



Full Length Article

Experimental investigation of CO₂ injection into coal seam reservoir at in-situ stress conditions for enhanced coalbed methane recovery



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ABSTRACT

CO₂ geological sequestration is an effective method to reduce the concentration of CO₂ in the atmosphere. The injection of CO₂ into CH₄-containing coal seams also named CO₂-ECBM (CO₂ Enhanced Coalbed Methane Recovery), allows the storage of CO₂ and the enhancement of CH₄ recovery. Due to complex geological environment of the site, dynamic variation of CH₄ and CO₂ can hardly be monitored in real time during CO₂ injection into coal seams. Therefore, this paper conducted experimental studies were carried out on cuboid coal samples with a size of 300 × 70 × 70 mm via a self-developed experimental device which can simulate in-situ stress and temperature conditions. In the experiment, different adsorbed gases (CH₄ and CO₂) were applied to determine the variation of permeability values under various load stresses and pore pressures. Results proved that the coal had greater adsorption capacity to CO₂ than to CH₄. The injected CO₂ occupied the original adsorption site of CH₄, thus leading to the rapid discharge of CH₄. Meanwhile, real-time dynamic monitoring was also performed on gas parameters in coal under different gas injection pressures to obtain variation laws of pore pressure, gas flow rate and concentration at the outlet with the increment of injected CO₂. The results reveal that with the increase of injected CO₂, pore pressure tended to equilibrium while CH₄ flow rate and concentration at the outlet gradually fall to zero. Therefore, CO₂ injection into the coal mass can effectively improve the recovery of CH₄. In addition, variations of gas flow rate and concentration at the outlet with the displacement ratio of gas volume under different gas injection pressures suggested that the enhancement of gas injection pressure boosted discharge efficiency of CH₄ from the coal mass and CO₂ consumption. Furthermore, inspired by the competition relationship between engineering efficiency and CO₂ consumption, we think a reasonable and effective economic cost model can be established as an effective method to guide the selection of gas injection pressure in the future. Hence, the experimental results have guiding significance for better understanding and application of CO₂-ECBM technology.

1. Introduction

Carbon dioxide is both an important raw material for plant photosynthesis that can elevate photosynthetic products, and a kind of greenhouse gas affecting heat balance of the earth [1,2]. With the heavy consumption of fossil energy after Industrial Revolution, the concentration of CO₂ in the atmosphere has increased year by year. At the current growth rate, the temperature of the earth is to witness an increase of 1.5–4.5 °C by 2030, which will exert a serious impact on global environment. Therefore, the reduction of CO₂ concentration in the atmosphere has become a hot research issue [3,4].

Geological sequestration and storage of CO₂, an effective measure of

reducing greenhouse gases, has drawn the attention of scholars and organizations all around the world [5–7]. Based on the estimation of the world's underground storage capacity of CO₂ by Bruant et al., about 7.0 × 10⁷ km² of sedimentary basin can be applied to storing 1.0 × 10¹⁴–2.0 × 10¹⁴ t of CO₂ which is equivalent to the CO₂ emissions by mankind over thousands of years [8]. At present, CO₂ storage sites mainly cover unmineable coal seams, depleted hydrocarbon reservoirs, deep aquifers and marine strata [9]. However, in fact, storing CO₂ into a CH₄-containing coal seam can bring lots of benefits such as promoting the CH₄ recovery and reducing gas disasters [10–13]. It offsets the cost of capture, compression, transportation, and storage of CO₂ by producing natural gas. Besides, unmineable coal seams are also

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available for storage because of their wide distribution [14,15]. This technology is called CO₂ enhanced coalbed methane recovery (CO₂-ECBM) [16]. During the CO₂-ECBM process, CO₂ first enters the fracture system because the injection pressure is greater than coal reservoir pressure. When CO₂ is entering the coal matrix, it will sorb to the coal inner surface, replacing part of the CH₄ by a combined sorption/diffusion process, due to the adsorption capacity of CO₂ exceeding that of CH₄. Therefore, the understanding of dynamic variation of CH₄ and CO₂ in the coal is of great significance to improve this technology during the CO₂ injection into coal seams with CH₄.

Carbon dioxide-Enhanced Coalbed Methane Recovery was first proposed by Macdonald of Alberta Energy during discussions with Gunter and coworkers in 1991 [3]. To date, many scholars have proved the feasibility of injecting CO₂ into a CH₄-containing coal seam which has a promising future through various experimental methods. Clarkson et al. [17] reported during CH₄ recovery that the injected CO₂ could depressurize CH₄ in the free gas or occupied adsorption space to accelerate CH₄ desorption from the coal seam, thus boosting the production of CH₄. Stevens et al. [18], who specialized in the process of enhancing CH₄ recovery by injecting CO₂, considered that the CO₂ injected was conducive to maintaining the pore pressure and guaranteeing the cleat and pore channels unobstructed for smooth CH₄ emission. Zuber et al. [19] also verified that CO₂ injection into coal seams contributed to the improvement of CH₄ production.

To further verify that CO₂ injection into the coal mass can enhance the emission rate of CH₄, some scholars have conducted physical simulation experiments on cylindrical coal samples, making detailed record of the CH₄ recovery volume after CO₂ injection. Fulton et al. [20] injected CO₂ into dry and water-saturated CH₄-containing coal samples under low pressures. In these experiments, cylindrical coal samples used were 8.89 cm or 9.555 cm in diameter and 5.08 cm or 10.16 cm in length; the gas inlet and outlet were on the same side of the coal; and the CO₂ injection pressure was 45–200 psia. Experimental results revealed that the CH₄ recovery after CO₂ injection was 9%–57% higher than that of natural emission. Reznik et al. [21] conducted the CO₂ injection test on coal samples under high pressures following Fulton's experiment. The CH₄ emission rate was about 30%, but CH₄ in the coal was totally discharged under gas injection pressure of 200–800 psia. Besides, in their another experiment to inject CO₂ and N₂ mixture into samples, CH₄ recovery plunged. Jessen K et al. [22] carried out different experiments with 100% N₂, 100% CO₂, 85% CO₂/15% N₂, 46% CO₂/54% N₂, and 24% CO₂/76% N₂, respectively. The cylindrical coal sample with a size of 250 mm in length and 42.5 mm in diameter is made of 0.25 mm coal particles whose permeability reached $3.1 \times 10^{-14} \text{ m}^2$. In addition, they also studied the effects of gases of different compositions on CH₄ recovery and breakthrough time, reporting that the injected gases could all achieve CH₄ recovery rate of over 94%, but the recovery rate would be lowered if the gas was mixed with N₂ (even with a trace of N₂). Furthermore, compared with pure CO₂, the gas mixed with N₂ injected into coal led to longer breakthrough time of CH₄ emission and less total injected volume. Dutka Barbara et al. [23,24] conducted CO₂-ECBM experiments on coal samples with a diameter of 96 mm and a length of 280 mm which consists of particles with a size of below 0.2 mm. They analyzed variation laws of pore pressure as well as CO₂ and CH₄ concentrations during CO₂ injection, concluding that CO₂ injection could improve CH₄ emission amount effectively.

Following physical simulation experiments that obtained favorable conclusions in the laboratory, field tests of CO₂-ECBM technology were also gradually carried out, and America took the lead. In 1996, the Allison Unit pilot, located in the Northern New Mexico part of the San Juan Basin, was selected as the first demonstration site for field test. After the implementation of CO₂-ECBM technology, CBM production was greatly improved and CH₄ recovery reached 77–95%, lasting for over 6 years [25]. To test and monitor the injectivity, capacity, heterogeneity, and performance of mature coalbed methane reservoirs, a

CO₂-ECBM field injection project was performed in Black Warrior field in addition to other similar programs in North America [26–29]. Besides, other countries and regional organizations followed America in conducting field practices. In Canada, pure CO₂, pure N₂, flue gas, and CO₂-rich flue gas were injected into the experimental area in Fenn Big Vaney of the Alberta Basin. Results proved that CBM recovery was enhanced by injection of other gases. Besides, it was also observed that coal permeability was lowered and improved by injecting pure CO₂ and pure N₂ or N₂-containing flue gas, respectively [30]. To assess the technical and economic feasibility of CO₂ sequestration in coal seams, the Recopol project was implemented in the EU where a total of 760 tons of CO₂ was injected in Upper Silesian Basin of Poland [31]. In Japan, field test was conducted in the Ishikari Basin of Hokkai-do, where dynamic changes of the coal seams were monitored in real time via measures like field monitoring. Results suggested that there was an upsurge in CH₄ recovery after CO₂ injection [32]. A field research in Qinshui Basin, Shanxi Province, China, indicated an improved CBM recovery and the good effect of CO₂ sequestration after injecting CO₂ into coal seams [33].

Numerous experimental studies have been conducted on CO₂ injection into coal mass for enhancing CH₄ recovery, and fruitful results have been achieved. However, the aforementioned physical simulation experiments adopted cylindrical coal samples made of coal particles with the size below the millimeter level. Besides, experimental conditions in the laboratory, especially the stress and temperature, differed hugely from the real environment of coal seams. Hence, there are some defects in the aforementioned results [7,34–36]. Furthermore, in the field tests of CO₂-ECBM technology, the complex on-site geological environment constrained both the study on dynamic variation laws of gas during the CO₂ injection process and further understanding of CO₂-ECBM technology.

In view of the above research status, a new experimental apparatus was developed to simulate the process of enhancing CH₄ recovery by injecting CO₂. And the main innovation of this paper is the experimental design. Specifically speaking, it is reflected in three aspects: Firstly, a rectangular coal sample with a size of 300 * 70 * 70 mm as the research object, which is more close to the real coal seam in terms of scale. Therefore, the obtained migration laws of gases in this coal sample is more reliable compared with that of cylindrical coal samples with a size of 50 * 100 mm. Secondly, to better simulate the environment of coal seam, the condition of temperature and stress can be set according to the actual situation in our experimental system. Thirdly, some different types of sensors and monitoring devices are used on this experimental apparatus, which can better understand the effect of CO₂ injection.

In this paper, to analyze the CH₄ and CO₂ adsorption capacity of coal, the steady-state method was adopted to calculate permeability values under different conditions. In addition, the pore pressure, gaseous composition and flow rate at the outlet were tracked and tested in real time to dynamically monitor the effect and CO₂ consumption after CO₂ injection into CH₄-containing coal. Besides, the competition relationship between gas injection pressure and engineering efficiency and CO₂ consumption was discovered from our experimental results. Inspired by this relationship, we think a reasonable and effective economic cost model can be established as an effective method to guide the selection of gas injection pressure in the future [37]. Hence, the experimental study in this paper has positive significance for better understanding and application of CO₂-ECBM technology.

2. Materials and Methods

2.1. Coal sample preparation

Coal samples were obtained from No.2 coal seam, Haishiwan Coal Mine of Yaojie coal field, Gansu province, and they belong to gas-fat coal. To investigate the adsorption properties and the composition of

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