

# Impact research of CH<sub>4</sub> replacement with CO<sub>2</sub> in hydrous coal under high pressure injection

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## Abstract

**Purpose.** Based on high-pressure gas injection technology that enhances coal seam gas drainage, the effect of CH<sub>4</sub> replacement with CO<sub>2</sub> in aquiferous coal has been studied.

**Methods.** Using the laboratory experimental method and the self-built high-pressure gas injection experimental device, high-pressure CO<sub>2</sub> is injected into coal with different moisture contents to replace CH<sub>4</sub> under different adsorption equilibrium pressures.

**Findings.** With an increase in coal moisture content, the adsorption capacity of coal for CH<sub>4</sub> and CO<sub>2</sub> gradually weakens, the adsorption capacity for CO<sub>2</sub> is always greater than that of CH<sub>4</sub>, and the CH<sub>4</sub> replacement rate and the CO<sub>2</sub> injection ratio gradually decrease. It is concluded that the CH<sub>4</sub> replacement rate and the CO<sub>2</sub> injection ratio are negatively correlated with the water content of coal. With an increase of the pre-adsorption equilibrium CH<sub>4</sub> pressure (0.5, 0.75, 1.0, 1.3 and 2.0 MPa), the CH<sub>4</sub> replacement rate and the CO<sub>2</sub> injection ratio first sharply and then slowly increase. The transition point is 1.3 MPa (pre-adsorption equilibrium pressure of CH<sub>4</sub>).

**Originality.** Based on the adsorption characteristics of coal seam gas injection, the influence of coal water content and gas injection pressure on CH<sub>4</sub> replacement rate and CO<sub>2</sub> injection ratio is analyzed, and the mechanism is studied.

**Practical implications.** The experimental results have important guiding significance for selecting reasonable gas injection pressure and the source of gas to drive its injection into underground coal seam.

**Keywords:** moisture, coal, equilibrium pressure, methane, carbon dioxide, replacement ratio, injection ratio

## 1. Introduction

Coal contains abundant pore fissure, is a kind of natural adsorbent [1], the performance of different gases adsorption capacity of different [2]-[6] its adsorption properties in addition to the affected by external factors (temperature, pressure), also is determined by its physical and chemical properties, the water content of coal impact on adsorption performance is an important factor, and the effect is more complicated. Zhao-feng, W. et al. [7] believed that the adsorption capacity of coal to gas decreased gradually with the increase of water content, and there was a critical water content affecting the adsorption capacity of coal. When the water content exceeded the critical value, the adsorption capacity would not be affected. Shu-gang, L. et al. [8] analyzed the influence of coal's adsorption capacity on CH<sub>4</sub> by its water content by using Langmuir monolayer adsorption theory, and the results showed that water-bearing coal samples still met Langmuir monolayer adsorption theory, and adsorption constants A and B were correlated with the change of water content in coal samples to some extent. Jia-hao, W. et al. studied the displacement effect of water on gas, and measured the displacement desorption amount, pressure relief desorption amount and residual gas content in coal under different moisture content conditions. The results showed that water can promote displacement desorption of adsorbed

gas in coal, and the greater the moisture content, the greater the displacement desorption amount.

By analyzing the research contents of scholars, the conclusions of gas adsorption of coal with different moisture content are also different under different experimental conditions, and such conclusions trigger scholars to think that the law of replacing CH<sub>4</sub> in coal with high-pressure gas injection in coal with different moisture content should be discussed. Since the successful experiment of gas injection to improve the recovery of coal bed methane (CBM), scholars have carried out many studies on gas injection to promote CH<sub>4</sub> drainage in dry coal [9]-[12]. Zhao-feng, W., Hong-min, Y., Li-wei, C. et al. [13]-[18] believed that the mechanism of promoting CH<sub>4</sub> in coal drainage by gas injection mainly includes displacement adsorption and desorption, carrying / displacement of gas injection flow, dilution and diffusion of gas injection, and expansion and antireflection.

The weak adsorptive gas N<sub>2</sub> can not promote the desorption of CH<sub>4</sub> from coal through direct competition of adsorption sites, but can only be achieved by injecting gas to change the partial pressure of gas and destroy the original equilibrium. The strongly adsorbed gas CO<sub>2</sub> can compete with the CH<sub>4</sub> adsorbed on the surface of coal for desorption of CH<sub>4</sub> from coal [19]-[22]. With the deepening of research, scholars have put forward many important conclusions on

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the CH<sub>4</sub> effect of replacing coal by gas injection in dry coal. However, the influence of coal water content on the replacement effect needs further research. The author took anthracite of typical high gas mining area in China as the experimental object to study the difference of replacement CH<sub>4</sub> effect of CO<sub>2</sub> on coal samples with different moisture content.

## 2. The experimental method

### 2.1. The experimental device

The device is mainly composed of high pressure gas supply system, constant temperature adsorption and desorption system, gas component analysis system, vacuum system and data acquisition system, and the safety of the experimental device meets the test requirements. The schematic diagram of the experimental device is shown in Figure 1.

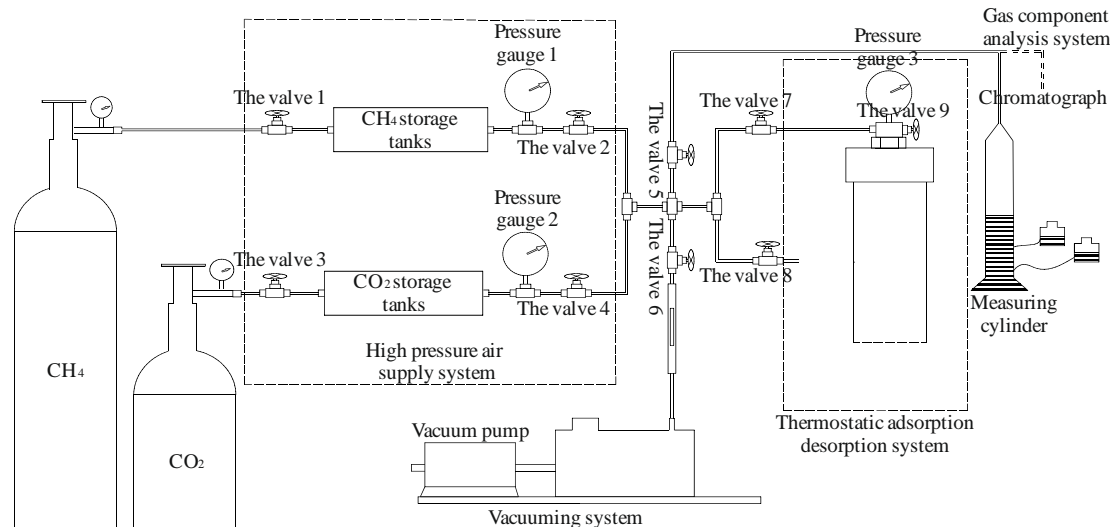


Figure 1. Schematic diagram of experimental device for CO<sub>2</sub> substitution of CH<sub>4</sub>

### 2.3. Coal sample preparation

The prepared granular coal was put into the air-blast drying oven, and the temperature was set at 105°C. After drying for 24 hours, the coal was put into a drying vessel for cooling, and a certain amount of water was added to stir it fully to make it uniform. Then the sample coal with the required moisture content was prepared by standing for 48 hours.

### 2.4. The experimental steps

Experiment 1: Check the air tightness of the system, vacuum the system, then fill CH<sub>4</sub> into each adsorption chamber containing coal samples with different moisture content and balance the adsorption to the set pressure, record the pressure change of gas storage vessel before and after injection, and finally calculate the adsorption amount of gas in the coal. Isothermal adsorption curves of CH<sub>4</sub> in coals with different moisture content were drawn based on calculation (the steps for drawing isothermal adsorption curves of CO<sub>2</sub> in coals with different moisture content were the same as above).

Experiment 2: The air tightness of the system was checked, the system was vacuumized, and then CH<sub>4</sub> was filled into the adsorption chamber containing coal samples to balance the adsorption to the target pressure, and quantitative CO<sub>2</sub> gas was filled into the coal sample chamber under high pressure. After replacement adsorption equilibrium, data were recorded and static gas samples were collected for gas chromatography analysis. Perform calculations, drawings and analyses based on recorded and analyzed data.

### 2.2. The experimental coal sample

The coal samples used in the experiment were selected from anthracite of Yangquan mine in Shanxi Province. The particle size of coal samples was 60-80 mesh, and the mass was  $180 \pm 0.01$  g. The experimental temperature was kept at a constant temperature of 20°C. The parameters of coal samples were shown in Table 1.

Table 1. Industrial analysis results of coal samples

Indicators	Measurement value
Moisture, %	1.67
Ash, %	17.03
Volatile, %	12.12
Truth versus density, g/cm <sup>3</sup>	1.52
Apparent relative density, g/cm <sup>3</sup>	1.41
Porosity, %	7.24

## 3. Experimental results and analysis

### 3.1. Adsorption law of gas in coal

In order to study the effect of coal with different moisture content on gas adsorption, experiments on CH<sub>4</sub> and CO<sub>2</sub> adsorption of coal with different moisture content were carried out. Langmuir equation was used to establish adsorption isotherms of pure CH<sub>4</sub> and CO<sub>2</sub> in coal samples with different moisture content, as shown in Figure 2. The amount of CO<sub>2</sub> absorbed by coal with the same moisture content is greater than that of CH<sub>4</sub>. With the increase of adsorption equilibrium pressure, the amount of CH<sub>4</sub> and CO<sub>2</sub> absorbed by coal increases, and the amount of adsorbed gas gradually slows down.

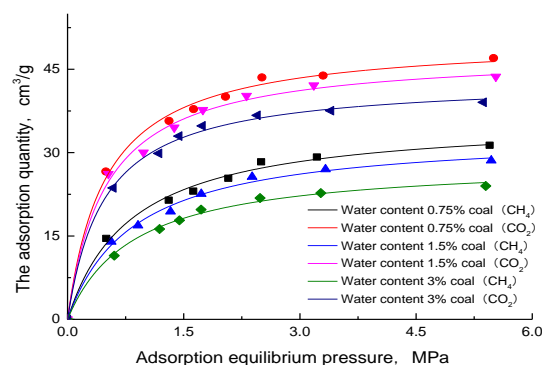


Figure 2. Langmuir isothermal adsorption curves of different gases in coal with different moisture content

That is, the adsorption capacity of gas CH<sub>4</sub> is less than CO<sub>2</sub>, which is consistent with the results of previous studies. The analysis shows that the frequency of gas molecules in the system impacting the coal surface increases with the increase of pressure, which leads to the increase of partial pressure of free gas in the coal body. Because the free state and adsorption state of gas are always in dynamic balance during the adsorption process, the number of adsorbed gas molecules increases. When the pressure is low, some gas molecules are difficult to enter the micropores, and with the increase of pressure, the micropore filling phenomenon will occur in the system. The reason why the growth rate of adsorption capacity gradually slows down with the increase of pressure is that when the pressure is high, there are more adsorption sites in coal body to achieve the dynamic balance between free phase and adsorption phase than in the state of low pressure, while the number of adsorption sites in coal body is fixed, so the growth rate of adsorption capacity slows down with the increase of pressure. Under the same adsorption equilibrium pressure, the adsorption law of coal with different moisture content is shown in Figure 3.

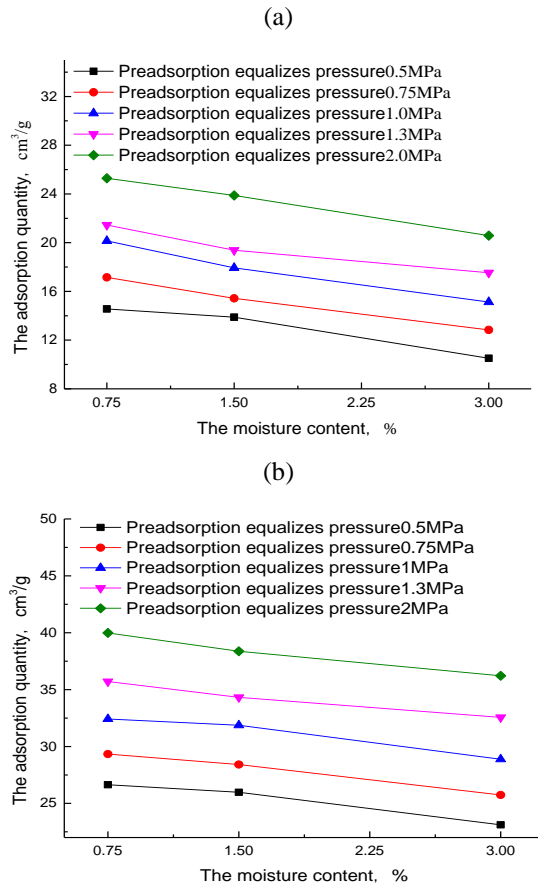


Figure 3. Variation of adsorption capacity of CH<sub>4</sub> and CO<sub>2</sub> in coal with water content at different adsorption equilibrium pressures: (a) CH<sub>4</sub>; (b) CH<sub>2</sub>

As can be seen from Figure 3, under the same pre-adsorption equilibrium pressure, the adsorption amount of CH<sub>4</sub> and CO<sub>2</sub> per unit mass of coal decreases gradually with the increase of coal water content.

Analysis reason, with the increase of water content, water in the coal molecular quantity also will increase, easy water molecules and coal breaking of chemical bonds and the inside of the coal matrix substrate surface hydrophilic functional

groups, to a certain extent, reduce the coal surface free energy, make the CH<sub>4</sub>/CO<sub>2</sub> – coal adsorption system heat release of the balance is less, and, The molecular force between water and coal is stronger than that of CH<sub>4</sub>/CO<sub>2</sub>, which can occupy the effective adsorption sites on the coal surface, thus weakening the adsorption capacity of aqueous coal for CH<sub>4</sub>/CO<sub>2</sub>.

### 3.2. CH<sub>4</sub> replacement rate of coal with different moisture content

High pressure injection is when coal sample chamber CH<sub>4</sub> adsorption equilibrium pressure reaches a constant, using the equivalent replacement of piston pump will be higher pressure gas chamber into the coal samples, its purpose is to after the replacement source gas injection, reduce the free volume fraction of CH<sub>4</sub> in the gas phase after injection pressure is reduced, resulting in CH<sub>4</sub> in coal adsorption quantity is reduced, the free volume increase, CH<sub>4</sub> is displaced from coal.

In order to study the difference of CH<sub>4</sub> replacement effect of CO<sub>2</sub> in coals with different moisture content under high pressure injection, the CH<sub>4</sub> replacement rate was selected to measure the CH<sub>4</sub> replacement effect of injection source gas in coals with different moisture content. CH<sub>4</sub> replacement rate refers to the ratio of the change in the amount of CH<sub>4</sub> absorbed by coal before and after gas injection to the amount of CH<sub>4</sub> absorbed by coal before gas injection, as shown in Formula 1:

$$R_{r,CH_4} = \frac{\Delta Q_{CH_4}}{Q_{1,CH_4}} \cdot 100\% = \frac{Q_{1,CH_4} - Q_{2,CH_4}}{Q_{1,CH_4}} \cdot 100\%, \quad (1)$$

where:

$R_{r,CH_4}$  – the replacement rate of CH<sub>4</sub>, %;

$Q_{1,CH_4}$ ,  $Q_{2,CH_4}$  – the adsorption capacity of CH<sub>4</sub> in coal before and after the experiment, cm<sup>3</sup>/g.

After the high-pressure injection test of coals with different moisture content, the replacement amount and replacement rate of CH<sub>4</sub> are shown in Figure 4. As can be seen from Figure 4, both the replacement amount and replacement rate of CH<sub>4</sub> increase rapidly before the equilibrium pressure of CH<sub>4</sub> pre-adsorption is 1.3 MPa, and slowly after that. This indicates that the pursuit of high gas injection pressure should not be pursued in the downhole gas injection to replace CH<sub>4</sub>, wasting resources and increasing costs.

After coal with different moisture content is injected under different high pressure, the replacement amount and replacement rate of CH<sub>4</sub> are shown in Figure 5. That under the same equilibrium pressure of CH<sub>4</sub> pre-adsorption, the displacement amount of CH<sub>4</sub> decreases with the increase of coal water content. Within the range of 0.75 to 3% moisture content in coal, CH<sub>4</sub> desorption rate decreases in a “downhill” pattern.

Based on the analysis, in the process of high pressure injection, injection of CO<sub>2</sub> cavity after total volume, total pressure, reduce the free volume fraction of CH<sub>4</sub> in CH<sub>4</sub> in the gas phase after the injection of CO<sub>2</sub> partial pressure is reduced, resulting in CH<sub>4</sub> in coal adsorption quantity is reduced, the free volume to increase, and free of CO<sub>2</sub> adsorption on coal, to establish a new equilibrium. In addition, AS an adsorbent gas, CO<sub>2</sub> is bound to have competitive adsorption with CH<sub>4</sub>, and the coal sample holes are bound to absorb a small amount of CO<sub>2</sub>, occupying part of the adsorption space and inhibiting part of CH<sub>4</sub> adsorption. The adsorption state of CH<sub>4</sub> tends to be transformed into a free state for various reasons, which explains why the injection of high-pressure CO<sub>2</sub> can make CH<sub>4</sub> desorbed out.

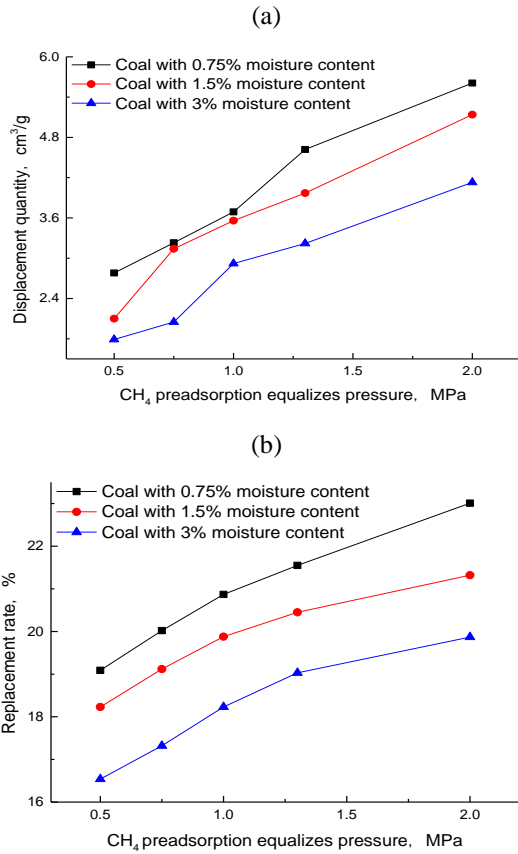


Figure 4. Variation of CH<sub>4</sub> replacement amount and replacement rate in coals with different water content with equilibrium pressure of CH<sub>4</sub> pre-adsorption: (a) displacement; (b) replacement rate

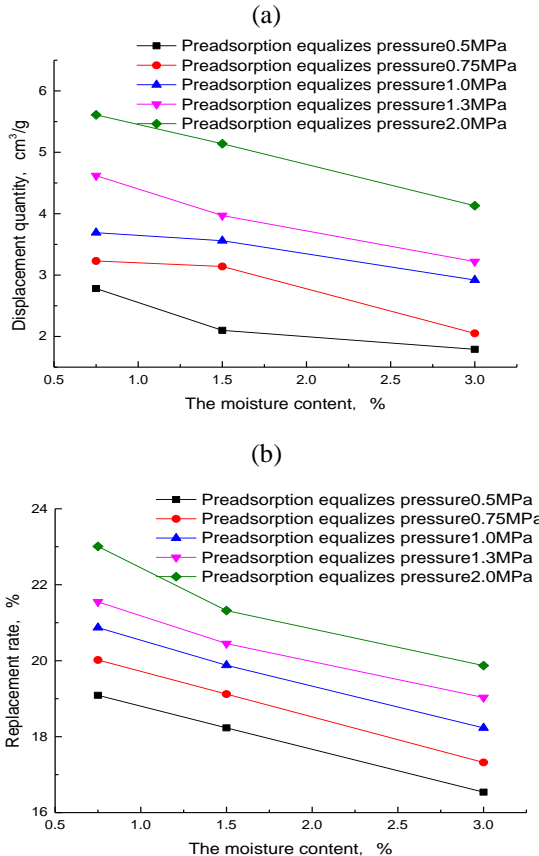


Figure 5. Variation of CH<sub>4</sub> replacement amount and replacement rate with water content of coal under different high pressure injection: (a) displacement; (b) replacement rate

The replacement rate of CH<sub>4</sub> in coal with high moisture content is low, because water can inhibit the adsorption and desorption of CH<sub>4</sub> in coal. For example, when the adsorption equilibrium pressure of CH<sub>4</sub> is 1.3 MPa, the adsorption amount of CH<sub>4</sub> (21.45 cm<sup>3</sup>/g) of coal with 0.75% moisture content is 1.11 times that of coal with 1.5% moisture content (19.38 cm<sup>3</sup>/g). It is 1.22 times that of 3% coal (17.53 cm<sup>3</sup>/g). The water molecular weight of coal with high moisture content is large, and water molecules can occupy effective adsorption sites on the coal surface, which weakens the adsorption capacity of CH<sub>4</sub>. Finally, with the increase of water content, CH<sub>4</sub> replacement rate weakens.

### 3.3. CO<sub>2</sub> injection ratio of coal with different moisture content

In order to study the replacement efficiency of CO<sub>2</sub> injection on CH<sub>4</sub> adsorption in coal, it is expressed by the replacement and injection ratio of CO<sub>2</sub> to CH<sub>4</sub> in coal, referred to as CO<sub>2</sub> injection ratio, which refers to the ratio of the replacement amount of CH<sub>4</sub> and the injected amount of CO<sub>2</sub> after balanced high-pressure injection. On the other hand, it represents the efficiency that CO<sub>2</sub> per unit volume can replace CH<sub>4</sub> in coal. See Equation 2:

$$R_{r,CH_2} = \frac{AQ_{CH_4}}{AQ_{1,CO_4}} \cdot 100\% = \frac{Q_{1,CH_4} - Q_{2,CH_4}}{Q_{1,CO_4}} \cdot 100\%, \quad (2)$$

where:

$R_{r,CH_2}$  – injection ratio, %;

$Q_{CO_2}$  is the injected amount of CO<sub>2</sub> before high-pressure gas injection, cm<sup>3</sup>/g.

Figure 6 and 7 show CO<sub>2</sub> injection after high-pressure injection experiment for coals with different moisture content.

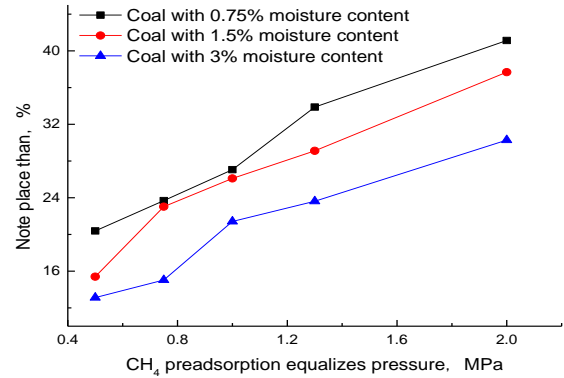


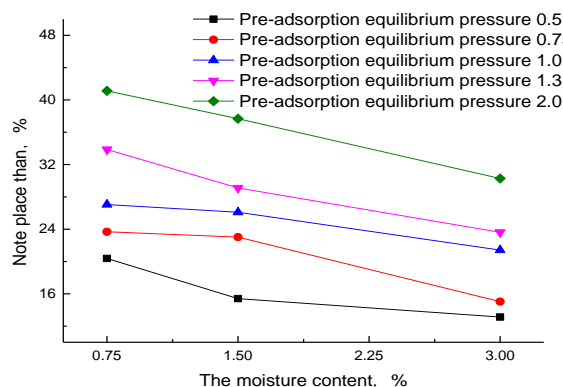
Figure 6. Variation of CO<sub>2</sub> injection ratio in coals with different water content with equilibrium pressure of CH<sub>4</sub> pre-adsorption

In the coal with the same moisture content under high pressure injection, CO<sub>2</sub> injection ratio increases sharply with the increase of CH<sub>4</sub> pre-adsorption equilibrium pressure and then increases slowly. Under the same CH<sub>4</sub> pre-adsorption equilibrium pressure condition, CO<sub>2</sub> injection ratio decreases with the increase of coal moisture content.

A comparative analysis of the replacement amount of CH<sub>4</sub>, replacement rate of CH<sub>4</sub> and CO<sub>2</sub> injection ratio after high pressure injection equilibrium showed that:

1. Increasing CO<sub>2</sub> injection pressure can make the replacement amount and replacement of CH<sub>4</sub> increase rapidly at first and then slowly, resulting in the similar phenomenon of CO<sub>2</sub> injection ratio as above.





**Figure 7. Variation of CO<sub>2</sub> injection ratio with coal water content at different CH<sub>4</sub> pre-adsorption equilibrium pressures**

Therefore, for the engineering technology of underground CO<sub>2</sub> injection to replace coal seam CH<sub>4</sub>, blindly increasing the gas injection pressure during high-pressure injection will lead to a decline in CO<sub>2</sub> replacement efficiency.

2. With the increase of coal water content, the replacement amount of CH<sub>4</sub> and CO<sub>2</sub> injection ratio decrease, and the replacement efficiency decreases. In other words, under the same gas injection condition, the displacement amount of coal with high moisture content is small and the displacement efficiency is low.

#### 4. Conclusions

In coal with the same moisture content, the adsorption capacity of CH<sub>4</sub> and CO<sub>2</sub> in coal increases with the increase of injection pressure; With the increase of coal water content, the amount of CH<sub>4</sub> and CO<sub>2</sub> absorbed by coal weakens, and the amount of CO<sub>2</sub> absorbed by coal is greater than that of CH<sub>4</sub>.

In the process of high pressure injection, both CH<sub>4</sub> replacement rate and CO<sub>2</sub> injection ratio are negatively correlated with the water content of coal. With the increase of CH<sub>4</sub> pre-adsorption equilibrium pressure, the CH<sub>4</sub> replacement rate and CO<sub>2</sub> injection ratio show a trend of sharp increase at first and then slow increase, and the transition point is 1.3 MPa (CH<sub>4</sub> pre-adsorption equilibrium pressure).

The replacement amount of CH<sub>4</sub>, replacement rate of CH<sub>4</sub> and CO<sub>2</sub> injection ratio are mainly affected by CO<sub>2</sub> injection volume, coal water content, coal adsorption to gas, “adsorption competition” between gases and “partial pressure” caused by high pressure injection.

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#### Дослідження впливу заміни CH<sub>4</sub> на CO<sub>2</sub> у водомісткому вугіллі під високим тиском закачування

Л. Чен, М. Чжао, К. Лі, Я. Лю

**Мета.** Дослідження впливу заміщення CH<sub>4</sub> на CO<sub>2</sub> у водомісткому вугіллі на основі технології закачування газу високого тиску, що покращує відведення газу вугільних пластів.

**Методика.** Використовуючи лабораторний експериментальний метод та власноруч виготовлений експериментальний пристрій для впорскування газу високого тиску, CO<sub>2</sub> високого тиску вводили у вугілля з різним вмістом води для заміни CH<sub>4</sub> при різних тисках рівноваги адсорбції.

**Результати.** Зі збільшенням вмісту води, здатність вугілля адсорбувати  $\text{CH}_4$  і  $\text{CO}_2$  поступово слабшає, адсорбційна здатність  $\text{CO}_2$  завжди перевищує адсорбційну здатність  $\text{CH}_4$ , а ефективність заміщення  $\text{CH}_4$  і коефіцієнт закачування  $\text{CO}_2$  поступово зменшуються. Зроблено висновок, що швидкість заміщення  $\text{CH}_4$  та коефіцієнт закачування  $\text{CO}_2$  негативно корелюють із вмістом води у вугіллі. Зі збільшенням передадсорбційного рівноважного тиску  $\text{CH}_4$  (0.5, 0.75, 1.0, 1.3 та 2.0 МПа) швидкість заміщення  $\text{CH}_4$  і коефіцієнт введення  $\text{CO}_2$  спочатку різко, а потім повільно збільшується. Точка переходу становить 1.3 МПа (передадсорбційний рівноважний тиск  $\text{CH}_4$ ).

**Наукова новизна.** Виявлено за адсорбційними характеристиками закачування газу у вугільний пласт особливості впливу вмісту води у вугіллі та тиску закачування газу на ефективність заміщення  $\text{CH}_4$  та коефіцієнт закачування газу  $\text{CO}_2$ , а також досліджено механізм взаємодії.

**Практична значимість.** Результати експерименту мають важливе орієнтовне значення для вибору розумного тиску закачування газу та джерела газу для закачування його в підземний вугільний пласт.

**Ключові слова:** вологість, вугілля, рівноважний тиск, метан, вуглекислий газ, коефіцієнт заміни, коефіцієнт впорскування