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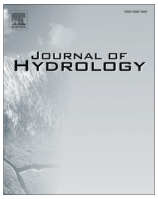

Research papers

CO_2 breakthrough pressure and permeability for unsaturated low-permeability sandstone of the Ordos Basin

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ABSTRACT

With rising threats from greenhouse gases, capture and injection of CO_2 into suitable underground formations is being considered as a method to reduce anthropogenic emissions of CO_2 to the atmosphere. As the injected CO_2 will remain in storage for hundreds of years, the safety of CO_2 geologic sequestration is a major concern. The low-permeability sandstone of the Ordos Basin in China is regarded as both caprock and reservoir rock, so understanding the breakthrough pressure and permeability of the rock is necessary. Because part of the pore volume experiences a non-wetting phase during the CO_2 injection and migration process, the rock may be in an unsaturated condition. And if accidental leakage occurs, CO_2 will migrate up into the unsaturated zone. In this study, breakthrough experiments were performed at various degrees of water saturation with five core samples of low-permeability sandstone obtained from the Ordos Basin. The experiments were conducted at 40 °C and pressures of >8 MPa to simulate the geological conditions for CO_2 sequestration. The results indicate that the degree of water saturation and the pore structure are the main factors affecting the rock breakthrough pressure and permeability, since the influence of calcite dissolution and clay mineral swelling during the saturation process is excluded. Increasing the average pore radius or most probable pore radius leads to a reduction in the breakthrough pressure and an increase by several orders of magnitude in $scCO_2$ effective permeability. In addition, the breakthrough pressure rises and the $scCO_2$ effective permeability decreases when the water saturation increases. However, when the average pore radius is greater than 0.151 μm, the degree of water saturation will has a little effect on the breakthrough pressure. On this foundation, if the most probable pore radius of the core sample reaches 1.760 μm, the breakthrough pressure will not be impacted by the increasing water saturation. We establish correlations between (1) the breakthrough pressure and average pore radius or most probable pore radius, (2) the breakthrough pressure and $scCO_2$ effective permeability, (3) the breakthrough pressure and water saturation, and (4) the $scCO_2$ effective permeability and water saturation. This study provides practical information for further studies of CO_2 sequestration as well as the caprock evaluation.

1. Introduction

There is increasing evidence that CO_2 emissions from industrial production and modern human activities contributes to global warming (Nordbotten et al., 2005; Falkowski et al., 2000; Cox et al., 2000). Geological storage of CO_2 has been widely recognized as an important method to mitigate greenhouse gas emissions (Juanes et al., 2006). The major storage options for CO_2 are deep saline aquifers, unminable coal beds, and depleted oil and gas reservoirs (Shukla et al., 2010). In all these sequestration methods, a major concern is potential CO_2 leakage into the atmosphere. Therefore, we need to expand our understanding of the sealing capacity of the caprock in geological sequestration sites. To ensure the long-term safety of an underground storage facility, the integrity of the caprock is imperative for safe storage of the injected CO_2 (Hildenbrand and Krooss, 2003; Song and Zhang, 2013). Generally, the reservoirs are deeper than 800 m, where the pressures and temperatures surpass the critical point values for CO_2 (7.38 MPa, 31.1 °C) and CO_2 is stored as a supercritical (sc) fluid. In these conditions, the density of CO_2 is lower than that of the formation water (Mickler et al., 2013); therefore, the CO_2 will move up towards the caprock by gravimetric segregation and accumulate beneath the caprock. Thus, during the CO_2 injection process, when the reservoir pressure becomes too high and exceeds the threshold pressure, it will induce mechanical stresses

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and deformation in and around the reservoir formation (Rutqvist et al., 2007). Irreversible mechanical changes may be caused by the induced stresses, which will create new fractures or reactivate the existing faults. This will generate new flow paths through the reservoir and a strong concentration of CO_2 under the nearby caprock, which may weaken the integrity of the caprock, and thereby the effectiveness of the sequestration. Moreover, if the caprock is discontinuous or missing, the breakthrough pressure of the target formation will be the most important factor regulating the process of CO_2 upward movement (Lee et al., 2010). The sealing quality of a rock is also affected by the CO_2 effective permeability after capillary leakage. It depends on the displacement pressure required to permeate CO_2 through the water wetting porous medium (Hildenbrand et al., 2002). Therefore the effective permeability is influenced by the degree of water saturation and the non-wetting phase of the CO_2 inside the pores (Wollenweber et al., 2011).

Gas breakthrough experiments can be used to measure the breakthrough pressure and the CO_2 effective permeability in a core flooding system (Thomas et al., 1968; Schowalter, 1979; Gallé, 2000; Hildenbrand et al., 2002; Hildenbrand and Krooss, 2003; Li et al., 2005; Boulin et al., 2013; Rezaeyan et al., 2015). Three gas breakthrough methods have been used in experimental studies: the step-by-step method, continuous injection method, and residual capillary pressure approach. The step-by-step approach, which is based on the high-accuracy definition of breakthrough pressure, is widely used (Thomas et al., 1968; Schowalter, 1979; Boulin et al., 2013). Almost all previous breakthrough pressure studies focused on a CO_2-saturated rock system; however, this is not always the case found in the field. Low-permeability sandstone rock may exist in an unsaturated condition during the CO_2 injection and migration process. In this condition, part of the pore volume is occupied by CO_2, and the breakthrough pressure and permeability will change considerably depending on the degree of water saturation. Furthermore, once accidental leakage occurs, CO_2 will migrate up into the unsaturated zone (Bickle et al., 2007; Oldenburg et al., 2009a; Lary et al., 2012; Teasdale et al., 2014; Yang et al., 2014). Recent studies on CO_2 transport in the unsaturated zone (Oldenburg and Unger, 2003; Oldenburg et al., 2009b) showed that the unsaturated zone could still attenuate CO_2 leakage (Jeffrey et al., 2016).

The breakthrough pressure in a pore throat can be measured based on the Young–Laplace equation,

$$P_{bt} = \frac{2\gamma_{w,CO_2}\cos\theta}{R},$$

where $\gamma_{w,CO2}$ is the water–CO_2 interfacial tension (IFT), θ is the contact angle and R is the characteristic pore radius in the rock (Watts, 1987; Tokunaga et al., 2013). The Young-Laplace equation shows that the breakthrough pressure is affected by the IFT, contact angle, and pore radius; thus, the breakthrough pressure is related to the pore structure and mineral components of the rock (Li et al., 2005).

In addition, previous studies investigated the permeability and gas breakthrough in compacted bentonite with 0–80% water saturation (Tanai et al., 2012), and in synthetic Ca-smectite clay with 70–100% water saturation (Kaneta, 1998; Gallé, 2000). These experimental studies showed that the degree of water saturation and clay swelling have a significant effect on the breakthrough pressure and gas permeability. Therefore, to better understand the pressure bearing capacity and CO_2 transport properties, CO_2 breakthrough experiments under various degrees of water saturation in low-permeability rocks with different lithology and pore structure are required.

It was suggested that Ordos Basin is the most suitable basin in China for CO_2 geological storage (Chang et al., 2014; Li et al., 2015; Tian et al., 2015; Xie et al., 2015; Wang et al., 2015). And the low-permeability sandstone of the Ordos Basin can be regarded both as caprock and reservoir rock, investigation of its breakthrough pressure and permeability is necessary for future geological sequestration projects. So in this study, we conducted breakthrough experiments on five low-permeability sandstone cores from Ordos Basin in China at various degrees of water saturation, with different pore structure, permeability, porosity, and mineralogy. We presented the results of our investigation about the effects of the water saturation, as well as the pore structure and mineral composition on the CO_2 breakthrough pressure and permeability of these five core samples. It turns out that the influence of calcite dissolution and clay mineral swelling can be ignored, suggesting the water saturation and pore structure to be the main factors that have impact on the breakthrough pressure and permeability. The results of this study provide us with useful information for further studies of CO_2 geological sequestration as well as its storage security.

2. Materials and methods

2.1. Rock cores

The Ordos Basin is the second largest sedimentary basin in China and has a huge potential for CO_2 geological sequestration (Chang et al., 2013; Li et al., 2015; Tian et al., 2015; Xie et al., 2015). The basin has multiple layers of sealing caprock and abundant storage formations, as demonstrated by its huge hydrocarbon resources (Li and Hou, 2005; Hu, 2009; Liu et al., 2014). The basin has a stable and simple geological structure which is characterized mainly by subsidence (Wang, 2002; Hou and Zhang, 2008), with no large faults, even after several crustal movements. The Ordos Basin is one of the most important energy supply areas in China; it has many energy-related industries, such as steelworks, refineries, and power plants, which emit millions of metric tons of CO_2 every year into the atmosphere (Chang et al., 2013, 2014). Thus, it will be possible to store CO_2 within Ordos Basin and do not need to transport it to other sites, which can reduce CO_2 emissions by minimizing transportation costs.

The Ordos Basin has been considered as a prospective potential for large-scale CCS deployment. At the Shenhua Group CCS site, multiple sandstone formations with relatively low permeability are used for CO_2 injection and storage (Li et al., 2016a; Li et al., 2016b; Zhang et al., 2016). Five sandstone cores of relatively low permeability were chosen for our experiments. They were extracted from monitoring well No. 2 from the Shenhua Group CCS Project, which is located in the northeastern part of the basin, near Ejin Horo Banner, Inner Mongolia. And they were obtained from Triassic and Permian sandstone formations, of the Zhifang Group, Heshanggou Group, Liujiagou Group, Shiqianfeng Group, and Shihezi Group (Fig. 1). The cores were extracted from depths of 1244.3 m, 1537.75 m, 1691.87 m, 1939.95 m, and 2167.82 m, and are referred to as samples #1, #2, #3, #4, and #5, respectively (Fig. 2).

The dimensional specifications (diameter and length) and properties (porosity and permeability) of the core samples are listed in Table 1. For consistency, the porosity and permeability were both determined with helium, as described in Section 2.3.1. The mineral content of the rock cores was determined by X-ray diffraction (XRD) analysis (Table 2). Moreover, conventional mercury injection capillary pressure (MICP) measurements were conducted on all the core samples to determine the specific pore size distributions, and the distribution curves are shown in Fig. 3.

2.2. Experimental apparatus

The equipment used for the experiments in this study are shown in Figs. 4 and 5. The system used to measure the