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Research paper

Water-rock interaction and methanogenesis in formation water in the southeast Huaibei coalfield, China

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ABSTRACT

The hydrogeochemical characteristics of aquifers in coal-bearing strata can provide an important foundation for understanding the evolution of the aquifers and recognizing the formation process and occurrence of biogenic gas. By analyzing the hydrogeochemical characteristics of water produced from the Cenozoic, the limestone, and the coal strata aquifers and the coalbed gas wells, this study focused on water-rock interactions and methanogenesis in the formation water of the Suzhou mining area, Huaibei coalfield, China. Tectonically, this is one of the most complex coalfields in China. The dissolution of evaporites, silicates, and carbonates controlled the chemical composition of the formation water. The Cenozoic aquifers have a relatively stable water quality type and balanced ionic composition, but more complex water-rock interactions occur in the coal strata and the limestone aquifers. Extensive methanogenic activity and methanogenesis could be recognized by the meteoric water recharge, positive $\delta^{13}C$ signatures for dissolved inorganic carbon (DIC; 15.4‰–26.2‰), lower sulfate concentrations (<2 mM), and higher levels of dissolved methane and DIC. The generation and enrichment of secondary biogenic gas in the coal seams was after the microbes' inoculation along with the freshwater infilling the fracture network.

1. Introduction

The prospective resource of coal-bed methane (CBM) in the Huaibei coalfield could reach 315.9 billion cubic meters (Jiang et al., 2010). As one of the CBM exploitation demonstration sites in China, the exploitation in the southeast of the coalfield has just started. Spatially, thermogenic gas, mixture gas, secondary biogenic gas, and a gas-weathered zone were distributed regularly from the bottom to the top and from the center to the margin of the cola-bearing sedimentary basin (Bao et al., 2014; Li et al., 2015). Considering that the hydrocarbon index ($C_{HC} = C_1/(C_2 + C_3)$) is much higher than 1000 and the $\delta^{13}C_{CH4}$ are partly lighter than -60‰, the study area should be very rich in secondary biogenic gas (Li et al., 2015).

Previous studies have shown that most of the microbial CBM in coal seams formed during later periods of coalification and developed in relationship to the injection of ancient meteoric water (McIntosh et al., 2002; Healy et al., 2011; Shuai et al., 2013; Pashin et al., 2014). Hence, researchers have paid greater attention to the generation mechanisms of microbial methane in ground water and their controlling factors (Scott et al., 1994; Aravena et al., 2003; Flores et al., 2008; Gao et al., 2013; Ju et al., 2014). To adequately understand the microbial activity and the formation process of microbial gas, it is necessary to analyze the chemical and isotopic compositions of related solutes in aquifers and the redox state of the water (Whiticar et al., 1986; Whiticar, 1999; Schlegel et al., 2011; Golding et al., 2013).

Moreover, the hydrogeochemical characteristics of the formation water may provide useful information about its evolutionary process, such as the degree of the water-rock interactions and the sources of the solutes (Négrel et al., 1993; Gaillardet et al., 1999; Yang et al., 2008; Li et al., 2014). Additionally, the water produced

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during CBM exploitation may induce potential threats to surface water, soils, and ecosystems because of the higher salinity and heavy metal concentrations (Cheung et al., 2010; Healy et al., 2011; Yang et al., 2011). Thus, it is essential to research the geochemical features of the produced water and evaluate its influence on the environment surrounding the gas wells.

Based on the hydrogeological features of the study area and the long-term groundwater quality data, this study analyzed the associated chemical and isotopic compositions of the formation water and the produced water from CBM gas wells through systematic sampling. The aims are to (1) research the water-rock interactions and evolution process of the formation water and (2) evaluate the microbial activity and the formation pathway of microbial methane and its enrichment features.

2. Geological background

The Huaibei coalfield lies in northern Anhui Province (E: 115°58′–117°12′, N: 33°20′–34°28′), and its coal-bearing area is approximately 4100 km^2 (Jiang et al., 2010). Tectonically, this coalfield lies on the southeastern margin of the ancient North China Plate. Affected by the Dabie-Tanlu-Sulu orogenic evolution, the structural styles of the coalfield include linear tight folds and imbricate thrust faults (Wu et al., 2011). The main coal-bearing structures in the study area are the Sudong and the Sunan synclines (Fig. 1).

As one of the most geologically complex coalfields in China, the origin, occurrence, and distribution of CBM in the Huaibei coalfield differ in many ways from those coalfields with simple tectonic evolutions. Based on its tectonic-thermal-burial history, the process of CBM generation in the Huaibei coalfield occurred in three stages: (1) thermogenic gas generation, when the buried depth of coal seams increased rapidly; (2) gas escape, when the coal-bearing strata moved upward; and (3) the generation of secondary biogenic gas at a suitable temperature and burial depth (Wu et al., 2011).

2.1. Coal-bearing strata and gas content

The minable coal seams in this mining area mostly lie in the Permian strata (Table 1). The total thickness of the coal-bearing strata exceeds 1300 m, and the largest thickness for a single coal seam is 21 m (Zheng et al., 2008). According to Wu and Li (2005), the main coal seams have a relatively high gas content (6.9–25.5 m^3/t) and gas-bearing saturation (98%–100%).

2.2. Hydrogeology

The coal seams in the study area are all covered by unconsolidated Tertiary and Quaternary sediments. According to the lithology and the distribution of the aquifers, the formation water can be classified into three types (Table 1): the upper aquifers in the Cenozoic unconsolidated formations, the middle aquifers in the Permian coal strata, and the lower aquifers in the Carboniferous limestone strata (Gui, 2005; Chen et al., 2013a, 2013b).

2.2.1. Cenozoic aquifers

From north to south and east to west, the thickness of the Cenozoic sediments increases gradually and reaches hundreds of meters in the study area (Gui, 2005). Sand layers, gravel layers, and interlayers of claypan are the main components of these formations, and the hydraulic connections among the four aquifers in the Cenozoic formations are negligible because of the stable distribution and large thickness of the water-resistant interlayers.

The fifth aquifer, which lies in a Jurassic conglomerate layer, has a very high water-bearing capacity. However, this aquifer is controlled by paleotopography and exists only in shallow and marginal areas in the study area. Hence, the Cenozoic unconsolidated formations often directly overlie the coal-bearing strata.

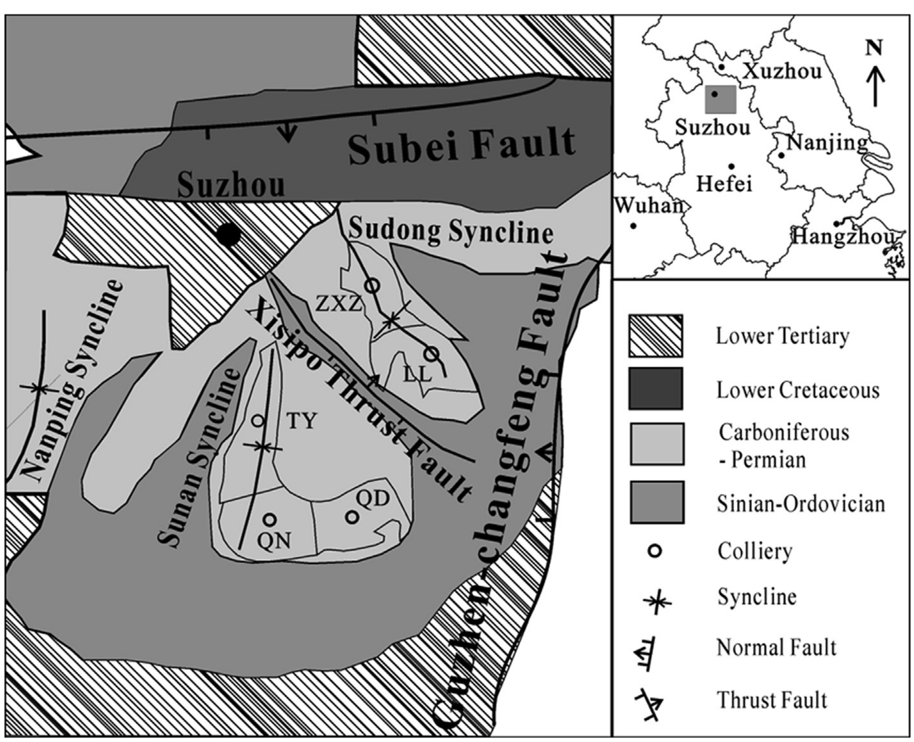

Fig. 1. Mining location and the main structure outline map (QD: Qidong colliery; QN: Qinan colliery; TY: Taoyuan colliery; ZXZ: Zhuxianzhuang colliery; LL: Lulling colliery).