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# Experimental study of $CO_2$–brine–rock interaction during $CO_2$ sequestration in deep coal seams

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ABSTRACT

$CO_2$ sequestration in deep coal seams is a potential option for reducing greenhouse gas emissions. Once $CO_2$ is injected into coal seams, sealing capability of the cap rock is critical. To investigate and quantify reactions over time between $CO_2$, cap rocks and brine, associated with selected cap rocks of the No. 3 coalbed of the Qinshui Basin in China, batch experiments were conducted for reacting powdered rock samples (180–220 μm) with $CO_2$ and brine, as well as $CO_2$-free brine, at 160 °C and 15 MPa. The analysis of leachate chemistry indicated significant mobilization of major elements from dissolution of carbonate and silicate minerals in the coal measure strata. Analysis of reacted solids by XRD and SEM also revealed appreciable changes in mineralogical compositions. For lithic sandstone after reaction with $CO_2$–brine, the contents of quartz, plagioclase, illite and chlorite increased considerably, whereas the contents of illite/smectite, biotite and kaolinite decreased more or less. The calcareous mudstone reacting with $CO_2$–brine and $CO_2$-free brine all showed major mineralogical alteration after 12 days of treatment. The modeling results identified key chemical processes, but they also showed that the models are not capable of covering all possible contingencies. The precipitation of carbonate minerals could also enhance the security of $CO_2$ sequestration in deep coal seams.



## 1. Introduction

Climate change has become a hot point involved in global environmental issues. Controlling greenhouse gas emissions and protecting global climate is a major issue at present. Of all types of greenhouse gases blamed for climate change, emissions of $CO_2$ from human activities has one of the largest impacts on climate change gases, accounting for 63% of the temperature effect from all total greenhouse gases. Additionally, $CO_2$ is retained in the atmosphere for more than 200 years (Metz et al., 2005). The question of how to reduce the content of $CO_2$ in the atmosphere is the current problem to be solved. It is well known that geological $CO_2$ storage (GCS) is undoubtedly the most realistic and effective disposal method. At present, storing $CO_2$ by injection into subsurface oil and gas reservoirs (Winter and Bergman, 1993; Bachu and Shaw, 2003), deep saline aquifers (Xu et al., 2004; W. Zhang et al., 2009; X. Zhang et al., 2009; Lei et al., 2015; Tian et al., 2015) and deep unmineable coal beds (Massarotto et al., 2010; Dawson et al., 2011) is the most valid and economic choice for reducing $CO_2$ emissions into the atmosphere. Of these methods, coal bed geological disposal is one of the most favorable, as enhanced coal bed methane recovery can be accomplished simultaneously with sequestration of $CO_2$. This technique is known as $CO_2$-ECBM (Gale and Freund, 2001; White et al., 2005; Czerw, 2011; Baran et al., 2015).

Throughout the world, coal measure strata are important source layers of oil and gas, and they are important reservoirs, especially for natural gas resources. By the statistics, many large gas fields in the world are associated with coal measure strata. Coal measure strata include a large number of abandoned coal seams due to technical and economic reasons, and these unmineable coal seams are potential geological structures that can be used for geological $CO_2$ sequestration. Only a handful of $CO_2$-ECBM tests in the world have been implemented. These tests showed great potential for both $CO_2$ sequestration and ECBM production. For instance, the United States built the first $CO_2$-ECBM pilot project in the San Juan basin, and more than 100,000 t of $CO_2$ has been injected into the Fruitland coal seam since 1996 (Weber et al., 2012). Canada is in the process of a controlled trial of a $CO_2$-ECBM test in Alberta (Gentzis, 2000), and the evaluation of regional projects is widely being carried out throughout the world. In addition, Australia, Japan, The Netherlands and other countries have implemented $CO_2$ injection to enhance CBM exploitation, and they have carried out related researches (Li et al., 2004; Golding et al., 2013).

There are many coal-bearing rock series widely distributed in continental sedimentary basins (Qinshui Basin and Ordos Basin, etc.) in China, which are more suitable for $CO_2$ sequestration. Since 2002, under the support of the governments of Canada and China, the China United Coal Bed Methane Corporation carried out the theory and technological research of $CO_2$ injection/burial, enhanced coal bed methane

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recovery, and successful implementation of a single well $CO_2$ injection test in the southern Qinshui Basin of Shanxi Province in 2004, obtaining a series of key parameters (Wong et al., 2007, 2010; Wei et al., 2007). Under the support of the China Ministry of Science and Technology, the China United Coal Bed Methane Corporation carried out a technological research on deep coal seam $CO_2$ injection and coal bed methane exploitation in 2007, to achieve the commercialization of this technology. This is the first research project of deep coal bed $CO_2$-ECBM in China, and 234 t of $CO_2$ was injected into the No. 3 coal seam.

In recent years, in allusion to the basic theory of $CO_2$-ECBM, a series of studies on multicomponent gas competitive adsorption on coal surfaces (Tang et al., 2004; Zhang et al., 2004; Zhou et al., 2013), coal matrix expansion and contraction induced by adsorption (Karacan, 2003; Qin et al., 2005; Pan and Luke, 2007), and numerical simulation (Sun, 2005; Wu and Zhang, 2007; Siriwardane et al., 2012; Vishal et al., 2013) were carried out. However, long-term studies have ignored the fact that, once $CO_2$ is injected into coal seams, it will dissolve in formation water and form carbonic acid. Thus, the minerals in coal seams and roof-floor rocks were dissolving and the components of coal were changing. Secondary aluminosilicate and carbonate minerals could be formed at the same time, so that $CO_2$ can be safely sealed underground for a long time (Li et al., 2013).

To reveal the physicochemical processes and evaluate the security of $CO_2$ sequestration in deep unmineable coal seams, it is necessary to carry out the related experimental investigations on $CO_2$–brine–rock reactions, to discuss the geochemical behavior and possible mechanism of mineral traps in the long-term interaction between $CO_2$ and coal measure strata, and to provide a natural analogy as a research object and as a geological basis for $CO_2$-ECBM projects.

## 2. Geological setting

The Qinshui Basin is a near longitudinal-trending tectonic basin between Taihang Mountain and Lvliang Mountain, located in the middle of the North China Platform. The basin has experienced the tectonic movements of the Indosinian Period (Late Permian to Triassic), the Yanshan Period (Jurassic to Early Cretaceous) and the Himalayan Period (Late Tertiary). The basin's fold and fault structures are well developed, and most of the tectonic lines are in the NE-NNE direction. The study area, in the north block of Shizhuang, is located in the southeast of the Qinshui Basin. The main geological structure is folding. Faulting is not well developed, and existing faults are no longer than 100 m. Collapse columns occur occasionally. The overall structure of the study area is a western-leaning monocline (Fig. 1). The block is divided by the longitudinal-trending syncline structure into three tectonic units. The east unit is a gentle monoclinal structure, with only a few small faults and with dip angles generally less than 5°, and it slowly uplifts towards the SE. The central unit is a severely squeezed anticline-syncline pair with a range of 210 m, and the axis orientation is NNE. The west unit is a complex groove structure, with several large faults, and the coal seam has a discernible undulation (Huang et al., 2010; Ye et al., 2012).

The Upper Carboniferous Taiyuan Formation and the Lower Permian Shanxi Formation are the main coal-bearing strata in the study area, containing a total of 6–11 coal seams. Among them, the thicknesses of the No. 3 coal seam of the Shanxi Group and the No. 15 coal seam of the Taiyuan Group are larger and have a stable spatial distribution. They are the main coal seams for coal bed methane exploration. The burial depth of the No. 3 coal seam is from 830 m to more than 1600 m due to dipping strata, and the thickness is 4–6 m with an

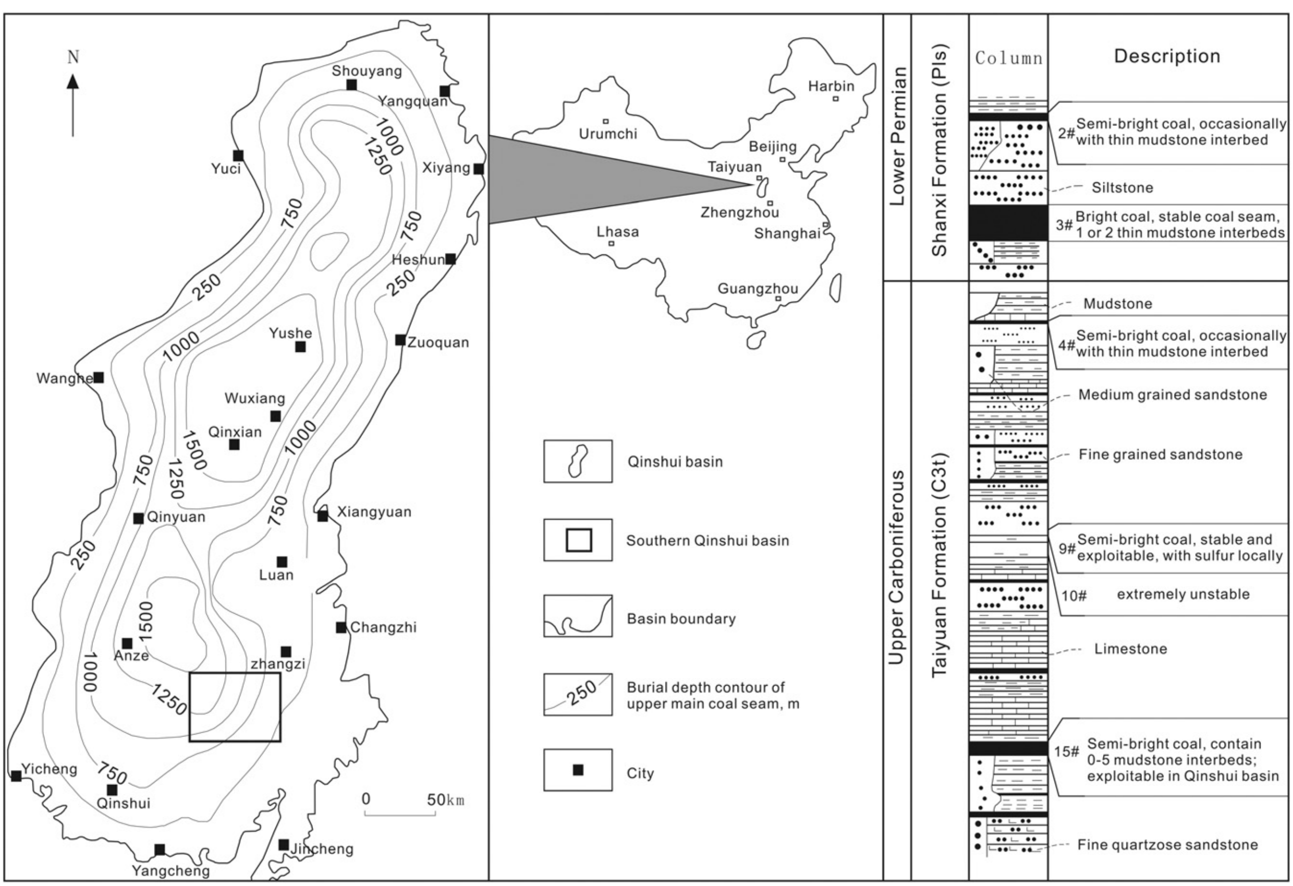


**Fig. 1.** Burial depth contour map of the No. 3 coal seam of the Qinshui Basin and the stratigraphic section of coal-bearing strata.
Modified from Wei et al. (2007) and Cai et al. (2015).