) react with gypsum to form ettringite (a calcium trisulfoaluminate hydrate Ca₆[Al(OH)₆]₂·(SO₄)₃·26H₂O) or monosulfoaluminate hydrate ((CaO)₃(Al₂O₃)(CaSO₄)·12H₂O) [10,12]. Previous studies have shown that, as an alkaline material, oilwell cement is highly vulnerable to acid attack from CO₂ fluids [10,11,13,14]. Generally, the carbonation of Portland cement causes an increase in porosity and permeability and a reduction in the mechanical strength of cement. The carbonation of Portland cement mainly includes the following chemical reactions [11,15,16]:



When CO₂ is dissolved in water (Eq. (1)), it forms carbonic acid (Eq. (2)) that reacts with hydration products in hydrated Portland cement such as CH and CSH (Eqs. (3) and (4)). The precipitation of carbonates can form a protective layer, inducing a decrease in permeability and porosity and then hindering the further penetration of carbonic acid into the cement [16–18]. As the CH and alkali phases are depleted, the pH of the pore solution falls, and bicarbon-

ate (HCO₃⁻) becomes the dominant species (Eq. (5)). This process eventually causes the calcium in the major cement phases to be dissolved out of the cement and only a porous silica gel remains [10,19].

Various laboratory experiments and models have been developed to understand the effects of temperature, pressure, pH, curing period, additives, water to cement ratio (w/c), and water salinity on the cement alteration phenomena and their rate under CCS conditions. In field working conditions, however, damage to materials is often due to many factors (chemical and mechanical) acting in a combined manner. The phenomenon in which a material is exposed to chemical corrosion while under simultaneous exposure to mechanical stresses is defined as stress-corrosion [20,21].

It has been reported that accelerated strength loss and spontaneous failure may occur in Portland cement mortar and concrete samples if they are simultaneously exposed to a chemical corrosion solution and to flexural/tensile stresses [21–27]. In a CCS well, besides chemical attack, mechanical degradation is another factor that can lead to cement sheath loss of integrity [28–31]. However, previous studies of well cement degradation under CCS conditions have focused mainly on individual chemical attack, ignoring the coupled function of CO₂ attack with mechanical stress. Thus, there is a need to study the coupled effects of CO₂ attack with mechanical stress on well cement, to increase understanding of the behavior of well cement under CCS conditions and provide more information for the safety assessment analysis of CCS wells.

Tensile failure is one of the main failure types of cement sheath for which is usually a hard-brittle material with low tensile strength. In this study, well cement samples were exposed to the action of CO₂ attack coupled with tensile stress. After exposure, the cement samples were characterized by optical microscopy, dye testing, relative tensile strength analysis, scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS) analysis, and X-ray diffraction (XRD).

2. Experiment

2.1. Materials

The materials used in this experiment were API (American Petroleum Institute) class G well cement (produced by the Jiajiang Cement Company, China), silica fume (produced by the Omax Company, China), AMPS-amide-carboxylic acid polymer based filtrate reducer G33S, and acetone-formaldehyde condensate based disperser USZ (produced by the Weihui Company, China). The chemical compositions of the class G well cement and silica fume are given in Table 1. The composition of the cement samples is shown in Table 2.

Table 1
Chemical composition of the class G well cement and silica fume analyzed by X-ray fluorescence (mass fraction, %).

| Material | SiO ₂ | CaO | Al ₂ O ₃ | Fe ₂ O ₃ | K ₂ O | MgO | Na ₂ O | SO ₃ | MnO | Loss on ignition |
|---------------------|------------------|-------|--------------------------------|--------------------------------|------------------|------|-------------------|-----------------|------|------------------|
| Class G well cement | 20.38 | 61.79 | 3.37 | 4.15 | 0.45 | 1.95 | 0.23 | 4.71 | 0.36 | 2.61 |
| Silica fume | 94.1 | 0.42 | 0.54 | 0.27 | 0.28 | 0.25 | 0.345 | 0 | 0 | 1.89 |

Table 2
Composition of cement sample.

| Materials and composition | | | | Water/solid ratio | Slurry density (g/cm ³) | Tensile strength (MPa) |
|---------------------------|-------------|-----------|-----------|-------------------|-------------------------------------|------------------------|
| Class G well cement | Silica fume | G33S | USZ | | | |
| 100% BWOC* | 10% BWOC | 0.6% BWOC | 0.1% BWOC | 0.44 | 1.90 | 2.89** |

* BWOC = by weight of cement.

** This value is based on the average of five samples and the standard deviation is 0.23 MPa.

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