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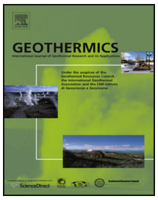

Experimental study of static and dynamic interactions between supercritical CO_2/water and Australian granites

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ABSTRACT

Recent research in Enhanced Geothermal Systems (EGS) have given rise to the interests of a CO_2-based EGS concept due to the unique thermo-physical properties of supercritical carbon dioxide (scCO_2) in EGS applications. However, available studies related to CO_2-based EGS are mostly theoretical investigations and relevant experimental study is highly scarce. To support the development of the new concept, this study conducts both static and dynamic fluid-rock interaction experiments between scCO_2/water and three different Australian granites. A tailored fluid-rock integration apparatus was designed to conduct the above investigation. The pulverised granites were exposed to scCO_2/water for up to 15 days at the simulated reservoir temperatures of 200 °C, 250 °C and pressures of 20 MPa and 35 MPa. The results of static fluid-rock interactions show that the elements of Na, Si, K, Ca, Mg, Fe, Al were found dissolved into the scCO_2-rich geofluid at an average rate of 4.5, 2.7, 1.6, 0.5, 0.3, 0.2, and 0.1 ppm/day, respectively. The dynamic fluid-rock interactions shows that the average rate of mineral dissolution in the pure water was around 183 ppm/day of Si, 14 ppm/day of Na, 12 ppm/day of Al, and 4 ppm/day of K, while only 0.4–2.5 ppm/day of Si, 0.4–1.6 ppm/day of Na, and 0.1–0.3 ppm/day of K for using scCO_2-rich stream as the geofluid. The typical composition of the trace elements dissolved in both pure water and scCO_2-rich geofluids were also identified. Fluid-rock equilibrium analyses shows that the geofluids obtained after the 15 days of static fluid-rock interaction may have reached/were approaching geochemical equilibrium for some elements (e.g. Si), whilst for the flow-through experiments the reacted geofluids were far from geochemical equilibrium. The examination of the fluid-rock interaction using the three Australian granites highlighted the importance of mineral composition to fluid-rock interaction. The research provides valuable experimental data and insights for understanding the CO_2-based EGS system.

1. Introduction

An urgent need for a worldwide shift from conventional fossil fuels to cleaner energy sources (particularly renewable energy) is continuously on the high-priority agenda of many governments/societies. This is driven by mainly the increasing pressure from fossil fuel usages on the earth climate which created a notorious “global warming” effect (Bernstein et al., 2007; Zhou et al., 2014). The work presented here is concerned with the advancement of understanding of an Enhanced Geothermal System (EGS) (Tester et al., 2006; Zhou et al., 2013), which uses supercritical CO_2 (scCO_2) and/or H_2O as its energy carriers to produce clean thermal/electrical energy. The primary focus of this article, however, is given to EGS reservoir engineering instead of power generation, and in particular, fluid-rock interactions at elevated temperatures and pressures.

Current and past EGS reservoir studies are mostly based on the use of water for hydraulic fracturing and as an energy medium (Baria et al., 1999; Brown et al., 1999; Brown, 2000; Hori et al., 1999; Kuriyagawa and Tenma, 1999; Parker, 1999; Richards et al., 1994; Sanjuan et al., 2006; Savage et al., 1992; Tenma et al., 2008; Tenma and Iwakiri, 1998). However, a major drawback of using water as the transportation fluid is that water, as a universal solvent, easily dissolves rock minerals and create scales in wells and surface facilities (Yanagisawa, 2007; Yanagisawa et al., 2006, 2007, 2008). Brown (2000) presented a novel EGS concept which utilised scCO_2

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as a geofluid (i.e. geothermal fluid). It is suggested that $scCO_2$ has a unique property which allows it to flow in/out of the reservoir with largely reduced pumping power, owing to its significant density difference between the hot and cold states — a phenomenon also called "thermosiphon". In addition, $scCO_2$ is a poor solvent for rock minerals eliminating or significantly reducing mineral precipitation on surface equipment.

More specifically, fluid-rock interaction and its impacts on the performance of EGS system have been investigated primarily for H_2O-based EGS. Literature on CO_2-rock interaction under conditions pertinent to EGS is scarce. Nevertheless, in Table 1 we summarised research work highly related to CO_2-rock interactions. For example, Lin et al. (2007, 2008) conducted the experiments of pure CO_2-rock interaction in a hydrothermal autoclave at 100 °C and 10 MPa. The investigation indicated minute amount of H_2O presented in the pores of quartz and granite rocks. Bertier et al. (2006) and Shiraki and Dunn (2000) conducted experiments in sandstone formation using CO_2 and revealed slight dissolution of ankerite/dolomite and aluminium silicates, which may help enhance the bed porosity/permeability. Kaszuba et al. (2003, 2005) carried out experiments in a flexible cell hydrothermal apparatus that is used to simulate saline aquifer storage at 200 °C and 200 bar injected with CO_2. They found that geochemical reactions took place followed by the precipitation of carbonates. Reichman et al. (2008), during their demonstration of a pilot CO_2-based EGS system in Australia, advises that the corrosive nature of CO_2-water mixture in the production wells requires corrosion resistant protection measures. Studies have also been conducted on CO_2 sequestration by mineral carbonation in geothermal fields (Kaieda et al., 2008, 2009; Ueda et al., 2005). This is achieved by saturating water with injected CO_2 and then capture and precipitate CO_2 at a deep depth in the form of carbonate minerals. These studies provided insights on the possible CO_2—H_2O-rock interactions at water-rich environments. However, in a $scCO_2$-based EGS system, $scCO_2$ is the dominant fluid and there will be a wide region where CO_2 with minute amount of H_2O could interact with the host rocks. In a more recent study, a simulation conducted by Pruess and Spycher (2010) suggests that in a fractured reservoir water dissolution into CO_2 will persist in the CO_2 stream for many years. Sugama et al. (2010a,b, 2011), on the other hand, explored wet carbonation of American rock minerals and their reaction by-products: bentonite, kaolinite, and soft-clays. They exposed those materials to $scCO_2$/water for 72–107 h at 200 °C, 250 °C and 17.23 MPa. Under the performed static fluid-rock interaction, the authors examined mineralogical changes of the rock minerals in great details and established sound mechanisms for wet carbonation of rock minerals.

The above literature review clearly shows that most of the research is related to theoretical and experimental works of CO_2 geosequestration at relatively low temperatures (Allen et al., 2005; André et al., 2007; Bachu and Adams, 2003). Studies on the fluid-rock interactions for $scCO_2$-based EGS at moderate-high temperatures and comparison with H_2O-based EGS are lacking. Moreover, Sugama et al. (2010a,b, 2011) only investigated fluid-rock interaction under static conditions and based on the mineral characteristics of granites from the United State. A dynamic/flow-through environment which may lead to more serious corrosion and mineral dissolution has not been investigated.

To support the development of CO_2-based EGS, in this study we examined the possible interactions between $scCO_2/H_2O$ and Australian granite rocks by exposing three different Australian granites (pulverised) to both pure water (for simulating H_2O-based EGS system) and CO_2–rich geofluids (for simulating $scCO_2$-based EGS system). The experiments were performed under conditions pertinent to a typical EGS reservoir for up to 360 h at 200–250 °C and 20 MPa/35 MPa. The static/batch experiments were performed to simulate the reaction of stagnant fluids with the granite bodies during the exploration, shutdown and maintenance periods of an EGS plant, whilst the dynamic/flow-through experiments were carried out to simulate the fluid flow in the reservoir from the injection to production well during the operating period of an EGS plant.

The experiments carried out in this study are to achieve the following three specific objectives: (i) identify possible fluid-rock reactions and reaction mechanisms for both simulated H_2O- and $scCO_2$-based EGS systems during the stagnant/operating phases; (ii) investigate the extent of mineral dissolution in the two geofluids during the stagnant/operating phases at varied reservoir temperatures and pressures; (iii) evaluate the effects of geofluid flow rate and granite mineral composition on the fluid-rock interaction. The ultimate goal is to advance the understanding of a $scCO_2$-based EGS system and verify the merits of $scCO_2$ over water.

2. Methodology

2.1. Granite samples

Granite rock samples were collected from three different places in Australia to represent different geological conditions of EGS reservoirs. They are respectively collected from the granite outcrop at Moonbi near the New England Highway in NSW, Australia and the drill-core samples from the intersected granite from Mossgiel 1 borehole (at a depth of ∼1795 m) and Nambucurra 1 borehole (at a depth of ∼261 m) at Murray-Darling Basin, NSW, Australia (see Fig. 1). The raw granite samples were cleaned, crushed and pulverised using tungsten carbide ring mill (N.V. Tema Ring Mill) for

(a)

(b)

(c)

Fig. 1. (a) Moonbi granite outcrop, New England Highway, NSW, Australia, (b) Mossgiel drilling core section at 1793–1796 m depth, (c) Nambucurra 1 drilling core section at 260.9–261.4 m.