Scientific Research Publishing

Natural Resources, 2024, 15, 69-81
https://www.scirp.org/journal/nr
ISSN Online: 2158-7086
ISSN Print: 2158-706X

The Effectiveness of the Continuous and Cyclic Method on CO_2-ECBM

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How to cite this paper: Tambaria, T.N. and Sugai, Y. (2024) The Effectiveness of the Continuous and Cyclic Method on CO_2-ECBM. *Natural Resources*, **15**, 69-81.
https://doi.org/10.4236/nr.2024.153006

Received: February 5, 2024
Accepted: March 12, 2024
Published: March 15, 2024

Abstract

This study examines the impact of different CO_2 injection methods on coalbed methane recovery. Specifically, this study investigated the effectiveness of continuously injecting CO_2 versus injecting CO_2 that had been soaked for two weeks. The objective was to ascertain which approach was more successful in enhancing CO_2 Enhanced coalbed Methane (CO_2-ECBM). The experiment involved injecting 3 MPa of CH_4 into dry coal samples, allowing it to adsorb until reaching equilibrium, and then injecting 5 MPa of CO_2 to recover adsorbed CH_4. The continuous method recovered CH_4 without detectable effluent concentration for 5 hours, but desorption efficiency was only 26% due to fast flow. On the other hand, the desorption efficiency of the cyclic method was only 12%, indicating trapped CH_4. A comparison of desorption efficiency per unit of time shows the continuous method is more effective than the cyclic method. The results of this study demonstrate the continuous method is more effective for the desorption of CH_4, and its efficiency can be improved by briefly soaking CO_2 on coal and then reinjecting it to maximize CH_4 recovery. It is advisable to limit the soaking time to prevent excessive swelling of the coal matrix, which can hinder seam flow and harm long-term gas production.

Keywords

Coalbed Methane, CO_2 Injection, Desorption Efficiency

1. Introduction

The world is facing an energy crisis, and there is a growing debate on reducing the risk of future disruptions and promoting energy security. Due to high demand and extravagant consumption, fossil fuel reserves are being depleted at an alarming rate. Unfortunately, fossil fuel power plants are still being used to sup-

port excess cooling demand during extreme summer heat, which increases emissions by 15% - 40% [1]. It will be challenging to achieve the goal of reducing CO_2 emissions to 23 Gt by 2030 and to zero by 2050, a trajectory necessary to limit the temperature increase to less than 1.5˚C in 2100 [2]. This difficulty is compounded by climate policy delays after the COVID-19 pandemic, and geopolitical factors have led to increased fossil fuel investment in some regions [3].

Coalbed methane (CBM) is a gas found within coal seams that shows promise as an alternative energy source. CBM is a cleaner and more affordable fuel option than traditional fossil fuels, making it a vital resource for meeting global energy demands [4]. Its primary component is methane (CH_4), but it also includes secondary components like carbon dioxide (CO_2), nitrogen (N_2), and trace amounts of higher hydrocarbon gases (C_2-C_4) like ethane, propane, and butane in different proportions [5]. Various techniques exist for extracting CBM, but ECBM via gas injection is the most effective method for increasing CBM production [6]. CO_2 injection has successfully boosted CH_4 production by replacing adsorbed CH_4 on pore size due to CO_2's high affinity with coal [7]. CO_2 injection increases the overall CH_4 desorption and enhances the CH_4 desorption rate per unit time by 14% and per unit mass of coal by 35% [8]. CO_2-ECBM is a promising technique that boosts coal bed methane production and facilitates CO_2 storage in coal seams. This method of CO_2 storage is technically viable and environmentally friendly [9]. Raising the pressure and volume of CO_2 injection can significantly amplify the amount of CO_2 sequestration in the coal seam [10]. It could lead to a more efficient and effective method of reducing carbon emissions. Furthermore, coal seams offer a long-term CO_2 storage solution that can aid in combating climate change and air pollution [7] [11] [12].

The efficiency of gas recovery is influenced by various factors, such as the time of injectant gas breakthrough, flow velocity within the coal seam, and the proportion of the seam gas displaced [13]. To improve gas recovery efficiency, there are several gas recovery processes, such as CO_2 injection continuous method and CO_2 cyclic method (**Figure 1**). In the CO_2 continuous method, the CO_2 floods and CH_4 displacement were greater than 99%, but it can result in rapid breakthroughs of CO_2 to the production well through highly permeable formations, gravity separation, and a significant reduction in sweep efficiency [14] [15]. Additionally, if CO_2 is injected continuously for an extended period, the coal seam will gradually become saturated with CO_2 from the well to the distal area [16]. The CO_2 cyclic method is more favorable when the condensate is near the wellbore as it can increase the recovery factors and minimize the risk of gas breakthrough into the production wells [15] [17]. However, the soaking time between the injection well and production well is crucial, and research suggests using shorter or no soaking is more efficacious [18]. It is important to note that the limitation of each method in CO-ECBM makes them need a more profound understanding. In addition, applying high pressure during gas production can lead to higher production rates. However, this can also have a negative effect on the long-term productivity of gas production by reducing the flow ability through the

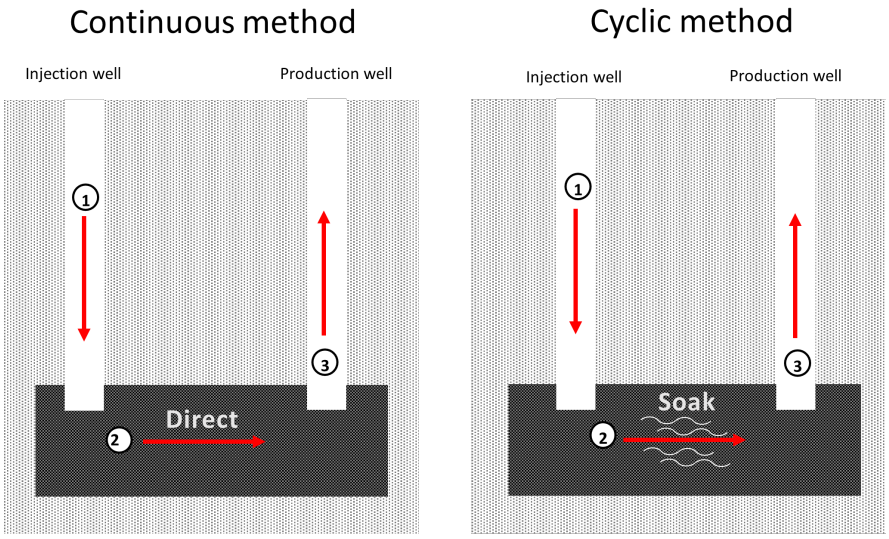

Figure 1. Gas recovery process.

seam [19]. It has been found that the self-diffusivity of adsorbed gas molecules is lower under the strongest confinement effect and decreases with increased pressure [20]. Furthermore, the effectiveness of CO_2 continuous and cyclic below critical pressure has not yet been conducted.

2. Experimental Methods

The CO_2 continuous and cyclic tests were made using Chinese coal. The samples were analyzed for their proximate properties using ASTM D1373-73, D3174-73, and D3175-77, and the findings were recorded Table 1. The coal was then crushed, pulverized into 1.86 μm particles, and dried for over 12 hours in an oven at 378.15 K. After that, it was cooled in a vacuum state and placed in a 75 mL sample cell until the total empty volume reached 18.46 mL.

2.1. Experiment Measurement for CH_4 Adsorption on Coal

The coal samples were loaded into the sample cell, which was then carefully arranged in the experimental device per the schematic diagram (Figure 2). The sample cell was seamlessly positioned between a pressure gauge and a filter, and two valves efficiently regulated the gas flow in and out of the equipment. The sample cell was injected with CH_4 to initiate the adsorption process, and the pressure gauge accurately measured the resulting pressure. A filter was strategically

Table 1. Proximate analysis of Chinese coal used in the experiment.

Proximate parameter (%, a.r)	
Moisture	4.4
Ash	3.87
Volatile matter	38.97
Fixed carbon	52.76

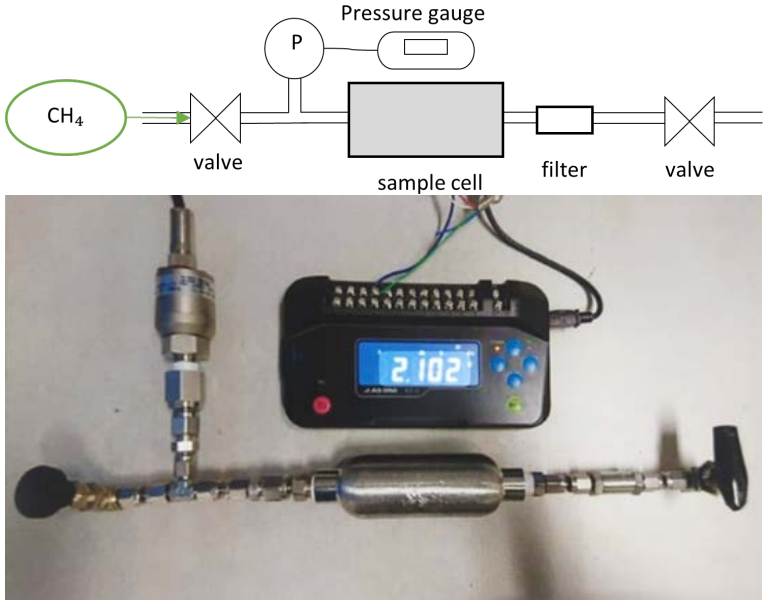

Figure 2. Schematic diagram of experimental equipment.

employed to prevent the coal powder from escaping with the gas from the equipment. Furthermore, the average weight of the equipment in a vacuum was meticulously recorded as 807.625 g.

Methane gas was injected into the equipment at room temperature while maintaining a pressure of 3 MPa. The pressure was kept constant for a period of 5 to 8 hours to ensure that an equilibrium state had been reached. Afterward, the weight of the equipment was measured using an electronic balance. The difference between the weight of the equipment in a vacuum state and the weight of the equipment when filled with methane was then recorded. Adsorption was taken into account during the process. The amount of CH_4 injected is calculated as [21].

$$n = \frac{(W - W_0)}{m} \tag{1}$$

where n is the amount of injected CH_4 (mol), W is the equipment weight after the gas injected and reached equilibrium (g), W_0 is the equipment weight in vacuum state (g), m is the gas molecular weight (g/mol).

Furthermore, it is possible to determine the quantity of gas that is not adsorbed to the coal by utilizing the following equation as [22].

$$PV = Zn_f RT \tag{2}$$

where P is the gas pressure (MPa), V is the equipment internal volume (mL), Z is the compression coefficient, n_f is the free gas amount (mol), R is the gas constant (J/K mol), T is the temperature (K).

The equation for determining the quantity of gas adsorbed on coal surface in the equipment has been formulated as [22].

$$n' = n - n_f \tag{3}$$

where n' is the adsorbed gas amount (mol).

The equations can measure the quantity of gas adsorbed on coal but cannot determine the rate of adsorption. The only way to estimate the rate of adsorption is by making assumptions while considering other parameters such as pressure and temperature.

2.2. Continuous Test Experiment for CH_4 Recovery from Coal

The research involved the adsorption of methane to coal, followed by the injection of carbon dioxide to extract the methane from the coal. The equipment used for both continuous and cyclic tests featured a microflow control valve and a water displacement device at the outflow side, as depicted in Figure 3. The microflow control valve was employed to adjust the outflow in small increments.

The experiment involved injecting CO_2 at a pressure of 5 MPa and room temperature. The process of displacement was being carried out while keeping the outflow constant. The experiment continued until steady state conditions were achieved, where the inflow and outflow gas composition and rate were equal. The duration required to attain a stable state was meticulously recorded during the experiment. The weight of the equipment was measured using an electronic balance to determine the amount of CO_2 injected.

As part of the continuous testing process, the CH_4 gas released from the equipment was efficiently collected in 10 mL sample bottles. To facilitate the gas collection process, a 100 mL cylinder was placed in a water displacement device, and the released gas was collected into the cylinder at a fixed outflow rate of 4.650 mL/min. Each collected gas sample was meticulously analyzed using a gas thermal conductivity detector (TCD) chromatograph. To accurately measure gas production, a needle was connected through the rubber stopper of the sample bottles, and the stopcock was opened to let the gas displace the liquid in the cylinder. Finally, gas samples were directly taken from the sample bottles using a specialized chromatography syringe. In the experiments assessing natural CH_4 desorption by CO_2 replacement, the desorption efficiency and desorption efficiency per unit time was defined as [8].

$$De = \frac{Q}{Q_0} \tag{4}$$

$$Dep = \frac{De}{TE} \tag{5}$$

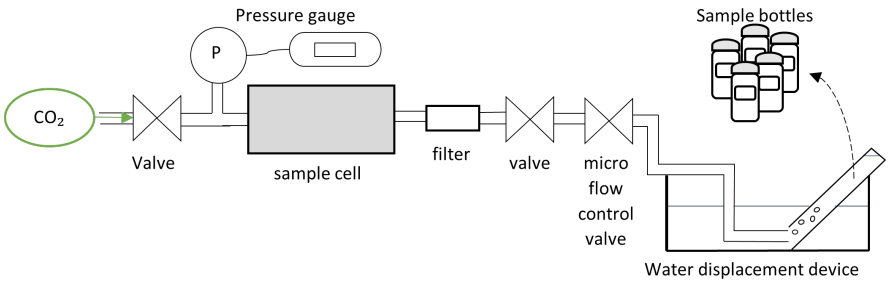

Figure 3. Schematic diagram of CH_4 recovery experiment.

where De is the desorption efficiency (%), Q is the total desorption amount (mol), and Q_0 is initial adsorption amount (mol), Dep is desorption efficiency per unit time (%), and TE is desorption time (h).

2.3. Cyclic test Experiment for CH_4 Recovery from Coal

In this experiment, the cyclic method was employed as a means of ensuring an adequate soaking time or cyclic phase between injection and adsorption. As such, the test involved the saturation of CO_2 for a duration of two weeks, during which the valves were kept firmly shut to create a sealed state. To obtain a comprehensive data set, samples of gas were collected every 12 hours, with a total of 28 samples being obtained over the two-week period.

3. Experimental Results and Discussion

3.1. CH_4 Adsorption on Coal

Coal has the capability to adsorb methane easily, and it takes around 40 minutes to reach an equilibrium state. However, research into CH_4 adsorption simulation has shown that several micropores are closed and unable to adsorb CH_4 [23]. This condition leads to variations in the state of adsorbed and free CH_4 [8]. In this study, CH_4 adsorption was calculated by considering the excess CH_4 adsorption amount and free CH_4. As shown in **Figure 4** higher injection pressure leads to a greater amount of CH_4 adsorption. A significant increase was observed between 0 and 1 MPa, with a slower increase between 1.5 and 3 MPa. The observed phenomenon results from increased pressure, which facilitates adsorption and enhances the degree of spontaneity of CH_4 adsorption [24].

3.2. Analysis of CH_4 Recovery in Continuous Test

In this test, the CO_2 injection was continuous and recovery process was stopped after reaching steady state where the CO_2 inflow and outflow gas compositions

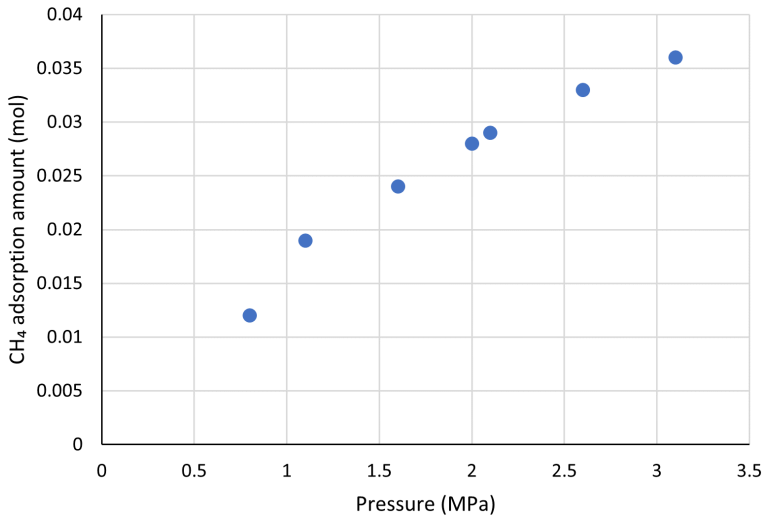

Figure 4. Relationship between pressure and CH_4 adsorption on the coal.

and rates are equal. To attain the steady-state condition, the collection of samples in bottles was carried out for a total of 8 hours. This process was repeated 15 times to ensure its accuracy and reliability. The effects of alterations in CH_4 concentration on the CO_2-ECBM behavior over time during gas injection. The results of the continuous testing, as shown in Figure 5, indicate that CO_2 initially caused a surge in methane production for 30 minutes, followed by a slight decrease for the subsequent 90 minutes, and then a gradual decline until methane production ceased at 300 minutes. In conclusion, the recovery rate of CH_4 is faster in the early stage and slower in the later stage. During the early stages, the high pressure of CO_2 facilitated a substantial amount of CH_4 to flow into the outflow system. As time passed and CO_2 injection rates remained constant, it became increasingly difficult to locate CH_4. By the total desorption calculation, the desorption efficiency from continuous test is 26% with desorption efficiency per unit time is 3.25%.

3.3. The Results of Cyclic Test

It was observed during cyclic testing that the rate of CH_4 adsorption was faster than that of CO_2. CO_2 injection under 5 MPa took about 5 hours to reach equilibrium. The results of the cyclic experiment were different from the continuous test. In the continuous test, the CH_4 concentration decreased over 150 hours and then slightly decreased until 350 hours (Figure 6). The CO_2 concentration was measured in this study to observe the sorption process on the coal matrix over the long term. The results showed that the CO_2 concentration slightly increased until 350 hours. The cyclic testing process showed different results from the continuous test, based on the duration of the test. In continuous testing, the peak of desorption occurs in a short time, while in cyclic testing, the peak of desorption remains stable over several hours. During continuous testing, CH_4 was only detected for 300 minutes, whereas cyclic testing continues for a longer period of time. According to calculations, the desorption efficiency from cyclic tests is 12%, with a desorption efficiency per unit time of 0.03%.

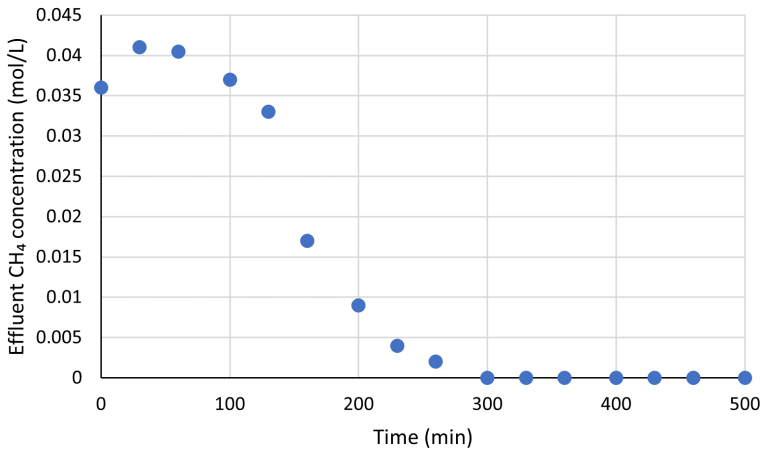

Figure 5. Changes in outflow CH_4 concentration over time during CO_2 injection.

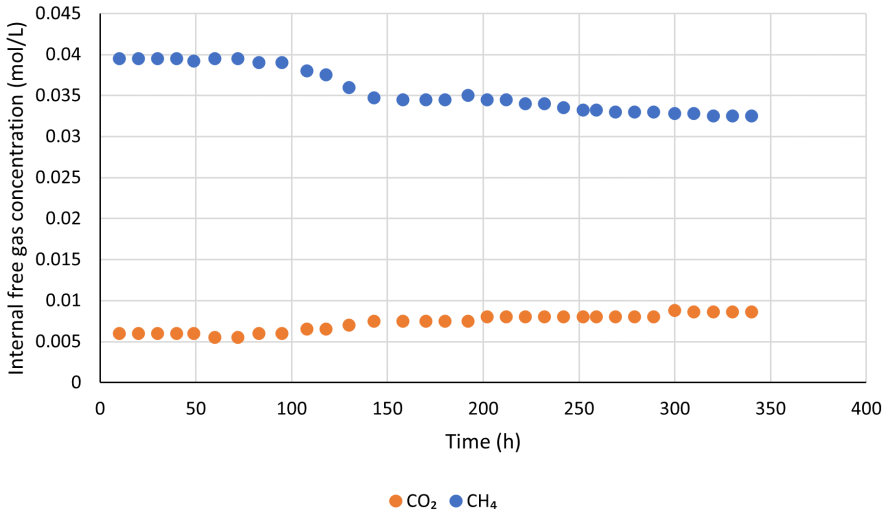

Figure 6. Change in free gas amount during shut in after CO_2 injection.

3.4. The Effective Method for ECBM

In continuous test, several stages occur during CH_4 production, as shown in Figure 7. During the initial phase, that methane CH_4 in its free state, adsorbed onto the coal surface and subsequently absorbed into the coal matrix. The second stage shows the significant impact of CO_2 pressure, which prompts the outflow of CH_4. Following the complete drying and vacuuming of the experimental sample, it was observed that the original coal sample lacked any water content except for adsorbed CH_4, thereby facilitating the flow of CH_4 to the outflow. Most of the remaining CH_4 was transported to the fracture space of the coal sample through large pores, where it transformed into a completely free state [8]. The process of CH_4 desorption from the coal matrix results in the contraction of the matrix, leading to a reduction in the horizontal stress and, subsequently, an increase in the permeability of cleats [25]. During the third stage, the phenomenon of Fickian diffusion takes place within the smaller pore sizes in the mesopores, leading to a sustained increase in mesopore diffusivity [26]. The current condition enables a greater accessibility of the coal matrix, thereby facilitating the adsorption of CO_2.

During the last stage of the process, the conditions stabilize, and no traces of methane are detected in the outflow. As the adsorption of CO_2 increases, there is also a corresponding increase in porosity [27]. As the permeability of coal decreases, the rate of CO_2 flow gradually slows down, leading to a breakthrough at the outlet, and results in a CO_2 concentration of 90% [28]. Based on the test results, it is evident that the continuous test successfully recovered CH_4. The continuous test successfully recovered CH_4, but recent research suggests that during the process of employing flooding CO_2 ECBM, a significant proportion of CH_4 is carried away by CO_2, instead of being removed primarily via the replacement method [28]. In addition to the adsorbed and absorbed methane, the swelling of the coal matrix can make it difficult to remove CH_4. Using continuous methods

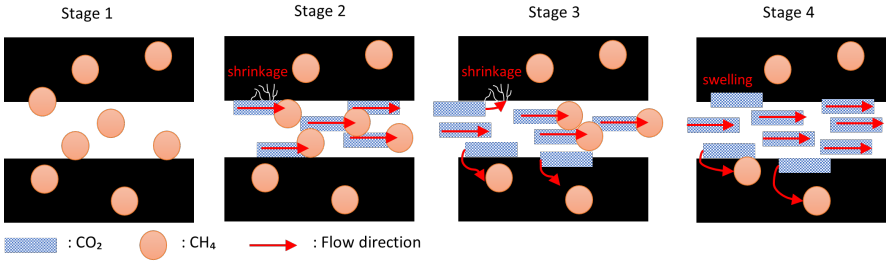

Figure 7. Mass transport process for CO_2 and CH_4 during continuous test.

can speed up the process, but it can also decrease the likelihood of CO_2 adsorbing and replacing CH_4 on the coal matrix.

During the observation of the CH_4 recovery process, it was found that the cyclic test demonstrated distinct stages that were not observed during the continuous test. Although the initial stage of both tests was the same, the subsequent stages of CH_4 recovery differed (**Figure 8**). During the second stage, CO_2 was injected and allowed to soak into coal samples, allowing CH_4 to desorb from coal into a free state. Coal samples can release CH_4 due to pressure differences and CH_4 could diffuses through the matrix pores faster than CO_2, indicating that CH_4 flows faster than CO_2 under the same pressure gradient [8]. In the case of CO_2, a stable condition was created because the CO_2 pressure in the coal near the injection well quickly reached the same level as the injection pressure and remained constant with the CO_2 concentration in the coal matrix gradually increased over time [16].

On last stage where CH_4 decrease, and CO_2 slightly increase into the end of the process. This condition occurs when the gas concentration in coal increases gradually, causing a decrease in gradient between it and the boundary concentration, which in turn reduces the amount of CO_2 adsorbed by coal [16]. Additionally, once the process of adsorption is finished, the quantity of CO_2 that gets desorbed is measured [29]. On the other hand, the competition between different mechanisms of gaseous methane diffusion and the swelling of the coal matrix caused by gas adsorption was raised [26]. The desorption curve experiences a decline indicating that only a limited number of methane molecules can desorb, with the residual gas being retained [30]. This condition worsens when small pores reach maximum adsorption capacity, leading to

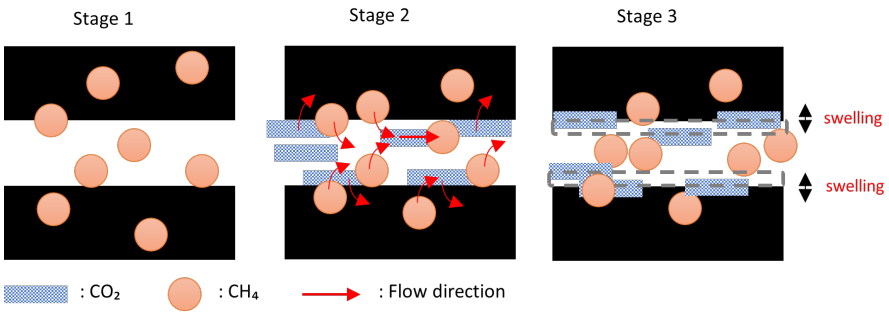

Figure 8. Mass transport process for CO_2 and CH_4 during cyclic test.

dominant swelling effects in the coal matrix [31]. Due to various factors, the CH_4 trapped in coal seams cannot be released smoothly, which may hinder CH_4 recovery improvement.

The results obtained from continuous and cyclic tests have revealed significant differences in the desorption efficiency and desorption per unit time, as shown in Figure 9. This calculation has been formulated considering the desorption efficiency of adsorbed methane on coal. The results show that the continuous method has better desorption efficiency and time than the cyclic method. However, the desorption efficiency in continuous test is not significant. This condition is caused by pressure, which pushes CO_2 to outflow faster, resulting in a shorter time for CO_2 to adsorb and absorb, thus leading to ineffective CH_4 desorption. During cyclic testing, the injection pressure no longer has an effect after a certain point due to the long time it takes for the process to complete. This results in CO_2 adsorption and swelling, which makes it difficult for CH_4 to desorb and flow out of the system. If this process were to be applied at an industrial scale, it is possible that the quantity of methane produced may fall below the threshold of economic viability, rendering the process unfeasible and requiring its discontinuation. Based on these results, it can be concluded that pressure and time are the primary factors impacting the efficiency of CH_4 recovery.

This study showcases the effectiveness of CH_4 desorption using an extreme example. Other publications have used cyclic testing, with a few hours of soaking time, repeated multiple times, to maximize CH_4 recovery [32] [33]. To optimize the research, a synergistic approach involving both continuous and cyclic methods can be employed, whereby CO_2 is subjected to a brief soaking period, followed by reinjection to attain maximum CH4 recovery. Furthermore, the research can be improved by considering coal seam permeability, porosity evolution, adsorption capacity, and geological conditions.

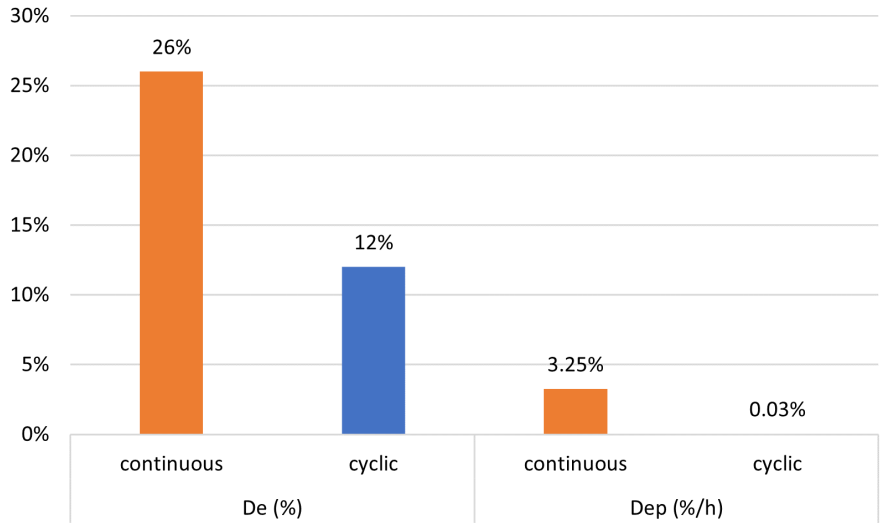

Figure 9. Total desorption efficiency and desorption rate per unit time during different experiments.

4. Conclusions

The efficiency of continuous and cyclic on CO_2-ECBM was investigated in this study and the following major conclusions can be drawn:

- The continuous test successfully recovered CH_4 with no detectable effluent concentration during the first 5 hours. However, desorption efficiency was only 26%, indicating that CO_2 flow was too fast to displace all adsorbed CH_4 from coal.
- In a two-week cyclic experiment, methane levels decreased, suggesting CH_4 was trapped and difficult to remove due to swelling. The desorption efficiency from cyclic test was lower than the continuous test, indicating this method has a low chance of CH_4 recovery.
- The continuous method is more effective in terms of time to recover CH_4 as it shows a higher desorption efficiency per unit time compared to the cyclic method.
- The CH_4 desorption can be more effective using the continuous method. It can be further improved by briefly soaking CO_2 on coal and then reinjecting it to maximize CH_4 recovery. This research suggests a shorter soaking time to prevent greater coal matrix swelling that may negatively affect long-term gas production by reducing the flow ability of the seams.

Further analysis is needed to determine the optimal duration for CO_2 soaking to increase CH_4 recovery and minimize long-term harm in production.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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