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CO₂ injection in coal: Advantages and influences of temperature and pressure

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ABSTRACT

Coalbed methane (CBM) recovery and CO2 storage have always been important topics of study for clean energy utilization and environmental governance. To enhance CBM recovery, CO2/N2 injection is applied in coal mining, especially CO2 injection enhanced coalbed methane (CO2-ECBM). CO2 injection not only improves the production of CBM but also helps to reduce the greenhouse effect. As there are differences between CO₂ injection and N_2 injection, the advantages of CO_2 should be studied in depth. The adsorbed methane accounts for more than 80% of total methane, and the amount of variation adsorbed methane is the key to CO_2/N_2 injection. In this paper, the desorption rate of adsorbed methane, the replacement ratio and the residual mixed gas content are discussed to evaluate the effect of CO₂/N₂ injection. After CO₂/N₂ injection, the desorption rate of adsorbed methane could be enhanced. The results also show that CO₂ injection is better than N₂ injection for enhanced coalbed methane (ECBM) recovery; however, the residual mixed gas content could be increased after CO_2 injection. Moreover, anthracite (QC) could be more suitable for CO2 injection because of the value of replacement ratio. The influences of temperature and injection pressure are also empirically studied. Increasing both temperature and pressure could not significantly enhance the desorption rate of adsorbed methane significant. Increasing the temperature could reduce the residual mixed gas content; however, it is difficult to induce ECBM recovery with increasing temperatures. Increasing the injection pressure could cause a slight increase of the desorption rate of the adsorbed methane; however, higher injection pressure corresponds to greater economic cost. Thus, the most suitable injection pressure is twice as much as the initial pressure of methane.

1. Introduction

The presence of coalbed methane (CBM) has always affected the safe exploitation of coal resources; however, it is also a source of clean energy [1,2]. CBM is the associated energy in the process of coal formation. With the development of clean energy utilization, an increasing number of countries are beginning to focus on CBM recovery. Because methane is both a clean fuel and a greenhouse gas, commercial exploitation of CBM has reduced greenhouse gas emissions and improved the utilization of resources especially in unminable coal. Some technologies for CBM recovery have developed rapidly, such as hydraulic displacement or air injection, especially CO_2/N_2 injection [3–5]. Gas injection and hydraulic technology are important methods for gas control in a coal seam, but there are some important differences

between the two. Hydraulic technology mainly uses high pressure water to create fractures in coal seam and improve the permeability of coal [6–8]. The intrusion of water can occupy the pores of coal and drive away the free gas. But water cannot enter the micropore and it can seal up the pores, so the gas in the pores cannot escape [9,10]. The gas molecule is smaller than water molecule, it can enter micropore [11,12]. The exchange sorption could be occurred between N₂/CO₂ molecules and CH₄ molecules. N₂/CO₂ injection could not only replace free methane, and reduce the content of adsorbed methane. Because gas molecules can enter the nano scale pores in coal, air injection has more advantages than hydraulic displacement. As a safe gas, CO₂ and N₂ are often injected in a coal seam to increase CBM production. To reduce the emissions of greenhouse gases, CO₂ storage in deep geological formations has been used in the enhanced coalbed methane (ECBM) approach

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[13-15].

Flaring of fossil fuels has resulted in the emission of CO₂, the amount can be reached 30 billion in 2010 according to the Energy Information Administration (EIA) [16]. CO2 injection in coal not only enhances CBM recovery, a process denoted as (CO2-ECBM), but also reduces environmental pollution [17]. The laboratory experiment was first conduced to study the emission rate of methane with CO₂ injection in column coal. CO2 injection is not the only method to increase emission of methane, as N2 injection is also very effective [18,19]. Adsorption and desorption are the basis of CO2/N2 injection in coal, and the adsorption capacities of N2, CO2, and CH4 in coal are different $(CO_2 > CH_4 > N_2)$ [20,21]. Shimada et al [22] conducted an adsorption experiment using powered bituminous coal, and found that the adsorption capacity ratio of $CO_2 \setminus CH_4 \setminus N_2$ is 5.5:2:1 when gas pressure reaches 6 MPa. The differences of adsorption capacity provides the possibility of CO₂/N₂ injection in coal; a large number of scholars have also focused on the adsorption mechanism of three gases in coal, aiming at explaining the mechanism of CO_2/N_2 injection [23]. The exchange sorption is the primary feature for CO_2/N_2 injection in coal; the CO_2 or N2 molecules are injected into the micropores of coal and occupy the adsorption sites of methane [24]. Exchange sorption impelled the adsorbed gas to be desorbed, causing mixed gas flow out of coal seam under high pressure difference. CO2 molecules have stronger adsorption capacity in coal. Therefore, CO2 injection will have more advantages than N2 injection in the process of exchange sorption. Many experiments have only considered the balance between input and output gas, and the replacement effect of adsorbed methane has not been further studied [23]. Adsorbed methane in coal occupies an important part in the total gas; thus, the effect of adsorbed methane by CO₂/N₂ injection should be further studied and comparison. Compared with N2 injection, the advantages of CO₂ injection should be analysed from the variation of adsorbed methane.

CH₄ recovery and CO₂ storage have been attracting attention recently in many countries [15,25]. Because of the high gas storage capacity of coal, CO₂ injection in coal is considered to be the better technology. The coal seam is more suitable for CO₂ storage and contributes to more environmental benefits [26–28]. Coal is a complex porous elastic medium, and a large number of micropores inside it provide the locations for gas storage [29–31]. A large number of natural fractures also provide gas flow channels for gas [32,33].

Exchange sorption is a complex physical and chemical process, and the influences of environmental factors are also very important. To study the mechanism of exchange adsorption, the adsorption and desorption experiments are usually conducted in the laboratory. Hamelinck et al. [34] studied the variation characteristics of the methane output using the adsorption test in the laboratory, and analysis of the potential commercial value of CO₂ injection. Coal is a special porous medium, coal matrix could be swelling and shrinkage during the gas adsorption and desorption [22,35]. Based on this factor, scholars have performed many experiments to evaluate CO₂ injection [36]. Zhou et al. [21] established the permeability model with the matrix deformation to predict the evolution of coal permeability, and compared the variation characteristics of CO2 and CH4 flowing in coal. The influences of temperature and pressure on gas adsorption and seepage are more significant and will affect the characteristics of exchange sorption in coal. The flow characteristics of CO₂ injection in coal are not the only factors; thus, this technology still faces many problems and requires further discussion. It is too simple to discuss the effect of CO₂ injection associated with sorption and permeability test; thus, more other data should be introduced into the experiment. More attention should be paid to the influences of environmental factors.

A large number of experiments have been performed on the changes of permeability and adsorption characteristics of coal in the process of CO_2 injection [21]. There is also lack of systematic research in evaluating the effect of CO_2 injection. The results show that CO_2 injection reduces permeability of coal, which is unfavourable for ECBM recovery [37]. However, the exchange sorption between CO_2 and CH_4 is stronger, helping to enhance desorption of adsorbed methane. Although N₂ injection has little effect on coal matrix deformation, the exchange sorption capacity is weak. Desorption of adsorbed methane in coal is an important evaluation index of CO_2/N_2 injection technology. In the previous studies, the volume changes of CO_2 input and CH_4 output are generally used in the CO_2 injection test [22]. However, including the free methane in tests and evolution of adsorbed methane has not been obtained enough attention. The deeper analysis of adsorbed methane would have more practical significance during CO_2/N_2 injection, with CO_2 injection being most widely used in CBM recovery. There are many factors affecting the exchange sorption; in particular, temperature and injection pressure have great influences on CBM recovery.

On the basis of summarizing previous test methods and results, the isothermal sorption experimental instrument is used to study the advantage and effect of CO_2 injection in this paper. The effects of N_2 injection and CO_2 injection on the displacement of adsorbed methane in coal are discussed, the advantages of CO_2 injection have been summarized using the rate of methane desorption, replacement ratio and residual mixed gas content. Three indicators can effectively reflect the change characteristics of adsorbed methane. The effect of temperature and pressure on displacement of adsorbed methane during CO_2 injection in coal is further analyzed using an experimental method. The results will help to guide CO_2 injection enhance coalbed methane (CO_2 -ECBM) in coal theoretically.

2. Experimental

2.1. Selection and preparation of coal samples

Three types of coal samples with different ranks were selected from Huaibei and Shanxi. Bituminous coal were obtained from Taoyuan coal mine (TY) and Qingdong coal mine (QD), anthracite coal was selected from Qincheng coal mine (QC). Three coal samples were obtained by drilling in coal seam, and sieved as 0.15–0.25CM particle size for testing. The proximate analysis and maximum vitrinite reflectance were tested, and the results are listed in Table 1.

2.2. Experimental device and method

The volumes of adsorption and desorption of gas in each particle coal sample are tested in the laboratory, and the volume of CH₄ displacement is obtained with CO₂/N₂ injection. The calculation method of gas quantity is adopted with the manometric method. The device mainly includes coal sample cell, thermostated water bath, vacuum pump and pressure gauge. The purity of three gases are more than 99.9%, the pressure transducers have the full-scale of 10 MPa with the accuracy of 0.05%, and the precision of water bath is accuracy of +0.1 °C. Experimental instrument is shown in Fig. 1.

Injecting adsorbing methane in the sample cell, the equilibrium pressure is 0.5 MPa at constant temperature. Next the mixture gas equilibrium pressure in sample cell reaches the target value, such as 1 MPa or 1.5 MPa, by CO_2/N_2 injection from reference cell. The volume of gases is obtained using manometric method. After adsorption equilibrium, the valve of sample cell is opened and the gas, which is dissolved saturated CO_2 , is collected by saturated brine discharge method.

Table 1
Proximate analysis and vitrinite reflectance.

Sample	M _{ad} (%)	A _d (%)	V_{daf} (%)	FC _d (%)	Vitrinite reflectance (%)
TY	1.27	5.66	37.12	58.26	0.78
QD	0.91	13.50	25.31	64.20	1.37
QC	1.51	11.59	7.70	82.78	2.78

 $M_{ad,}$ air-dried moisture; A_d dry ash; FC_d , dry fixed carbon; V_{daf} , dry ash free.

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