

Original Paper

A small-scale experimental study of CO₂ enhanced injectivity methods of the high-rank coal



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ABSTRACT

The attenuation of CO₂ injectivity has become the biggest technical barrier for the application of CO₂ enhanced coalbed methane recovery (CO₂-ECBM). Commonly, the intermittent CO₂ injection, N₂ displacing CO₂ and pre-fracturing are the potential CO₂ enhanced injectivity methods for coal reservoirs, but their mechanism and effectiveness remain to be clarified. This paper thus conducted small-scale experiments to simulate the working process of these engineering measures by an independently developed experimental device. Results show that the CO₂ injectivity of coal is remarkably improved by the intermittent injection mode since the CO₂ injection time is increased by folds and the loss of reservoir pressure can be complemented in time. The N₂ displacing CO₂ method promotes the desorption of CO₂ and reduces the swelling strain, with the result that the permeability of coal is improved by 74.82% and 64.95% compared with the methods of the primary subcritical CO₂ (SubCO₂) and supercritical CO₂ (ScCO₂) injection. However, the permeability reduces again with the secondary CO₂ injection. The permeability of the coal sample after pre-fracturing is averagely improved by 1–2 orders of magnitude, the irreversible permeability loss rate, average stress sensitivity coefficient and the permeability loss rate due to adsorption are averagely reduced by 95.885%, 61.538% and 96.297%, respectively. This indicates that the permeability of coal after pre-fracturing is no longer sensitive to both the effective stress and ScCO₂ adsorption, the injectivity is thus improved and stable. The CO₂ enhanced injectivity effects of the intermittent CO₂ injection, the N₂ displacing CO₂ and the pre-fracturing are various, which thus can be selected individually or jointly to improve the CO₂ injectivity according to the reservoir physical properties and geological conditions. This research deepens the understanding of the functional mechanism of CO₂ enhanced injectivity methods and provides some guidance for their selection and application in engineering practices.

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1. Introduction

Anthropogenic greenhouse gas emissions have become the principal consideration related to the global warming, as one of the primary greenhouse gases, CO₂ takes an inescapable responsibility

for climate change (Cheng et al., 2019; Li et al., 2015; Zhang et al., 2015). Consequently, reducing CO₂ emissions is gradually becoming the consensus of people. Carbon capture and sequestration (CCS) technology is an essential approach to reduce the CO₂ level in the atmosphere, which thus has aroused widespread

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concerns by scientists and governments around the world (Bakhshian et al., 2020; Zhao et al., 2017; Zhou et al., 2019). Currently, the depleted oil or gas reservoirs, saline aquifers and unmineable coal seams are the main geological sequestration sites (Xu et al., 2020; Zhang et al., 2017). Besides the superiority to realize geological CO₂ sequestration, injecting CO₂ into deep unmineable coal seams can also enhance coalbed methane recovery (CO₂-ECBM) with the displacement effect for CH₄, which will thus make up for the shortage of conventional energy (Ajayi et al., 2019; Niu et al. 2019a, 2020a, 2021). A large number of pilot tests have been implemented on a worldwide scale, distributing in the San Juan Basin, Illinois Basin, Williston Basin and Appalachian Basin in the United States, the Silesian coal basin in Poland, the Fenn-Big Valley in Canada, the Ishikari Coal Basin in Japan, the Qinshui Basin and Ordos Basin in China (Gunter et al., 2005; Oudinot et al., 2011; Pan et al., 2017; van Bergen et al., 2009; Yamazaki et al., 2006). However, one of the technical barriers for CO₂-ECBM is that the CO₂ injectivity is significantly lowered. As reported previously, the CO₂ injection rate was reduced by 40% during the initial injection stages at the Allison Unit (Reeves, 2003); a 70% reduction of the injection capability appeared during the first year of operation in the Ishikari Basin (Fujioka et al., 2010), and the loss of injection capacity was also experienced in the micro pilot field test in Qinshui Basin (Wong et al., 2007).

The most sensitive parameter influencing the CO₂ injectivity is permeability. It has been estimated that the low injectivity of CO₂ for a coal seam is caused by the reduction in permeability induced by coal swelling (Fujioka et al., 2010). For the deep unmineable coal seam, the CO₂ adsorption swelling of coal matrix will extend inward and compress the fracture space under the high overburden pressure, causing the attenuation of permeability in coal (Jin et al. 2019, 2020; Niu et al., 2017b; Zhou et al., 2020). The three-dimensional fracture network is extensively developed in the coal seams, mainly including the cleats developed in vertical bedding plane direction (VBD) and the bedding fractures distributed in parallel bedding plane direction (PBD) (Wang et al. 2018a, 2020a, 2020a). The structures of the two types of fractures are dynamically altered during the CO₂ injection process, and both of them contribute to the reduction of CO₂ injectivity (Niu et al., 2018). Moreover, the moisture content and temperature also affect the CO₂ injectivity by varying the permeability of coal (Kumar et al., 2012), but the influence level is moderate compared with the adsorption swelling of coal (Niu et al., 2019b; Wang et al., 2020b). All in all, reactions between the injected CO₂ and the coal matrix restrain the CO₂ injectivity and then affect the engineering effect of CO₂-ECBM.

To overcome this technical challenge, two major categories of optimizing injection scheme and coal reservoir stimulation are attempted to improve the CO₂ injectivity. Optimizing the injection scheme is mainly accomplished through changing the CO₂ injection rate or injection pressure and the composition of the injected gas, such as intermittent CO₂ injection (Li et al., 2016) and mixed gas injection (Fan et al., 2019). The intermittent CO₂ injection is often employed to enhanced oil recovery (EOR). For example, Singh (2018) compared four different injection schemes (i.e., constant rate, stepwise increasing rate, stepwise decreasing rate and cyclic rate), and considered that it would be preferable to store CO₂ using a cyclic injection scheme in storage reservoirs. Mixed gas injection means injecting N₂/CO₂ mixture or N₂ displacing CO₂, because the adsorption swelling strain induced by N₂ is far less than that caused by CO₂, the permeability loss affected by N₂ adsorption swelling is tiny. On this basis, Oudinot et al. (2017) considered that a larger amount of N₂ needs to be injected to limit the permeability loss (on the order of 85%). Other research intended to enhance CO₂ injectivity by improving permeability through stimulating the coal reservoir, such as hydraulic fracturing and waterless fracturing (Cao

et al., 2017; Fu and Liu, 2019; Liu et al., 2021; Osiptsov, 2017). Additionally, ultrasonic wave treatment and microwave irradiation treatment are capable of improving the porosity and promoting the formation of fractures of coal (Huang et al., 2019; Shi et al. 2017, 2019). However, these methods are either limited or have not been applied to engineering practices. Up to now, few scholars have conducted experimental simulations regarding the representative enhanced CO₂ injectivity methods (intermittent CO₂ injection, N₂ displacing CO₂ and pre-fracturing). Thus, the effectiveness verifications of these methods are deficient and their functional mechanisms are uncertain, which restricts the application of CO₂ enhanced injectivity technologies.

Given this, the methods of intermittent CO₂ injection, N₂ displacing CO₂ and pre-fracturing are first simulated by an independently developed experimental device, then, the injectivity enhancement effect is verified and the functional mechanism is clarified. This research will fill the gaps in studies of CO₂ injectivity on coal reservoirs and provide theoretical guidance for the implementations of CO₂-ECBM.

2. Experimental

2.1. Sampling

2.1.1. Preparation of core samples

Coal samples used were collected from Chengzhuang Coal Mine, Qinshui Basin, which is a successful commercialization area for CBM development (Li et al., 2018; Yun et al., 2012) and the target area for micro pilot tests of CO₂-ECBM (Pan et al., 2017). The coal blocks were obtained from the coal seams far away from the geological structural zone, which makes the samples more representative. The gathered coal blocks were wrapped by several layers of cling films and put in sampling bags to avoid the oxidation of coal surfaces. Then they were transported to the laboratory and stored in a constant temperature and humidity box.

Coal core samples with a length of 100 mm and a diameter of 50 mm were drilled from the coal blocks by a vertical drilling machine along the VBD (Fig. 1). Then the ends and profiles of samples were polished smoothly by the sandpapers. Cares were taken during the whole sample preparation process, aiming to obtain the relatively homogeneous coal samples without obvious fractures. The ASTM D2013 standard was consulted during the sampling stage. The proximate analysis, ultimate analysis and maceral composition analysis were conducted and shown in Table 1. The selected coals with maximum vitrinite reflectance ($R_{o,max}$) of 2.96% belong to high-rank coals. The size and weight were measured by a vernier caliper and a high-precision electronic balance and the results are shown in Table 2.

2.1.2. Preparation of coal sample with artificial crack

To study the permeability enhancement effect by fracturing, Kumar et al. (2015) and Wu et al. (2018) proposed to simulate the fracture crack through an artificial fracture by cutting the rock core. Accordingly, the coal sample with artificial crack was prepared by the following steps: (1) A wire cutting machine was adopted to cut the coal sample in half, and the cutting surfaces were polished by abrasive papers to form an idealized crack. (2) The quartz sands with a diameter of 0.15–0.42 mm were spread evenly on the crack surface. (3) The split-cores were re-mated (sample 4) and put into the rubber sleeve of the sample cell.

2.2. Experimental apparatus

Fig. 2 shows a schematic diagram of the experimental apparatus used. Two gas cylinders full of He and CO₂ are the gas sources in this

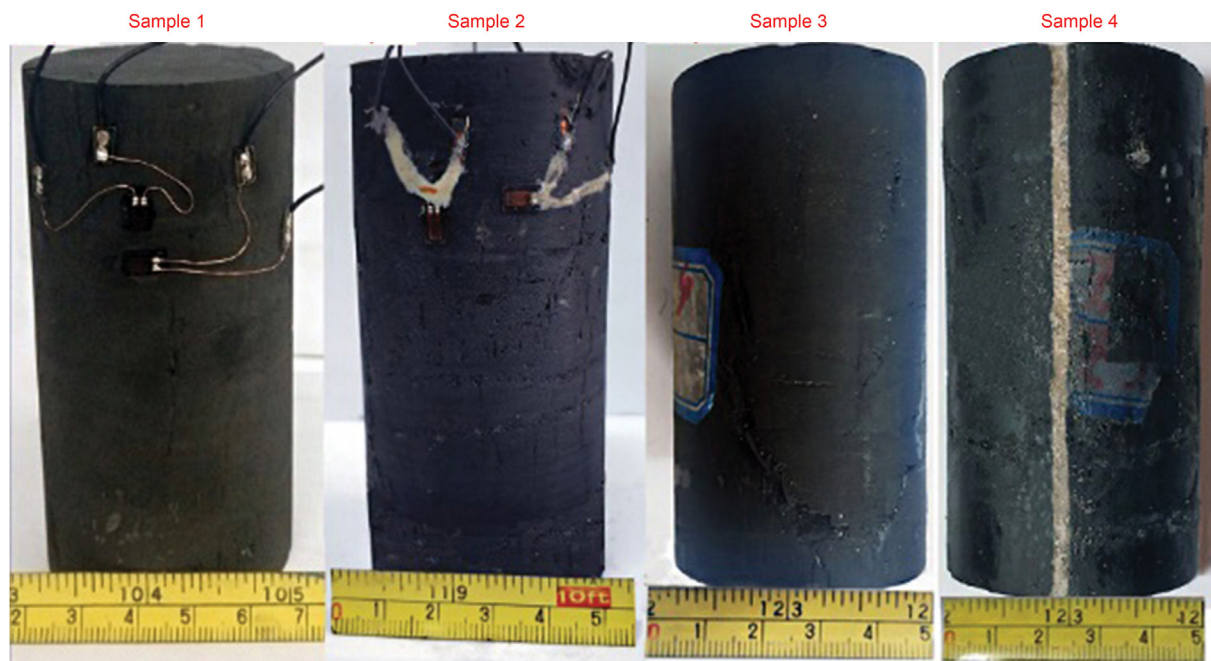


Fig. 1. The core samples used.

Table 1
Basic information of core samples taken from Chengzhuang Coal Mine.

$R_{o,max}$, %	Proximate analysis, wt%				Ultimate analysis, wt%				Maceral composition, vol%		
	M_{ad}	A_{ad}	V_{daf}	FC_{ad}	O_{daf}	C_{daf}	H_{daf}	N_{daf}	Vit	Ine	Min
3.27	92.84	2.31	3.27	81.72	3.27	92.84	2.31	3.27	75.80	21.40	2.80

Note: M_{ad} , moisture content of air-dried basis; A_{ad} , ash content of air-dried basis; V_{daf} , volatile content of dry ash-free basis; FC_{ad} , fixed carbon content of air-dried basis; O_{daf} , oxygen content of dry ash-free basis; C_{daf} , carbon content of dry ash-free basis; H_{daf} , hydrogen content of dry ash-free basis; N_{daf} , nitrogen content of dry ash-free basis; Vit, vitrinite; Ine, inertinite; Min, mineral.

experiment. The gas booster is used to increase the gas pressure, the maximum target pressure can reach 30 MPa. The vacuum pump can extract the residual gases in this apparatus until the pressure in the pipeline is reduced to -0.1 MPa, providing a vacuum environment for the test. The reference cell with a volume of 5000 mL is a gas storing device, a heating system is installed in it and provides a constant temperature environment in the whole experimental process. The pressure transducer can monitor the gas pressure, with a precision of 0.01 MPa. The air compressor, oil reservoir and axial piston pump are combined to provide an axial pressure for the coal sample, the maximum normal load is 2000 kN. The syringe pump is used to provide a confining pressure for the coal sample (the maximum value is 100 MPa). The axial linear variable differential transformer (axial LVDT) and the radial LVDT with a precision of $0.1 \mu m$ are used to measure the deformation of the sample, respectively. A mass flow meter and a gas chromatograph are adopted to monitor the gas flow rate and gas component content, respectively. The experimental apparatus is controlled by built-in

Table 2
Physical properties of coal core samples.

Sample ID	Length, cm	Diameter, cm	Weight, g	Density, g/cm ³
1	9.784	4.916	273.672	1.474
2	9.436	4.966	259.037	1.412
3	8.604	4.986	242.281	1.443
4	8.664	5.048	252.302	1.456

software on the computer, and all the data can be real-time recorded.

2.3. Experimental procedure

2.3.1. Experimental procedure for the continuous/intermittent CO₂ injection

This experiment was conducted to compare the effect of continuous and intermittent injection mode for improving the CO₂ injectivity. Sample 1 was used in this experiment. In the whole process, the instantaneous flow rate, injection pressure and the strain of the coal sample were real-time monitored. The experimental procedures were described as follows:

- (1) Sample 1 was installed in the sample cell, and then the axial pressure and the confining pressure were increased to 12 MPa and the experimental temperature was adjusted to 35 °C.
- (2) The vacuum treatment was performed with a vacuum pump for the whole device.
- (3) The pneumatic valve of the reference cell and the inlet valve of the sample cell were immediately opened to introduce CO₂ into the sample cell. The injection pressure was kept at 4 MPa. The injection process would be stopped until the inlet flow rate dropped to zero.

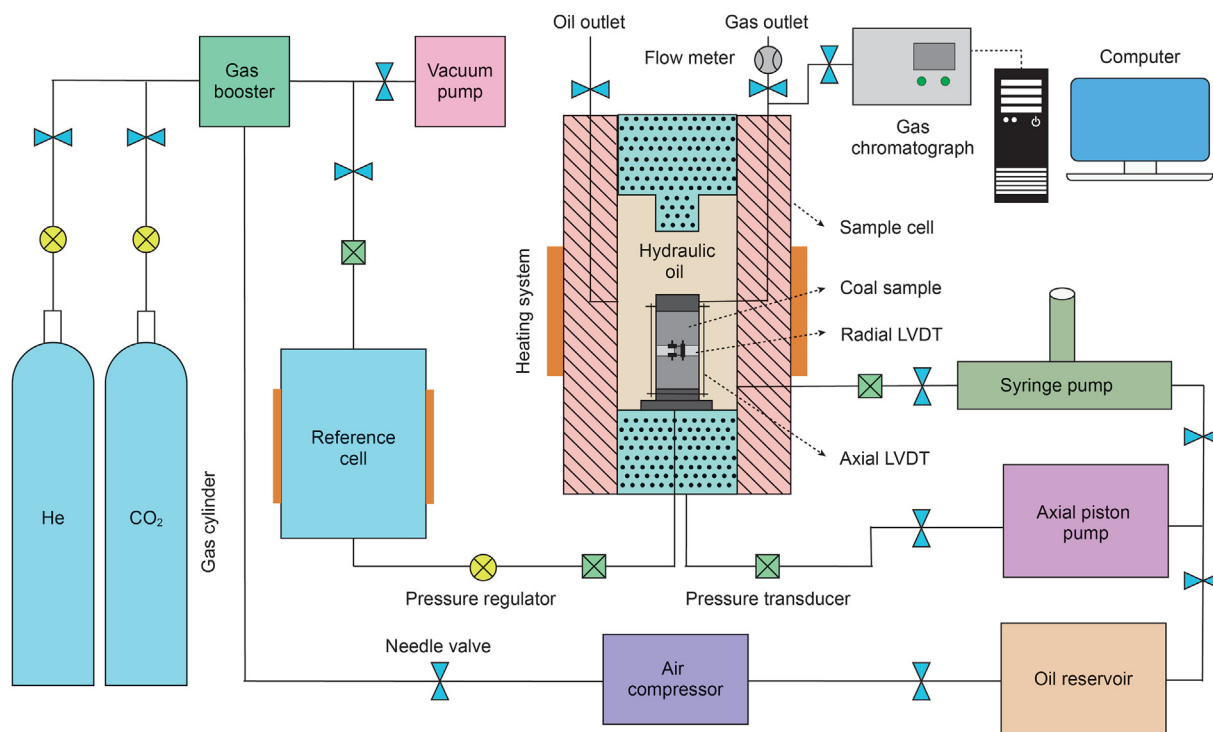


Fig. 2. Schematic diagram of the experimental apparatus.

- (4) CO₂ was expelled from the experimental device, the adsorbed CO₂ was extracted from the coal sample by the vacuum pump for 24 h until the desorption process was completed.
- (5) CO₂ was injected into the sample cell at an injection pressure of 4 MPa and kept for 0.5 h. Then, the inlet valve of the sample cell was closed and the injection process was suspended for 0.5 h. The injection/stop cycle was repeated for ten times until the inlet flow rate dropped to zero;
- (6) CO₂ was introduced into the reference cell at an injection pressure of 8 MPa, and steps (1)–(5) were repeated to accomplish the simulation of continuous/intermittent ScCO₂ injection.

2.3.2. Experimental procedure for N₂ displacing CO₂

This experiment was conducted to simulate the effect of N₂ displacing CO₂ for improving the CO₂ injectivity. Sample 2 was used in this test. The experimental procedures were described as follows:

- (1) Sample 2 was installed into the sample cell. The axial pressure and confining pressure were increased to 12 MPa and the experimental temperature was controlled at 35 °C.
- (2) The initial N₂ permeability of Sample 2 was measured at a confining pressure of 12 MPa, an injection pressure of 4 MPa and a back pressure of 2 MPa.
- (3) After the whole device was evacuated with the vacuum pump, CO₂ was injected into the sample cell at an injection pressure of 4 MPa, and the strain and permeability of the coal were real-time monitored.
- (4) The outlet valve was closed and CO₂ adsorption process was lasted for 24 h, the back pressure valve was regulated to control the outlet pressure to 2 MPa, and N₂ was injected to displace CO₂ at a pressure of 4 MPa, the strain and permeability of coal and the component content of outlet gas were

dynamically monitored until the content of N₂ is close to 100%.

- (5) When the process of N₂ displacing CO₂ was completed, CO₂ was reinjected into the sample cell at an pressure of 4 MPa, and the strain and permeability of coal and the component content of exit gas were recorded, until the content of CO₂ was close to 100%;
- (6) The N₂ and CO₂ pressure were increased to 8 MPa, the back pressure was adjusted to 6 MPa, and steps (1)–(5) were repeated to conduct the experiment of N₂ displacing ScCO₂.

2.3.3. Experimental procedure on the enhanced CO₂ injectivity by pre-fracturing

The permeability of coal seams is controlled by multiple factors during the CO₂-ECBM process, such as CO₂ adsorption, effective stress, moisture, temperature and metamorphism degree (Ju et al., 2016; Meng and Li, 2013; Zhi et al., 2019), and it is commonly considered that the former two are the most crucial factors (Niu et al., 2019b). Additionally, when CO₂ is injected into the deep unmineable coal seam, it turns to from the subcritical CO₂ (SubCO₂) to the supercritical CO₂ (ScCO₂) when the pressure is greater than 7.4 MPa and the temperature is larger than 31 °C. Therefore, in this section, the experiment is to ascertain the different impacts of effective stress and ScCO₂ on the permeability of coal before and after pre-fracturing. Samples 3 and 4 were adopted to conduct this test and the flow chart is exhibited in Fig. 3. The permeability of Samples 3 and 4 under a loading/unloading cycle was measured (5 → 7 → 9 → 11 → 13 → 11 → 9 → 7 → 5 MPa for Sample 3, and 5 → 8 → 11 → 14 → 17 → 14 → 11 → 8 → 5 MPa for Sample 4), aiming to investigate the influence of effective stress on the permeability of coal before and after pre-fracturing. The CO₂ permeability of Samples 3 and 4 before and after ScCO₂ adsorption was performed at various injection pressures, aiming to probe the influence of ScCO₂ adsorption on the permeability of coal before

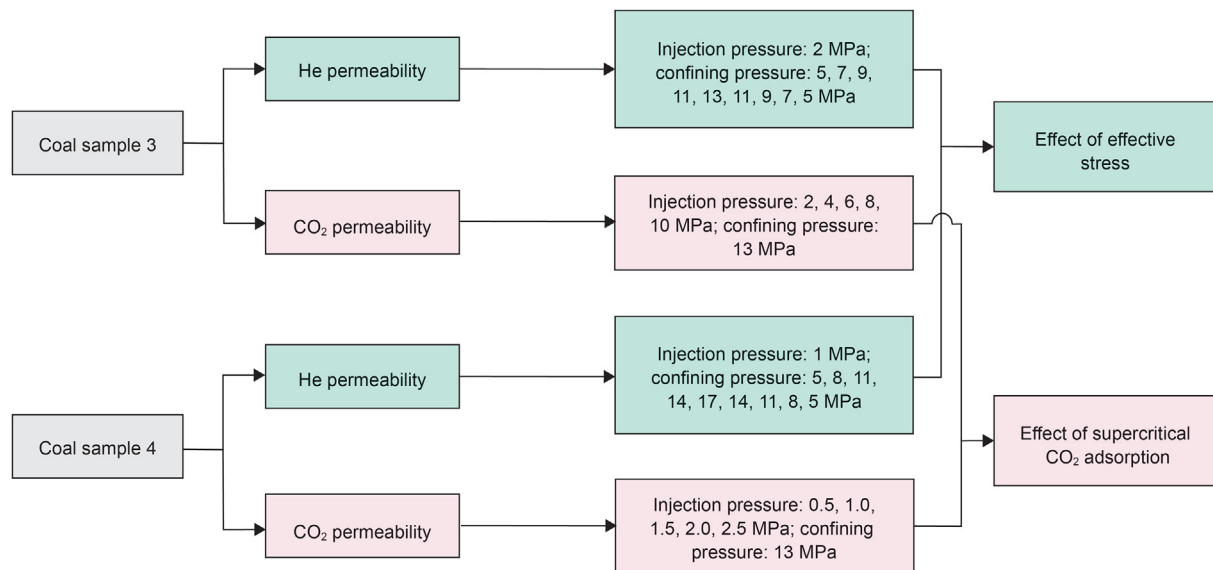


Fig. 3. Flow chart of permeability experiment on samples before/after pre-fracturing.

and after pre-fracturing. The adsorption time of ScCO₂ was 24 h, in the whole experiment, the axial pressure was 13 MPa and the experimental temperature was 35 °C. The favorable burial depth of coal seams for coalbed methane development in the Qinshui Basin is approximately 500–800 m (Shao et al., 2015), the selected experimental temperature/pressure match with the real conditions of these coal seams, according to Meng et al. (2011).

3. Results and discussion

3.1. Intermittent injection method

3.1.1. Continuous/intermittent SubCO₂ injection

Fig. 4 shows the changes of injection pressure, confining pressure, instantaneous flow rate and volumetric strain during the continuous/intermittent SubCO₂ injection. During the continuous injection of the CO₂ into the sample at an injection pressure of 4 MPa (Fig. 4a), the instantaneous flow rate showed several fluctuations and the peak values gradually decreased at the early stage (the injection time <1.6 h). With the progressive increase in the accumulated flow rate, the adsorption swelling strain of the coal matrix appeared and elevated to 0.50%, at this time, the instantaneous flow rate of CO₂ was decreased to zero. This indicates that the swelling strain affected the seepage path of the coal matrix and

further reduces the injectivity of the sample. The total injection volume of CO₂ is 1079.79 mL, the pre-calibrated free space volume is 149.70 mL, therefore, the net injection volume of CO₂ is 930.09 mL under the continuous SubCO₂ injection mode.

For the intermittent SubCO₂ injection mode, ten injection/stop cycles were conducted, and the interval time was 0.5 h. Fig. 4b shows that the instantaneous flow rate of CO₂ increased to a peak value quickly when the entrance valve was opened, and then it was rapidly attenuated. When the entrance valve was closed, the instantaneous flow rate was reduced gradually, and the CO₂ pressure also decreased because the adsorption process was continued. With the increase in the number of injection/stop cycles, the peak value of the instantaneous flow rate gradually decreased, and it reduced to zero when the volumetric strain reached 0.60%. The total injection volume of CO₂ is 1301.15 mL, and the net injection volume of CO₂ is 1151.45 mL after deducting the free space volume under the intermittent SubCO₂ injection mode. The net SubCO₂ injection volume by intermittent injection mode is improved by 23.80%, compared with that by continuous injection mode.

3.1.2. Continuous/intermittent ScCO₂ injection

The experimental results of continuous/intermittent ScCO₂ injection are shown in Fig. 5. As can be seen from Fig. 5a, after the injection of ScCO₂ at 8 MPa, the instantaneous flow rate of CO₂ also

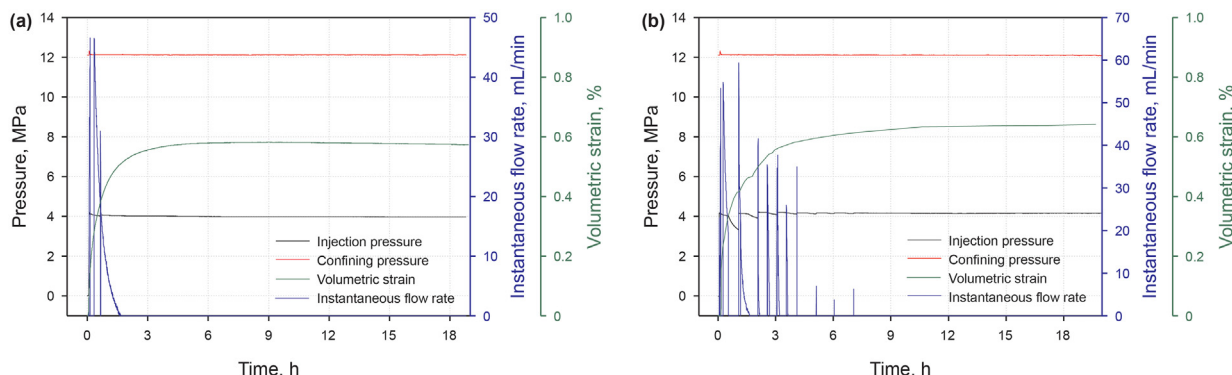


Fig. 4. Experimental results of continuous/intermittent injection of SubCO₂ into the sample. (a) Continuous injection, and (b) intermittent injection.

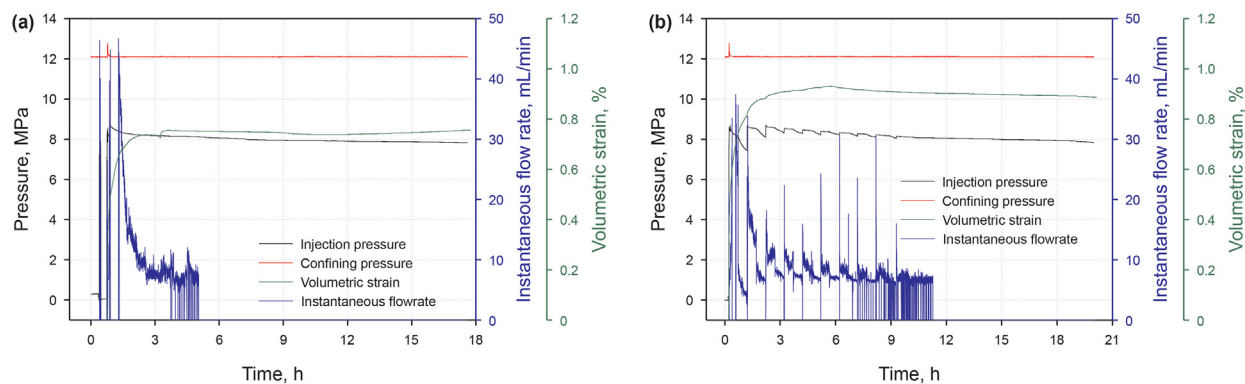


Fig. 5. Experimental results of continuous/intermittent injection of ScCO₂ into the sample. (a) Continuous injection, and (b) intermittent injection.

presented several fluctuations, however, unlike the SubCO₂ injection, the injection time became longer, and the instantaneous flow rate reduced to zero when the injection time was equal to 5 h. The time when the maximum volumetric swelling strain is reached was earlier than the time when the instantaneous flow dropped to zero. Although the adsorption swelling deformation reduced the injectivity of CO₂, the injection process was extended if the CO₂ injection pressure was high enough. The total injection volume of CO₂ was 2622.85 mL, and the net injection volume of CO₂ was 2323.45 mL under the continuous injection mode after deducting the free space volume.

For Fig. 5b, the instantaneous flow rate of CO₂ has not decreased to zero during the process of ten injection/stop cycles, which was different from the continuous ScCO₂ injection mode. The intermittent injection mode largely lengthened the injection process, manifesting in that the injection time of intermittent ScCO₂ injection was 2.25 times that of the continuous ScCO₂ injection. Additionally, although the intermittent injection mode was widely adopted, the effects of SubCO₂ and ScCO₂ were different. For the intermittent SubCO₂ injection, the sample seemed almost impossible to be injected at the seventh injection/stop cycle; however, for the intermittent SubCO₂ injection, the injection process continued for 1.96 h after ten injection/stop cycles. Through the intermittent injection mode, the total injection volume of CO₂ was 5297.45 mL, the net injection volume of CO₂ was 4998.05 mL after deducting the free space volume. The net injection volume of ScCO₂ was improved by 115.11% by intermittent injection mode, compared with the continuous injection mode.

3.1.3. Mechanism of intermittent injection to enhance CO₂ injectivity

The above analysis shows that the CO₂ pressure is gradually decreased when CO₂ injection is ceased. For the CO₂-ECBM pilot test, when the injection volume of CO₂ reaches the maximum storage capacity of the coal reservoir, the free CO₂ is regarded as the gas source of the coal reservoir when the injection well is closed. As the CO₂ adsorption continued, the volume of free CO₂ in the coal reservoir was reduced gradually, causing a decrease in reservoir pressure. The CO₂ was re-injected into the coal reservoir when the injection well was opened, hereof, the pressure difference appeared, which further promoted the adsorption of CO₂. With the increase in the injection/stop cycles, the cumulative injection volume of CO₂ was accreted gradually. In brief, the intermittent injection mode offsets the pressure loss during the shutting-in process of the injection well and results in the increase in CO₂ injectivity for the coal reservoir.

Also, the CO₂ injection time is prolonged through intermittent

injection mode. For the SubCO₂ and ScCO₂, the injection times of intermittent injection were 4.44 and 2.25 times of continuous injection. The extension of the injection process by intermittent injection mode is due to that the permeability of the coal reservoir can be recovered in the injection interruption period, which can be authenticated from the field engineering practices. For instance, Dutta and Zoback (2012) found that the loss of permeability can partially be overcome by intermittent injection for 6 months followed by a soak period. The extended injection process can provide sufficient time for CO₂ to flow and store in the coal matrix thoroughly, causing the improvement of CO₂ injectivity.

All in all, the intermittent injection possesses the following advantages: (1) compensating the loss of reservoir pressure by continuous injection; (2) prolonging the injection process of CO₂; (3) promoting the permeability recovery during the shut-in period. It enlightens us that the intermittent CO₂ injection is a potential and effective enhanced CO₂ injectivity method.

3.2. N₂ displacing CO₂ method

3.2.1. Variation of gas components during N₂ displacing CO₂

Fig. 6 shows the variations of gas component content during the processes of the primary SubCO₂ injection, N₂ displacing SubCO₂ and the second SubCO₂ injection. When the experimental time was 1.47 h, SubCO₂ at an injection pressure of 4 MPa was introduced into the whole device, CO₂ content monitored at the end of the device was above 99%, but it did not reach 100%, which is because that the CO₂ concentration in the purchased gas cylinder is 99.9% and a minor error existed. Then the CO₂ adsorption process started and continued for 24.67 h, which can be considered that the adsorption equilibrium has been reached (Dutta and Zoback, 2012). When the experiment proceeded to 26.14 h, N₂ was injected into the sample cell at an injection pressure of 4 MPa, and the displacement process began. The gas composition changed periodically, i.e., (1) When experimental time fell in the range of 26.14–29.71 h, the CO₂ content rapidly reduced from 99.37% to 8.13% and the N₂ content sharply raised from 0.63% to 91.87%, which indicated that the injection of N₂ start to displace the pre-adsorbed CO₂. (2) When the experimental time ranged from 29.71 h to 37.24 h, similarly, the CO₂ content decreased (from 8.13% to 1.55%) and the N₂ content increased (from 91.87% to 98.45%), while the whole trend was quite slow. During the time segment of 37.24–45.03 h, the gas content changed slightly and essentially tended to be constant. Subsequently, the SubCO₂ was re-injected into the coal sample at 45.74 h, resulting in the re-increase of CO₂ content and the re-decrease of N₂ content.

The changes in gas component content in the processes of the

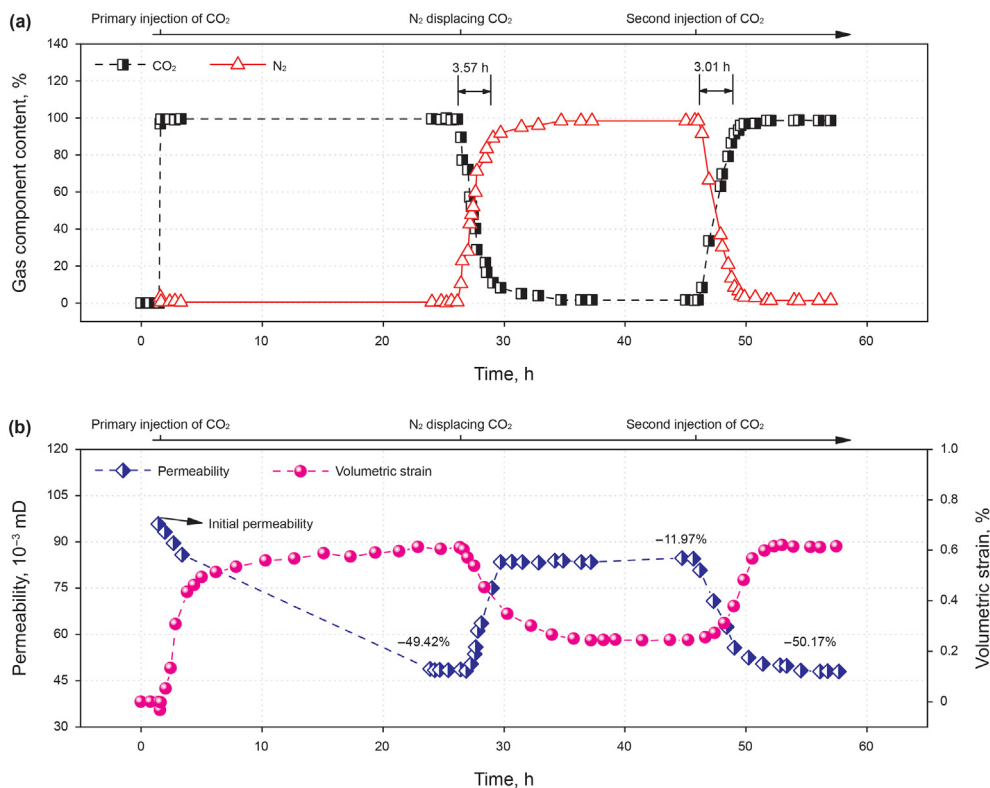


Fig. 6. Gas component content, permeability and volumetric strain of coal in the processes of the primary SubCO₂ injection, N₂ displacing SubCO₂, and the second SubCO₂ injection.

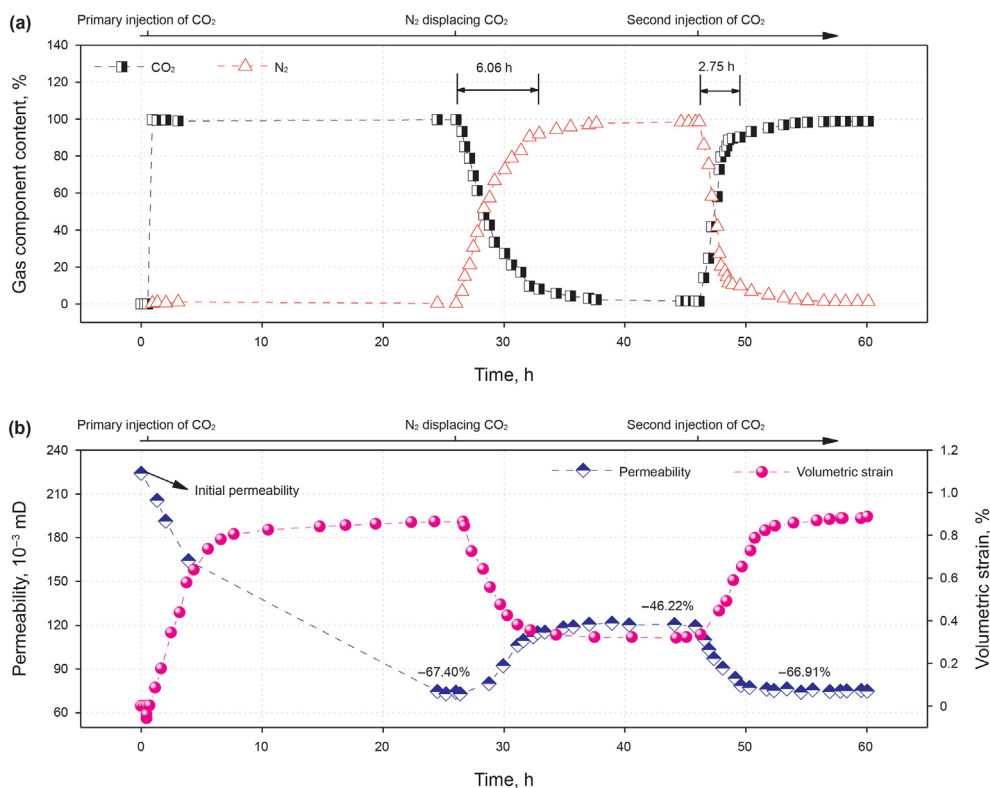


Fig. 7. Gas component content, permeability and volumetric strain of coal in the processes of the primary ScCO₂ injection, N₂ displacing ScCO₂ and the second ScCO₂ injection.

primary ScCO₂ injection, N₂ displacing ScCO₂ and the second ScCO₂ injection are shown in Fig. 7. The ScCO₂ was injected into the sample cell at the pressure of 8 MPa when the experimental time was equal to 0.55 h, the CO₂ content reached the maximum value, and then N₂ was injected at 8 MPa to displace ScCO₂. The CO₂ content decreased and the N₂ content increased, the gas content changed expeditiously at the experimental time of 26.06–30.63 h and changed slowly at the experimental time of 30.63–37.64 h, showing a phased evolution law. Thereafter, the ScCO₂ was re-injected into the coal sample, and the CO₂ content reached the maximum again.

The experiment in this section shows that in the processes of CO₂ injection, N₂ displacing CO₂ and second CO₂ injection, the evolutionary trends of gas content are analogous, regardless of the phase state of CO₂. Nevertheless, there exists differences in the displacement time and displacement efficiency. The displacement time was 3.57 h when the CO₂ content decreased to 10% for N₂ displacing SubCO₂, and at the same conditions, the displacement time reached 5.55 h for N₂ displacing ScCO₂, indicating that it was more difficult for N₂ displacing ScCO₂. For the second SubCO₂ injection, the duration when the CO₂ content increased to 90% was 3.01 h, and at the same conditions, the duration reduced to 2.75 h for the second ScCO₂ injection.

The gas adsorption capacity follows the sequence: ScCO₂ > SubCO₂ > N₂, therefore, ScCO₂ will be the winner during the competitive adsorption process, which manifests in the phenomena that ScCO₂ molecules rapidly supersedes N₂ molecules and occupy their adsorption sites with the higher efficiency.

Fractures in the coal are mainly seepage passages, and pores in the coal are the adsorption and diffusion sites for gas (Chen et al., 2021). Comparing with the displacement effect of CH₄ by CO₂, N₂ displacing CO₂ is more close to the flushing process. Before the injection of N₂, the CO₂ adsorption is in equilibrium, and the CO₂ pressure in the fractures is equal to that in the coal matrix when N₂ is injected into the coal, the total gas pressure in the fractures is composed of CO₂ partial pressure (p_1) and N₂ partial pressure (p_2). With the continuous injection of N₂, p_2 increases and p_1 decreases. When $p_2 > p_1$, CO₂ is flushed out of the coal under the action of gas pressure difference, accompanied by the approximate linear reduction of CO₂ content. When the CO₂ content in the fractures is less than that in the coal matrix, CO₂ is desorbed and diffused from the coal matrix to the fractures under the action of gas concentration difference, and finally, N₂ displacing CO₂ is completed. In this stage, the CO₂ content is reduced slowly.

Figs. 6 and 7 also illustrate the volumetric deformation of coal occurred in the processes of the primary CO₂ injection, N₂ displacing CO₂ and second CO₂ injection. For Fig. 6, during the primary SubCO₂ injection, the volumetric strain reduced instantaneously to -0.03%, which reflects the global compression effect of CO₂ for the coal body. The swelling strain induced by the SubCO₂ adsorption increased with the increase of experimental time, the maximum swelling strain reached 0.59%. During the N₂ displacing SubCO₂, the desorption of CO₂ caused the matrix shrinking of coal (Niu et al., 2017a), although N₂ adsorption also resulted in matrix swelling, the ultimate swelling strain was still reduced to 0.25%, which was greater than the initial strain value. And during the second SubCO₂ injection, the coal matrix swelled again and the maximum swelling strain reached 0.61%, nearly equaling to the swelling strain induced by the primary SubCO₂ injection.

For Fig. 7, the changes of the volumetric strain of coal occurred in the processes of the primary ScCO₂ injection, N₂ displacing ScCO₂ and the second ScCO₂ injection were similar to those of SubCO₂, but the values of swelling strain varied. During the primary ScCO₂ injection, the maximum swelling strain of the coal was 0.84%, which was higher than that induced by SubCO₂ adsorption. When N₂ was

injected into the coal saturated with ScCO₂, the coal shrank and the volumetric strain decreased to 0.32%. Similarly, when ScCO₂ was injected into the coal again, the volumetric strain re-increased to 0.87%. The deformation of the coal body is closely related to the adsorption/desorption of gas (SubCO₂/ScCO₂ and N₂), and the difference in adsorption capacity of various types of gases is the fundamental cause of the dynamic changes of the swelling strain of the coal during the primary CO₂ injection, N₂ displacing CO₂ and the second CO₂ injection.

3.2.2. Variation of permeability of coal during N₂ displacing CO₂

Generally, the gas flow in coal is laminar (Chao et al., 2019; Gao et al., 2017; Sharma et al., 2017). Thus, Darcy's law was used to calculate the permeability of the sample from the measured steady-state flow rates (Ranjith et al., 2011):

$$k = \frac{2Q\mu p_0 L}{A(p_1^2 - p_0^2)} \quad (1)$$

where Q , μ , p_i , and p_0 are the gas flow rate, the gas viscosity, the inlet gas pressure and outlet gas pressure, respectively; A and L are the cross-sectional area and the length of the core sample, respectively; k is the permeability of the sample. In the processes of the primary CO₂ injection and the second CO₂ injection processes, the CO₂ is adopted to calculate the permeability of coal, while, the N₂ is used to measure the permeability of the coal in the N₂ displacing CO₂ process.

The permeability evolution of the coal is what we are most concerned about in the processes of the primary CO₂ injection, N₂ displacing CO₂ and the second CO₂ injection. Corresponding results are shown in Figs. 6 and 7. As can be seen from Fig. 6, the initial coal permeability at an injection pressure of 4 MPa was 95.78×10^{-3} mD when the experimental time exceeded 1.47 h. When the SubCO₂ was continuously injected in coal, the coal permeability was gradually attenuated to 48.45×10^{-3} mD, with the decrease percentage of 49.42%; when N₂ was injected into the coal saturated with SubCO₂, the coal permeability was gradually recovered to 84.70×10^{-3} mD at the experimental time of 44.72 h, which was promoted by 74.82% compared with the permeability in the primary SubCO₂ injection. After that, the second SubCO₂ injection induced a decrease in coal permeability once again, the minimum value was 48.05×10^{-3} mD, and the permeability reduced by 50.17% compared with the initial value.

For Fig. 7, the permeability of the coal under conditions of the primary/second ScCO₂ injection and N₂ displacing ScCO₂ also showed the same variation law. The initial permeability of the coal at 4 MPa injection pressure was 224.18×10^{-3} mD, the primary ScCO₂ injection lowered the permeability to 73.09×10^{-3} mD with a reduction of 67.40%; the N₂ displacing ScCO₂ recovered the permeability to 120.56×10^{-3} mD, and the coal permeability was increased by 64.95% compared with the permeability at the primary ScCO₂ injection; however, the coal permeability reduced to 74.18×10^{-3} mD with the second ScCO₂ injection, approaching to that in the primary ScCO₂ injection.

The simulation experiments indicated that N₂ displacing CO₂ indeed promoted the recovery of permeability in the coal, nevertheless, its permeability recovery rate (64.95%) was lower than that of N₂ displacing SubCO₂ (74.82%), which may be related to the various difficulties of N₂ displacing CO₂ at different phase states. The permeability enhancement effect of coal seams by N₂ displacing CO₂ has been reported in previous research, e.g., Zhang et al. considered that the permeability was improved by 73.51%–106.61% after N₂ displacement at a gas pressure of 5–14 MPa (Zhang et al., 2018); for the CO₂-ECBM field test in Ishikari basin, the injection of N₂ increased the daily injection rate of CO₂ more than four times

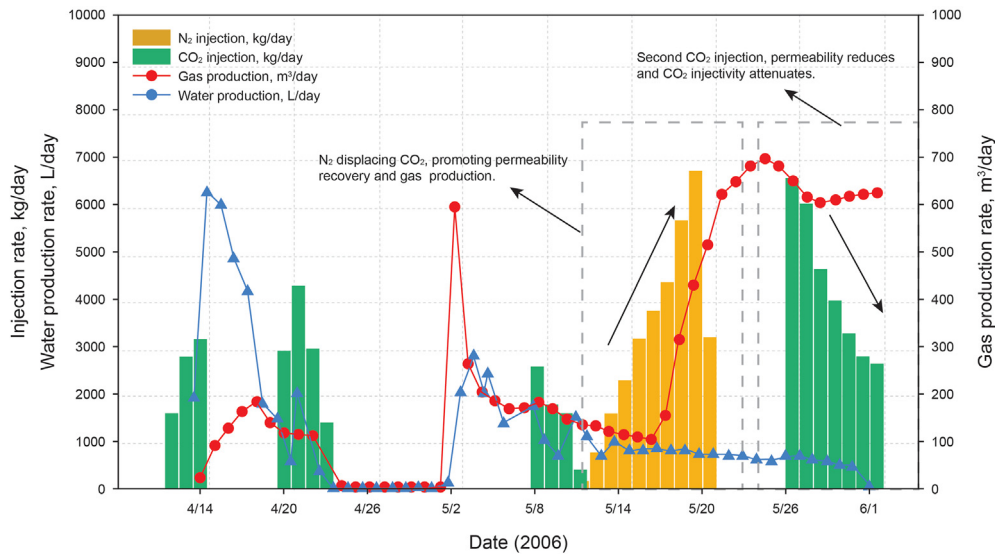


Fig. 8. Production and injection curves from the pilot test of CO₂-ECBM (Fujioka et al., 2010).

(Fujioka et al., 2010) (Fig. 8), however, this positive effect was only temporary, once the CO₂ was reinjected, the CO₂ injectivity still reduced slowly. This can be verified by simulation experiments in this section, i.e., the second CO₂ injection has conversely driven N₂ out of the sample, and decreased the permeability to the level of the primary CO₂ injection.

3.2.3. Mechanism of N₂ displacing CO₂ to enhance CO₂ injectivity

In the CO₂-ECBM field project, there exists an interaction of three gases after N₂ injection, i.e., CO₂, CH₄ and N₂. Because the adsorption capacities of coal for different gases are quite different, the gas composition in the coal continuously changes. Based on the kinetic equation of multicomponent gas adsorption, this process can be expressed as (Ji, 2015):

$$\frac{dx}{dt} = \eta_v \theta_{N_2} \theta_{CH_4} \theta_{CO_2} = \frac{\eta_v b_{N_2} b_{CH_4} b_{CO_2} p_{N_2} p_{CH_4} p_{CO_2}}{(1 + b_{N_2} p_{N_2} + b_{CH_4} p_{CH_4} + b_{CO_2} p_{CO_2})^2} \quad (2)$$

where dx/dt is the gas displacement/desorption rate; η_v is the gas displacement/desorption constant; θ_{N_2} , θ_{CH_4} and θ_{CO_2} are respectively the coverage rate of N₂, CH₄ and CO₂ at adsorption equilibrium; b_{N_2} , b_{CH_4} and b_{CO_2} are the adsorption constants of N₂, CH₄ and CO₂, respectively; p_{N_2} , p_{CH_4} and p_{CO_2} are the partial pressure of N₂, CH₄ and CO₂ respectively.

Because the adsorption capacity of coal for N₂ is lower than those of CH₄ and CO₂, Equation (2) can be simplified by using the inequality of $b_{N_2} p_{N_2} \ll 1 + b_{CH_4} p_{CH_4} + b_{CO_2} p_{CO_2}$ as:

$$\frac{dx}{dt} = \frac{\eta_v b_{N_2} b_{CH_4} b_{CO_2} p_{N_2} p_{CH_4} p_{CO_2}}{(1 + b_{CH_4} p_{CH_4} + b_{CO_2} p_{CO_2})^2} \quad (3)$$

Equation (3) shows that the N₂ injection promotes the partial pressure of N₂ and enhances the desorption rate of CH₄ and CO₂. The desorption process is accompanied by the matrix shrinking effect, which results in the expansion of narrow fractures or reopening of closed fractures, and thus the permeability of the coal is promoted. The ameliorated permeability accelerates the entrance of CO₂ into coal pores by seepage-diffusion-adsorption effects, which is in favor of CO₂-ECBM. However, during the process of N₂ displacing CO₂, the discharge of the desorption CO₂ will reduce the

CO₂ storage capacity, which counts against the purpose of CO₂ geological sequestration.

3.3. Pre-fracturing method

3.3.1. Influence of effective stress on the permeability of coals before/after pre-fracturing

Generally, the coal permeability decreases under loading conditions and increases under unloading conditions, but it is almost impossible to completely recover the permeability to the initial level. According to the description by Meng and Li (2013), the irreversible permeability loss rate (R_{ip}) under a loading/unloading cycle is defined as:

$$R_{ip} = \frac{k_0 - k'_0}{k_0} \times 100\% \quad (4)$$

where R_{ip} is the irreversible permeability loss rate, k_0 represents the initial permeability of the sample; k'_0 represents the permeability of the sample after a loading/unloading cycle. Generally, the stress sensitivity coefficient (α_k) is always adopted to characterize the stress sensitivity of coal reservoir (Geng et al., 2017), and defined as follows:

$$\alpha_k = -\frac{1}{k_0} \frac{\partial k}{\partial \sigma} \quad (5)$$

where k represents the permeability; k_0 represents the initial permeability; σ represents the confining pressure. Moreover, the permeability of a reservoir is verified to be highly susceptible to stress and has been confirmed by scholars in experiments to generally decrease exponentially with the increase in effective stress (Geng et al., 2017; Wang et al., 2018b; Zou et al., 2016):

$$k = k_0 e^{-3C_f(\sigma_h - \sigma_{h0})} \quad (6)$$

where C_f is the fracture compression coefficient; σ_h is the external stress and σ_{h0} is its initial value.

Fig. 9 shows the measured permeability of coal samples before/after pre-fracturing, every measurement was conducted twice to ensure data accuracy. For the coal sample before pre-fracturing, the increase in the effective stress decreased its permeability and the

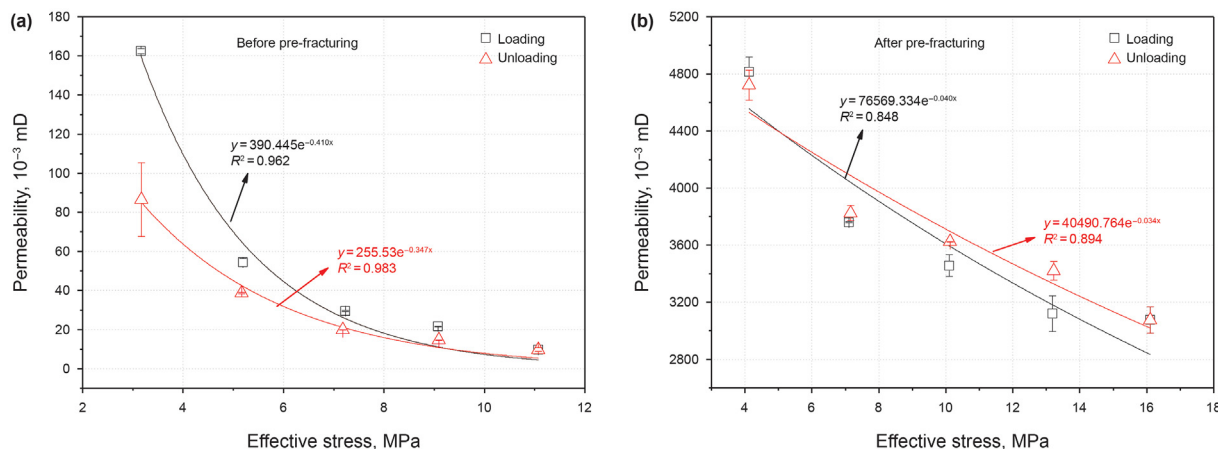


Fig. 9. Relationship between permeability and effective stress of samples before and after pre-fracturing.

decrease in the effective stress recovers its permeability, which was consistent with previous experimental results (Jiang et al., 2017; Ye et al., 2017; Zou et al., 2016). However, the permeability curve of the coal after pre-fracturing showed a huge difference, the curve almost changed to be straight-line though it still can be fitted by the exponential function (Eq. (6)) with higher R^2 . Additionally, in some cases, the permeability at the unloading stage was larger than that at the loading stage. The pre-fracturing treatment for samples affected its evolution of permeability.

For the loading stage, the permeability of the coal before pre-fracturing ranged from 109.331×10^{-3} mD to 10.223×10^{-3} mD, and that of the coal after pre-fracturing fell in the range of 4812.750×10^{-3} to 3076.614×10^{-3} mD; for the unloading stage, the coal permeability ranged from 9.785×10^{-3} mD to 86.512×10^{-3} mD before pre-fracturing, and 3076.614×10^{-3} mD and 4720.176×10^{-3} mD after pre-fracturing. It can be seen that the pre-fracturing treatment for the coal improved the permeability by 1–2 orders of magnitude. Although previous scholars have also confirmed that the permeability increases after pre-fracturing, the increasing magnitude was variously reported in different pieces of literature, e.g., Kumar et al. demonstrated that when the monolayer proppant was sandwiched in the coal fracture, the He permeability may be as high as ~10 fold than the initial value (Kumar et al., 2015); Wu et al. considered that the permeability of the proppant supported fracture was 2–3 orders of magnitude higher than that of the original sample; and for shale (Wu et al., 2018), Tan et al. (2017) deemed that the permeability of propped fractures was about a few hundred or even a few thousand times higher than those of natural fractures. The increasing magnitude of permeability depended on the proppant packing pattern, the layer number, particle size and the type of proppant. Generally, the hardness of proppant affects the compressibility of fractures and then influences the permeability of the coal sample; increasing the proppant layer, enlarging the proppant particle size and making the packing pattern sparse can create more space for the flow of gas, and thus, the permeability enhancement effect induced by pre-fracturing will be superior. In conclusion, there is no doubt that the pre-fracturing method is capable of dramatically enhancing the permeability of coal reservoirs.

The irreversible permeability loss rate and average stress sensitivity coefficient were calculated according to Eqs. (4) and (5). The average irreversible permeability loss rate of coal samples before and after pre-fracturing were 46.775% and 1.925%, respectively, and the pre-fracturing has reduced the irreversible permeability loss rate by 95.885%; the average stress sensitivity coefficient

of the sample before and after pre-fracturing were 0.117 and 0.045, respectively, and the pre-fracturing has lowered the average stress sensitivity coefficient by 61.538%. The irreversible permeability loss rate and the average stress sensitivity coefficient of coal samples after pre-fracturing were largely weakened, and it can be deduced that the permeability of the coal is insensitive to the effective stress after pre-fracturing treatment. The main reason can be attributed to that the stiffness of quartz sand is far greater than that of the coal matrix. Under the acting of effective stress, the quartz sand is hard to be compressed, it thus supports the fractures in coal and provides adequate seepage spaces for gas. Consequently, the high-permeability and low-stress sensitivity properties of the ameliorated coal reservoirs are highly favorable for CO₂ injection.

3.3.2. Influence of ScCO₂ adsorption on the permeability of coals before/after pre-fracturing

The adsorption swelling behavior affects the permeability in coal (Jia et al., 2018; Niu et al., 2017b; Wei et al., 2019), the seepage paths are cramped and the permeability is reduced through the physicochemical reaction between the CO₂ and coal. Based on the permeability pre and post the CO₂ adsorption, the parameter of permeability loss rate due to adsorption was defined as (Niu et al., 2019b):

$$R_a = \frac{k_0 - k_a}{k_0} \times 100\% \tag{7}$$

where k_0 represents the initial permeability of the coal sample, and k_a is the permeability of the coal sample when the CO₂ adsorption equilibrium state is reached.

The permeability changes of samples without/with pre-fracturing treatment before and after ScCO₂ adsorption are shown in Fig. 10. The permeability after ScCO₂ adsorption was lower than that before ScCO₂ adsorption, indicating that the adsorption swelling narrows the fractures and reduced the coal permeability. Nevertheless, the permeability of pre-fractured core sample did not show apparent permeability attenuation after ScCO₂ adsorption, and even some permeability values were larger than that of the sample without ScCO₂ adsorption. Based on Eq (7), the permeability loss rate due to adsorption of coal samples before and after pre-fracturing was obtained (Fig. 10). The pre-fracturing also decreased the permeability loss rate due to adsorption of the coal sample, manifesting in the fact that the average permeability loss rate due to adsorption of the pre-fractured core sample was lowered by 96.297% of the primary sample.

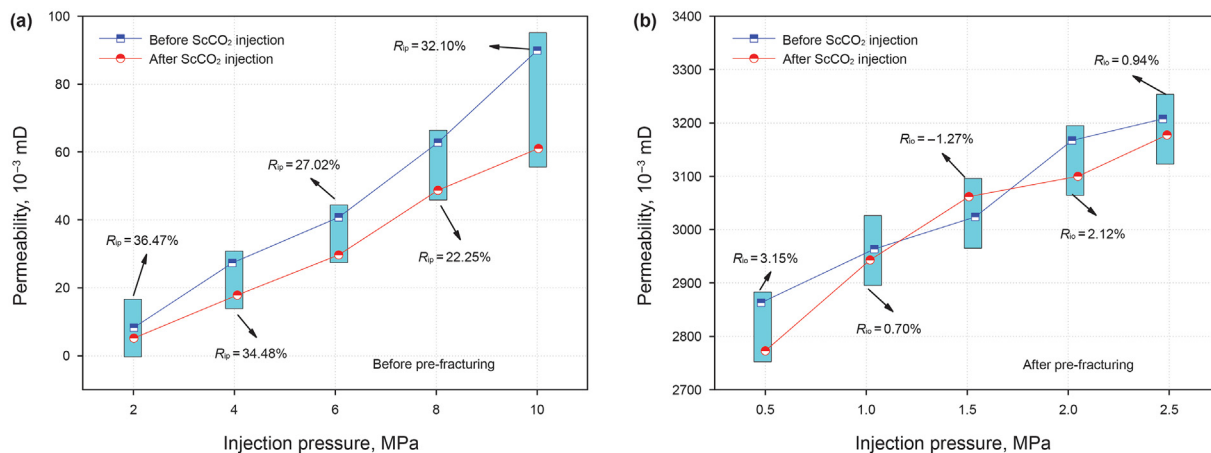


Fig. 10. The permeability loss rate due to adsorption of coal samples before and after pre-fracturing.

The decrease of permeability loss rate due to adsorption indicates that the permeability of the coal sample after pre-fracturing is also insensitive to ScCO₂ adsorption. This is because the quartz sand with higher stiffness can withstand the wall of fractures, increasing the difficulty for swelling deformation developing to the interior of fractures. Simultaneously, even if the fracture structure may also be influenced by adsorption swelling, this effect can be negligible due to the sharp increase in the permeability of coal after pre-fracturing.

3.3.3. Mechanism of pre-fracturing to enhance CO₂ injectivity

As described in the literature (Niu et al., 2020b), for CO₂-ECBM, CO₂ flows from the injection well, tectonic fractures or bedding plane fractures, cleats, and finally is adsorbed in the multiscale pores of coal. For structure undeveloped coal seams (e.g., coal seams in the Qinshui Basin), the bedding plane fractures and cleats all control the CO₂ injection because it is a continuous process (Perera et al., 2013). Research indicated that the bedding plane

seepage is mainly controlled by effective stress, contrarily, the cleat seepage is mainly affected by the adsorption of ScCO₂ (Niu et al., 2018).

The diagrammatic sketch of coal seams modified by fracturing is shown in Fig. 11. After fracturing, abundant crisscross cracks are induced in coal reservoirs. The cracks in the PBD enlarge the seepage space of CO₂, and cracks in the VBD link the cleats and increase its connectivity, then the global permeability of the coal seam is largely improved. When water, sand and additives are pumped under pressure into the coal seam, the fracturing sands are filled in the primary fractures and epigenetic cracks. As discussed above, the permeability stress sensitivity in the PBD and the permeability loss rate due to adsorption of fractures in the VBD all drop sharply. This means that the permeability of coal seams after pre-fracturing is independent of the effective stress and ScCO₂ adsorption, as the two factors are the main controlling factors for the permeability, we thus can consider the CO₂ injectivity is promoted evidently by the pre-fracturing method.

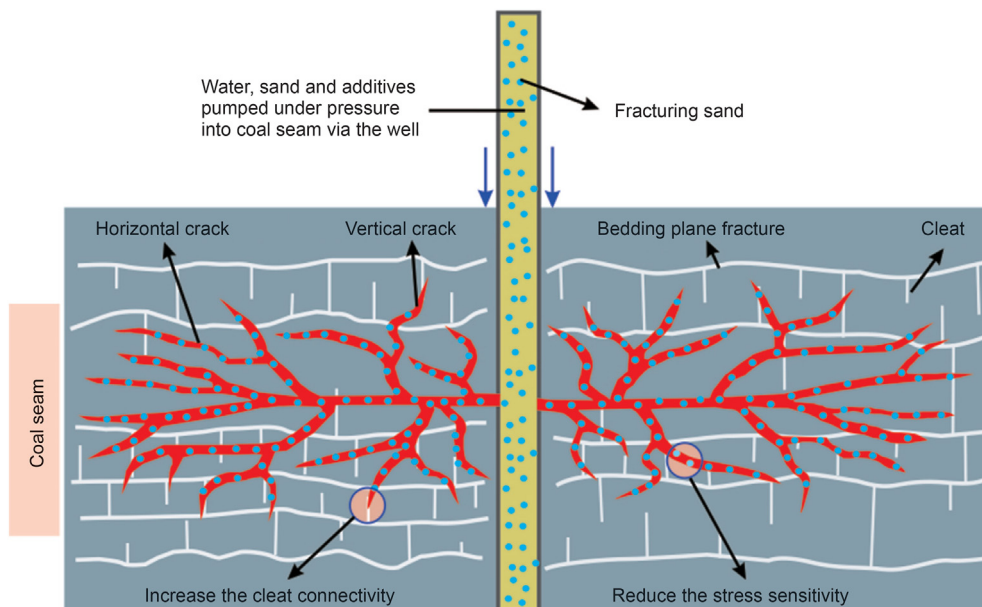


Fig. 11. The diagrammatic sketch of enhancing CO₂ injectivity for coal reservoir by pre-fracturing.

3.4. Comparisons of engineering measures for enhancing CO₂ injectivity during CO₂-ECBM

Intermittent CO₂ injection, N₂ displacing CO₂ and pre-fracturing are the common engineering measures to improve the CO₂ injectivity for the coal reservoir. All of them can amplify the CO₂ injection efficiency and injection capacity for CO₂-ECBM, however, the improvement level and effect are various. Pre-fracturing is the most efficient measure because it can thoroughly reconstruct the structure of the coal reservoir, which not only improves the three-dimensional permeability immensely but also makes it independent of the influences of effective stress and ScCO₂ adsorption. The effect of the intermittent injection is moderate because it can extend the injection process and remedy the reservoir pressure loss during CO₂-ECBM, while for an extremely low permeability coal seam, this method may be inefficient. The effect of N₂ displacing CO₂ is limited, although the permeability will recover to a certain extent, it will attenuate again when CO₂ is reinjected, also, it will reduce the sequestration capacity of CO₂ and is averse to the geological CO₂ sequestration.

The appropriate enhanced injectivity measures should be chosen according to the physical and geological conditions of coal reservoirs. For coal seams with high porosity and high permeability, intermittent CO₂ injection or N₂ displacing CO₂ may be adequate; while for coal seams with low permeability developed in complex geological conditions, these engineering measures need to be combined to obtain an excellent CO₂ injection effect.

4. Conclusions

The attenuation of CO₂ injectivity induced by adsorption swelling restricts the successful launch of CO₂-ECBM. In this paper, three commonly-used engineering measures, intermittent CO₂ injection, N₂ displacing CO₂ and pre-fracturing, were simulated by laboratory experiments and their effect and mechanism were clarified and examined. The following conclusions were drawn:

- (1) The phase state can affect the intermittent CO₂ injection process, for the intermittent SubCO₂ injection, the injectivity will reduce to zero within ten injection/stop cycles, while the injection process will last beyond ten injection/stop cycles for intermittent ScCO₂ injection. Compared with the continuous injection mode, the net injection volumes of SubCO₂ and ScCO₂ are improved by 23.80% and 115.11% by using intermittent injection mode. The CO₂ injection time is increased by folds and the reservoir pressure loss is complemented in time through intermittent injection mode, thus, the injectivity of CO₂ is highly improved.
- (2) When CO₂ is injected into coal, the adsorption swelling occurs and gradually reaches the maximum value, and the permeability is reduced by 49.42% for SubCO₂ and 67.40% for ScCO₂. When N₂ is injected into coal containing CO₂, the gas composition of CO₂ decreases and the adsorption swelling recoveries in some degrees, the permeability is increased by 74.82% and 64.95% compared with that in the primary SubCO₂ and ScCO₂ injection. The gas composition of CO₂, the swelling strain and the permeability all return to the values of those in the primary CO₂ injection. N₂ injection promotes the matrix shrinking effect induced by the desorption of CH₄ and CO₂, then the expansion of narrow fractures or reopening of closed fractures causes the increase of CO₂ injectivity.
- (3) After pre-fracturing treatment, the permeability of the coal sample is averagely advanced by 1–2 orders of magnitude, the irreversible permeability loss rate, average stress sensitivity coefficient and permeability loss rate due to adsorption

are averagely lowered by 95.885%, 61.538% and 96.297%, respectively. The permeability of the sample after pre-fracturing is no longer sensitive to the effective stress and ScCO₂ adsorption, which is the reason why the injectivity of CO₂ has been largely promoted.

- (4) The enhanced CO₂ injectivity effects of the intermittent CO₂ injection, N₂ displacing CO₂ and the pre-fracturing are various. For the CO₂-ECBM project, these engineering measures could be selected individually or jointly to improve the CO₂ injectivity, depending on the physical properties and geological conditions of reservoirs.

Acknowledgments

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