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Evolution of sealing efficiency for CO_2 geological storage due to mineral alteration within a hydrogeologically heterogeneous caprock

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In CO_2 geological storage (CGS) context, the evolution of the caprock sealing capacity has received increasing attention, particularly on a geological time span (thousands of years). At this time scale, geochemical reactions may enhance or weaken the caprock quality. It is widely recognized that, for the reservoir, geological heterogeneities affect the concentration and spatial distribution of CO_2, and then affect the extent of gas–water–rock interactions, which in turn alters the hydrogeological properties of the reservoir. However, much less attention of these effects has been paid to the caprock. In this study, we presented and applied a novel approach to evaluate the effects of permeability and porosity heterogeneities on the alteration of minerals, the associated evolution of the caprock sealing efficiency and the containment of supercritical CO_2 ($scCO_2$) within the caprock. Even though this is a generic study, several conditions and parameters such as pressure, permeability, and mineral composition, were extracted from a caprock layer of the Shiqianfeng Formation in the Ordos Basin demonstration site in China. For the sake of simplification, a 2-dimensional model was designed to represent the caprock domain. We firstly generated an appropriate heterogeneous random field of permeability with the average permeability taken from the uppermost mudstone layer of the Shiqianfeng Formation, and then the heterogeneity in porosity was incorporated using a joint normal distribution method based on the available data. Homogeneous mineral compositions of the reservoir and caprock were used in all simulations. Simulations of three cases were performed, including a homogeneous case, a case with only permeability heterogeneity and a case with both permeability and porosity heterogeneities. The results demonstrate dramatic influences of permeability and porosity heterogeneities on the migration of $scCO_2$ within the caprock, the alteration of minerals, and therefore the evolution of the caprock sealing quality. Specific to the data used in this study, hydrogeological heterogeneities facilitated the overall penetration of $scCO_2$ within the caprock and promoted the alteration of minerals, thereby weakening the caprock sealing efficiency over the simulation time.

1. Introduction

It is widely recognized that CO_2 geological storage (CGS) is an important option for mitigating anthropogenic emissions of greenhouse gases (Bachu and Adams, 2003), therefore combating global climate change (IPCC, 2007). Among the geological storage options, in terms of worldwide storage capacity, CO_2 storage in saline aquifers appears to be the most promising option (Thibeau et al., 2007).

To guarantee CO_2 geological storage to be a viable carbon management solution, one of the main issues to be addressed is the potential hazard, including damage to the caprock, CO_2 leakage, groundwater quality reduction due to acidic fluids intrusion, etc. (Armitage et al., 2013). A competent caprock with low porosity is vital to a successful CGS project, as the caprock is the first natural barrier. It needs to be at an adequate depth in order to maintain CO_2 in a supercritical state ($scCO_2$) and prevent leakage of CO_2 for an extended period of time (thousands of years) (Fleury et al., 2010), without significant compromise on sealing efficiency (Haszeldine et al., 2005; Lu et al., 2009; Rubert et al., 2009). However, hydrogeological parameters (e.g., permeability and porosity) naturally have spatial variations within the subsurface (Gelhar, 1993; Schnaar and Digiulio, 2009). It is well known that heterogeneity in natural porous formations controls groundwater flow and solute transport.

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Therefore, a comprehensive understanding of heterogeneity within a formation is necessary to predict solute transport through tracer tests and transport experiments (e.g., Mackay et al., 1986; Guven et al., 1992; Phanikumar et al., 2005; Salamon et al., 2007).

Heterogeneities in the hydrogeological parameters have been reported to profoundly affect the CO_2 storage capacity (Deng et al., 2012; Issautier et al., 2014; Ashraf, 2014; Meckel et al., 2015; Gershenzon et al., 2014), distribution and migration (Sundal et al., 2013; Ott et al., 2015; Jahangiri and Zhang, 2011; Gasda et al., 2013; Gershenzon et al., 2014), and injectivity (Jin et al., 2014; Gasda et al., 2013; Issautier et al., 2013; Deng et al., 2012).

Most of these works focused mainly on the target layer for injection, i.e. the reservoir. However, less attention has been paid to the caprock which is crucial to the safety of CGS. Only recently, a limited number of reports concerning the effects of heterogeneity on leakage of brine and CO_2 through the caprock have been published (e.g., Deng et al., 2012; Liu and Zhang, 2011; McLaughlin et al., 2014). These studies indicate that the heterogeneities in hydrogeological parameters (e.g., permeability and porosity) dramatically affect the migration and leakage of CO_2 within the caprock. Surprisingly, in these works an important facet of reactive transport process has often been disregarded, either due to a lack of data or to increasing computational efficiency.

After CO_2 injection, the mobility of the CO_2-rich multiple fluid depends on the distribution of porosity and permeability, capillary pressure (Leverett, 1941), relative permeability (Krevor et al., 2012), mineral composition (Tian et al., 2014a) etc. The concentration and spatial variation of CO_2 will affect the distribution of pH and the extent of gas–water–rock interactions (Hermanson and Kirste, 2013; Tian et al., 2014b). Dissolution of calcite and aluminosilicates, such as chlorite and feldspar, occurs in areas of low pH, results in a release of cations and pH buffering.

In addition, changes in hydrogeological properties as a response to the stated geochemical reactions resulting from hydrogeological heterogeneities have been explored. For example, De Lucia (2008) performed series of experiments with different reaction paths. It was found that, specific to the data used in the test cases, high local permeability enhances the local flow increasing the reaction, so that porosity decreases which in turn decreases the permeability. Any developing fingering triggers a local decrease in the flow velocity, which prevents fingering from further developing. Higgs et al. (2013) evaluated the response of reservoir heterogeneity to the introduction of high pressure CO_2 specific to the Kapuni Field in New Zealand. The results suggested that CO_2 circulation changes in sweet zones (regions with relatively higher permeability and porosity) and relatively tight zones due to varying degrees of grain sizes, which affects the net mineral alteration. This results in various degrees of compartmentalization through development of "sweet spots" and tight zones.

Supercritical CO_2 may infiltrate into the initially water-saturated caprock once the CO_2 capillary entry pressure is exceeded due to buoyancy differences between the CO_2 and in-situ fluids and the injection pressure. The invasion of $scCO_2$ may acidify the resident brine to a pH of about 3–5 (Liu et al., 2011). This low pH brine may invade the pore space and induce interactions with the minerals. For a long-term period, the acidified brine (even buffered by carbonate dissolution) may trigger reactions with the alumino-silicate minerals originally present in the caprock. Depending upon varying geochemical settings, these reactions may hinder or facilitate the rates of individual reaction, potentially contributing significantly to the evolution of permeability/porosity, thereby enhancing or weakening the sealing efficiency of caprock (Alemu et al., 2011). Therefore, the resistance of the caprock to geochemical processes is a site-specific phenomenon, and on a geological time scale, it is imperative to incorporate the effects of the heterogeneity of a geologic setting and its effects on the sealing efficiency of the caprock.

Several studies (e.g., Chen et al., 2013; Farajzadeh et al., 2011; Kong and Saar, 2013)involving geological heterogeneity considered the porosity as a constant variable based on a common assumption that the physical heterogeneity of aquifers needed to explain groundwater flow and transport is manifested entirely in the hydraulic conductivity field, and that variations in porosity have negligible effects except as a contributor to the hydraulic conductivity heterogeneity. However, Hu et al. (2009) suggested that it is also important to characterize and incorporate the porosity heterogeneity in transport modeling, because it could enhance irregular plume distributions, and delay and spread solute breakthrough curves. Therefore, the porosity heterogeneity should be incorporated into the geochemical simulations of CO_2-brine-caprock interactions.

It is a goal of the Chinese government and worldwide to reduce the total greenhouse gas emissions. The first, fully functional CGS demonstration pilot in China by the China Shenhua Group has recently been implemented. Precisely assessing the evolution of the caprock sealing efficiency with the geochemical processes coupled under condition of hydrogeological heterogeneity is imperative to assess the safety of a CGS project over the span of thousands of years. In this study, reactive transport simulations were performed in order to account for the effects of spatial permeability and porosity variations on the migration of CO_2 within the caprock, mineral alterations, and the evolution of caprock sealing efficiency. In addition, we also evaluated the relative impacts of incorporated heterogeneity in porosity. Using currently available data (including permeability, porosity and mineral composition) taken from the Shiqianfeng Formation in the Ordos Basin, we firstly plotted the heterogeneous distribution of permeability in a 2-D model that represents the conceptual model of caprock domain by employing a lognormal pattern. After the generation of heterogeneous permeability, the joint normal distribution theory was introduced to obtain the spatial porosity variation. By comparing the results of the simulations, the effects of heterogeneities on the above-mentioned concerns were concluded.

2. Geologic characteristics of the Ordos Basin, research location

The Ordos Basin is the second largest sedimentary basin in China, consisting of minor tectonic deformation, a multi-cycle structural evolution, and varied sedimentary facies. It is located in the western part of North China (Luo et al., 2014), and covers an area of $25 \times 10^4\ km^2$. The Ordos Basin spans the western part of the Lvliang Mountains, and is east of the Helan Mountains, north of the Qinling Mountains and south of the Yinshan Mountains. The basin is divided into six structural units: the Yimeng Uplift, Yishan Slope, Tianhuan Depression, western Fault-fold Belts, Weibei Uplift, and Jinxi Fold Belt (Fig. 1). The first fully functioning CO_2 capture and geological storage project in China will inject 10 Mt/a CO_2 into the saline reservoirs. The demonstration site is situated in the eastern section of the northern Yishan Slope (Fig. 1). The target zone is in the interior stable region of the basin, where a relatively complete sedimentary formation (Shiqianfeng Formation) from top to bottom is located. Lithological trapping is the main capture type expected for CO_2 immobilization within in the Ordos Basin saline reservoir Ordos Basin. Each formation in the target zone has different geological conditions in order to develop a layered lithological trap. According to Li et al. (2013), the formations below 1000 m are divided into six reservoir-caprock combinations. The Shiqianfeng Formation of the Upper Permian has the largest storage capability, consisting of multilayered sandstone-mudstone