

Experimental Study on CH₄ Displacement from Coal Seam Fractured by Liquid CO₂

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ABSTRACT

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In order to improve the gas drainage efficiency of low permeability coal seams, based on the theoretical analysis of liquid CO₂'s phase change permeability improvement, negative temperature damage, extrusion stress damage, and displacement effects on coal seams, this paper researches and develops a process system for liquid CO₂ fracturing coal seam CH₄ displacement, and carries out industrial tests on the 17246 coalface of B6 coal seam in Zhangji Coal Mine. The test results show that, in the injection process, liquid CO₂ first fills in the cracking holes and visible cracks with larger opening degree, then it enters the original weak plane areas, and finally penetrates into the coal matrix pores, the pressure at the orifice of the injection borehole presents fluctuation characteristics; when CO₂ flows into the coal mass cracks, and diffuses into the pores of the coal matrices, it's affected by flow resistance and adsorption, and the coal mass permeability in the fracture zone, plastic zone and elastic zone decreases in turn; the effective influence radius after liquid CO₂ is injected in the coal seams for permeability improvement displacement is more than 30 meters. The average single-hole gas concentration is increased by 2.15 times, the average single-hole pure gas drainage flow volume is increased by 2.27 times, and the average single-hole pure gas flow attenuation coefficient is reduced by 80.76%, which improves the gas drainage efficiency.

1. INTRODUCTION

More than 37% of the coal mines in China are high-gas coal mines or coal-gas outburst mines, 95% of which are low permeability coal seams, and the gas drainage in these coal seams is quite difficult [1-3]. The gas drainage technology in low permeability coal seams is still a major technical issue in the field of coal mine safety [4, 5]. Domestic and foreign scholars have done a lot of research on gas drainage technologies such as hydrofracturing [6-8], protective seam pressure relief [9, 10], deep borehole pre-fracturing and pressure relief [11], and conventional dense drilling [12], these research results are playing an important role in gas drainage in low permeability coal seams in China, but they still have certain limitations in terms of application process and applicability [13, 14]. CO₂ fracturing coal seam CH₄ displacement technology has received wide attention as a new type of gas drainage technology, it can not only achieve the purpose of geological storage of CO₂, but also promote the free analysis of the adsorbed gas, which increases the drainage efficiency of coal seam gas resources, improves the production condition of the coal mine, and prevents direct gas emission and global atmospheric environment pollution [15, 16]. Li et al. [17] applied a self-developed MCQ-II coal seam gas displacement device and found that: raising the injection temperature of the displacement gas can displace more CH₄ gas and store more CO₂ gas within per unit time; Liu et al.

combined with acoustic emission monitoring technology and CT scanning technology to analyze the law of crack propagation during supercritical CO₂ shale fracturing process, and they found that the permeability after shale fracturing had been increased by 3~4 orders of magnitude than that before fracturing; Ao et al. [18] carried out deformation test on shale adsorbed CO₂ of different pressures under constant temperature conditions, and explored the deformation law of shale adsorbed CO₂; Wu et al. [19] used a self-made triaxial adsorption-desorption test device to carry out the experimental study of coal seam CO₂ injection CH₄ displacement. However, at this stage, the CO₂ cracking coal seam flooding CH₄ technology is still in the feasibility and demonstration research stage in all countries of the world, there is few researches on the equipment, fracturing technology, and effective influence radius and drainage effect of liquid CO₂ fracturing coal seam CH₄ displacement technology.

This paper theoretically analyzes the permeability improvement and displacement effects of liquid CO₂ on the coal seams, it researches and develops techniques and equipment for the injection system, and conducts an industrial experiment in the B6 coal seam in Zhangji Coal Mine. The drilling construction parameters and borehole arrangement are designed. The key parameters in the injection process are recorded and analyzed. The gas drainage effect is analyzed by gas drainage concentration, pure flow volume, flow attenuation coefficient and effective influence radius.

2. THEORETICAL ANALYSIS

2.1 Increase the role of permeability

Liquid CO₂ (-35 °C) is a low temperature fluid with characteristics of low viscosity, low resistance, acidifying plugging removal, and phase change pressurization, etc. The permeability improvement effect of liquid CO₂ on the coal seams mainly includes phase change permeability improvement, negative temperature damage and extrusion stress damage [20], the coal mass permeability can be improved by injecting liquid CO₂ through coal seams. The phase change permeability improvement means that, after the liquid CO₂ contacts with the coal mass, during the heat exchange process, the pressure of liquid CO₂ temperature-rise phase change increases, under the action of stress, it forces the original cracks on of the coal mass to expand and extent, and generate new cracks. The temperature of liquid CO₂ is lower than that of the coal seams, and the negative temperature damage means that, after the liquid CO₂ contacts with the coal mass, the coal matrix skeleton is forced to shrink under the action of low temperature, so that the original cracks are expanded further, when the shrinkage stress is greater than the tensile stress of the coal rock, the internal structure of the coal rock body is destroyed, and new cracks are generated. The coal mass contains a certain amount of water, and the extrusion stress damage means that, after the liquid CO₂ contacts with the coal mass, it causes the water in the coal mass to freeze and expand, resulting in coal mass pore reorganization and crack expansion and extension.

2.2 Displacement effect

The coal seam is a dual medium body composed of a network of cracks and coal matrices, 80 % to 90 % of the CH₄ in the coal mass exists in an adsorbed state in the coal matrices, and the remaining CH₄ is stored in a coal mass crack network in a free state [21]. When liquid CO₂ of a certain pressure is injected into the coal mass saturated with CH₄, since the adsorption capacity of CO₂ gas component is stronger than that of CH₄, and the desorption capacity of CH₄ is stronger than that of CO₂, permeation and diffusion occur in the coal mass, and competitive adsorption occurs on the surface of coal mass. Under the combined action of permeation, diffusion and adsorption-desorption, the adsorbed CH₄ in the coal seams is replaced and driven out [22], and the displacement process of the two gas components in the coal mass is shown in Figure 1.

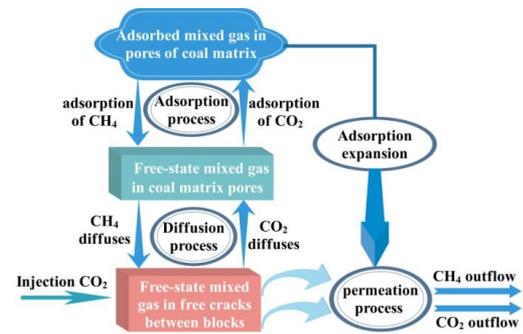


Figure 1. Displacement process of coal components

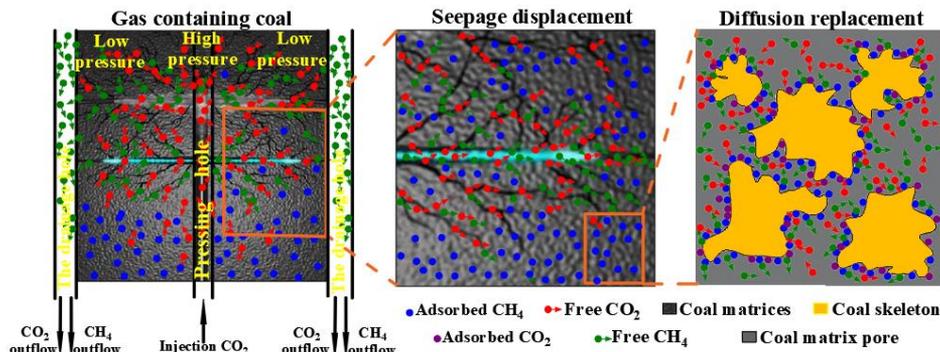


Figure 2. Schematic diagram of coal seam liquid CO₂ injection gas flow displacement

A schematic diagram of coal seam liquid CO₂ injection gas flow displacement is shown as Figure 2: With the injection of liquid CO₂, the pressure difference and volume fraction difference at both ends of the fracture channel are increased, after heating and phase change, CO₂ diffuses and permeates into the interior of the coal matrices, with its strong adsorption capacity, it replaces the CH₄ on the surface of the coal mass skeleton, and under the action of concentration gradient, the free gas diffuses into the cracks of the coal mass, and flows into the drainage holes along the fracture channels in the coal mass.

3. INJECTION SYSTEM PROCESS

The principle of liquid CO₂ fracturing coal seam CH₄ displacement technology is: with liquid CO₂ as medium, through a low temperature liquid pump, liquid CO₂ is injected into the coal seams with a certain pressure and flow volume,

under the combined action of phase change permeability improvement, negative temperature damage, and diffusion replacement, etc., the desorption and diffusion of gas adsorbed in the coal mass is promoted, and thus improving the gas drainage efficiency in the coal seams.

Based on the above analysis, a CO₂ fracturing coal seam CH₄ displacement system was researched and developed. The injection process system is shown in Figure 3. The system mainly includes a liquid CO₂ supply device, a supercharger, a data acquisition device and high voltage pipelines connecting the above three devices, etc. [23]. The liquid CO₂ supply device is a CPW-2.0 liquid CO₂ storage tank [24]; the supercharger adopts the SBP-3000/300 integrated low-temperature liquid pump, its rated power is 55KW, maximum working pressure is 30MPa, flow volume is 20~3000L/h; the data acquisition device model is AI-3706M, the feed voltage is 24VDC, and the power consumption is 5W; the connecting pipelines are made of mine high-pressure rubber hose of different diameters as needed.

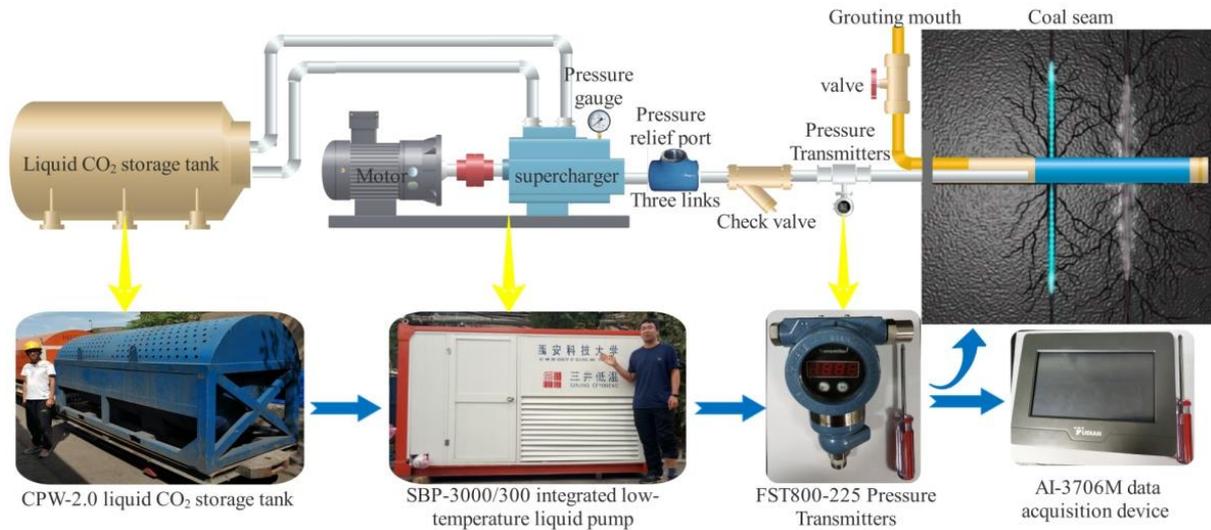


Figure 3. CO₂ permeability improvement CH₄ displacement system

4. FIELD TEST

4.1 Test plan

In the Zhangji Coal Mine of Huainan Mining Group, relevant tests and application research were carried out. The B6 coal seam of Zhangji Coal Mine is a soft and low permeability coal seam which is hard to be drained, its average thickness is 4.5 m, and the average coal seam inclination is 30°, the measured gas pressure is between 1.25~1.69 MPa, the gas content is 5.0~6.64 m³/t, the coal seam firmness coefficient f

is 0.2~0.4, the average attenuation coefficient of borehole gas flow before pressure relief is 0.48 d⁻¹, and the gas permeability coefficient is 0.054 m²/MPa²·d. The proposed technology was applied to carry out CO₂ fracturing permeability improvement on the 17246 coalface, so as to improve the drainage effects of the outburst dangerous coal seam, in this test, vertical to the coal wall, construct two fractured boreholes (7#, 14#) and multiple inspection holes (1#~6#, 8#~13#, 15#~20#), and the arrangement is shown in Figure 4. The borehole diameter is 113 mm, the depth is 140 m, and the maximum inspection influence radius is 30 m.

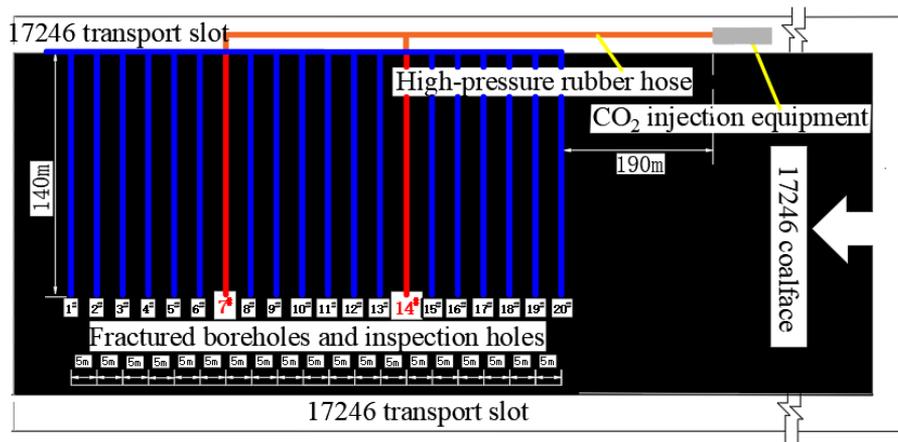


Figure 4. Arrangement of fractured holes and drainage holes

4.2 Test process

The entire liquid CO₂ injection process was carried out for 3 days in the order of “gas injection-liquid injection-pressure holding”. The liquid injection time, accumulated liquid injection volume, maximum pump pressure and orifice pressure of the 7# and 14# fractured holes were tested. The monitoring results are shown in Table 1.

With the construction of the boreholes, the three-directional stress equilibrium state of the coal mass was destroyed, the stress field of the coal mass was redistributed, and there were fracture zone, plastic zone and elastic zone in the coal mass around the boreholes. Liquid CO₂ was injected into the borehole at a constant flow rate. The pressure curves of the 7#

and 14# orifices during the injection process are shown in Figure 5 and Figure 6, and the pressure of the orifice repeatedly rose and fell. It's because when liquid CO₂ was injected, it first filled in the fracture channels of the fracture zone of the coal mass, the CO₂ flow injected into the fractured holes was larger than the CO₂ permeation flow, and the orifice pressure was increased. As the CO₂ volume in the coal mass fracture channels increased continuously, cracks in coal mass fracture zone, plastic zone and elastic zone continued to develop, CO₂ permeation channels increased, and the orifice pressure decreased. The descending sections of the 7# and 14# borehole injection process were linearly fitted and statistically calculated. The pressure dropping rate can be seen in Table 2. It can be seen from Table 2 that the pressure dropping rates of

the first day of the 7# hole were 0.013 and 0.004, respectively, and the pressure dropping rates of the first day of the 14# hole were 0.02, 0.016, and 0.005, respectively, it can be seen that the pressure dropping rate showed a decreasing trend, indicating that the liquid CO₂ first filled in the cracking holes and visible cracks with larger opening degree, then entered the original weak plane areas such as the stratification plane of the coal seams and the cutting cracks, and finally penetrated into the primary micro pores of the coal mass, similarly, same conclusions can be drawn on the second day and the third day; the average pressure dropping rates of the 7# hole injections on the first day, the second day, and the third day were 0.017, 0.011, and 0.01 respectively, and the average pressure dropping rates of the 14# hole injections were 0.014, 0.00937, and 0.0093, respectively, it can be seen that the average pressure dropping rates of 7 # and 14 # holes decreased day by day, indicating that when CO₂ was flowing in the cracks of the coal mass and diffusing in the coal matrix pores, due to the influence of flow resistance and adsorption, the permeability of the coal mass in the fracture zone, plastic zone and elastic zone around the borehole decreased in turn; The pressure dropping rate of the 7# hole of the second day decreased by 35.3 % than that of the first day, and the pressure dropping rate of the 7# hole of the third day decreased by 9.1 % than that of the second day, as for the 14# hole, the pressure dropping rate of the second day decreased by 33.1 % than that of the first day, and the pressure dropping rate of the third day decreased by 0.7 % than that of the second day, indicating that the

permeation and diffusion of liquid CO₂ into the coal mass was the fastest on the first day, followed by the second day, and the third day was the slowest.

Table 1. Key parameters of liquid CO₂ injection process

Fractured Holes	Key Parameters	Time		
		The First Day	The Second Day	The Third Day
7# fractured hole	Liquid injection time/min	18	16	19
	Accumulated liquid injection volume/m ³	1.56	1.75	1.4
	Maximum pump pressure/MPa	3.78	3.26	3.76
	Maximum orifice pressure/MPa	2.28	2.19	2.09
	Liquid injection time/min	9	15	80
14# fractured hole	Accumulated liquid injection volume/m ³	1.45	1.97	1.6
	Maximum pump pressure/MPa	3.67	3.89	3.94
	Maximum orifice pressure/MPa	2.62	2.38	2.28

Table 2. Statistics of pressure dropping rate of 7# and 14# holes during injection process

Drill Number	Pressure Dropping Rate								
	The First Day			The Second Day			The Third Day		
7#	0.013	0.004	---	0.013	0.011	0.008	0.0232	0.005	0.00238
14#	0.02	0.016	0.005	0.0106	0.01	0.0075	0.0196	0.00666	0.0018

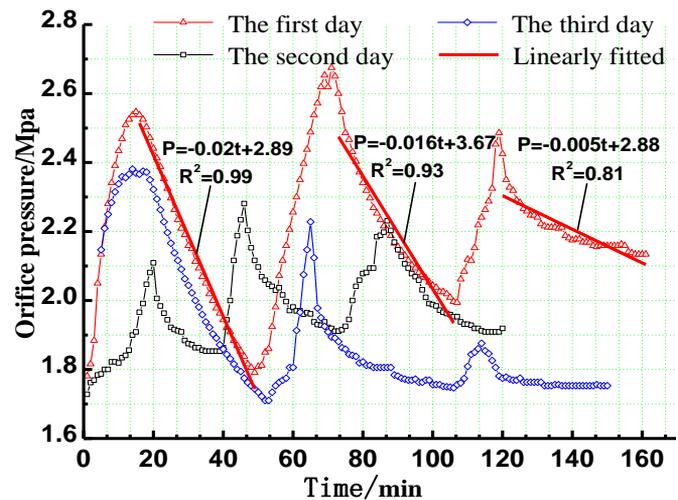
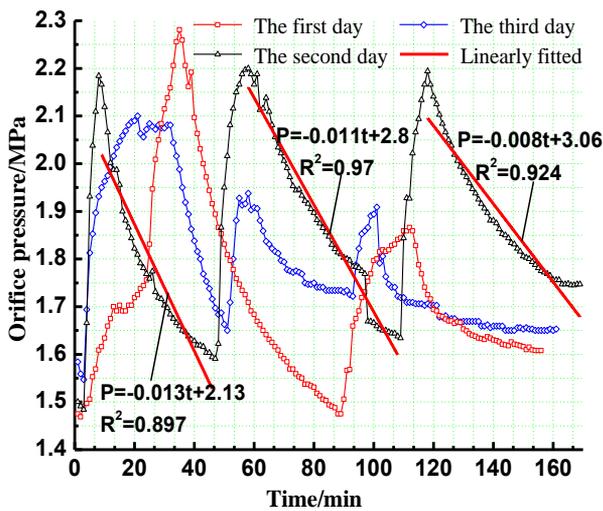


Figure 5. Pressure at orifice of 7# hole during injection process **Figure 6.** Pressure at orifice of 14# hole during injection process

5. EFFECT ANALYSIS

5.1 Influence radius analysis

When the liquid CO₂ injection was completed, after the

pressure of the boreholes dropped below 0.74 MPa, the CO₂ concentration in each inspection holes was detected for 36 days continuously, and the CO₂ concentration of the inspection holes on the first day, 12th day, 24th day, and 36th day is shown in Figure 7.

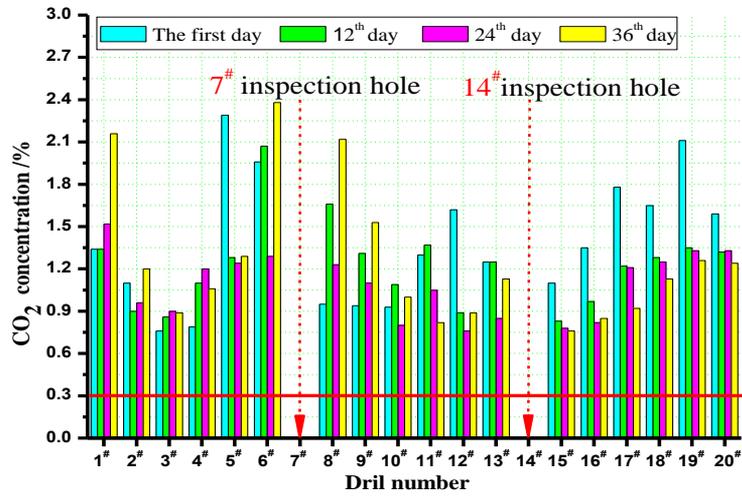


Figure 7. Measured results of CO₂ concentration at inspection holes

From the detection results of CO₂ concentration in the gas drainage holes around the 7# and 14# fractured holes we can know that, in the 1# drainage hole 30 meters away from the 7# fractured hole, the CO₂ concentration fluctuated in a range of 0.78 % to 2.56 % (average was 1.28 %); in the 20# drainage hole 30 meters away from the 14# fractured hole, the CO₂ concentration fluctuated in a range of 0.83 % to 2.59 % (average was 1.37 %); moreover, in the drainage hole 42.5 meters away from the 14# fractured hole, a CO₂ concentration as high as 3.5 % was manually detected by a portable device, this number is much larger than the CO₂ concentration in the drainage hole of original coal seams (0.3 %), indicating that the effective influence radius after the fracturing permeability improvement is more than 30 meters, the coal mass permeability can be improved by injecting liquid CO₂ through coal seams.

5.2 Analysis of gas drainage effect

Gas drainage concentration, pure flow volume and flow attenuation coefficient are important parameters reflecting gas drainage efficiency [4, 25]. When liquid CO₂ injection was completed, after holding the pressure for a period of time, the gas concentration and pure flow volume of the drainage holes were observed for 36 days continuously. The average single-hole CH₄ drainage concentration in the coal seams of the test area and the original area are shown in Figure 8, and pure flow volume in the coal seams of the test area and the original area are shown in Figure 9.

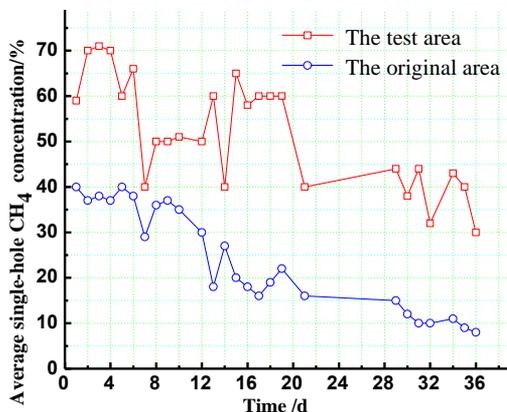


Figure 8. Average single-hole CH₄ drainage concentration

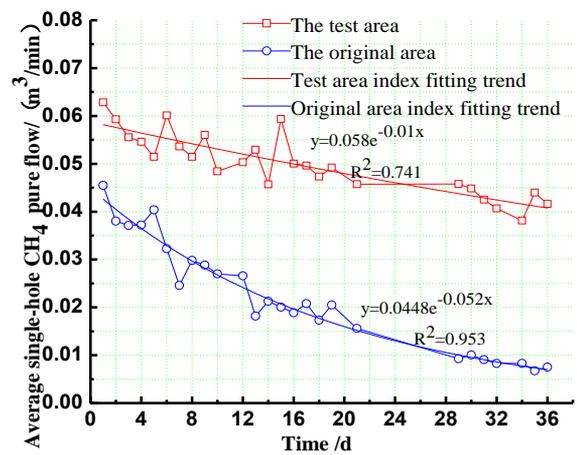


Figure 9. Average single-hole CH₄ drainage pure flow

As can be seen from Figure 8 and Figure 9, the effect of liquid CO₂ permeability improvement coal seam CH₄ displacement is quite significant, mainly reflected in the following three aspects:

(1) The gas drainage concentration increased significantly. It can be seen from Figure 8 that the maximum gas concentration in the test area was 71 %, maximum gas concentration in the original area was 41 %, which was increased by 1.78 times. The average single-hole gas drainage concentration in the test area was 51.96%, and the average single-hole gas drainage concentration in the original area was 24.15 %, which was increased by 2.15 times.

(2) The pure flow of gas drainage increased significantly. It can be seen from Figure 9 that the maximum pure flow volume in the test area was 0.063 m³/min, maximum pure flow volume in the original area was 0.045 m³/min, which was increased by 1.4 times. The average single-hole gas drainage pure flow in the test area was 0.05 m³/min, and the average single-hole gas drainage pure flow in the original area was 0.022 m³/min, which was increased by 2.27 times.

(3) The attenuation coefficient of gas drainage flow was significantly reduced. The gas drainage flow attenuation coefficient can be used as an index to evaluate the difficulty of gas drainage in coal seams. According to the attenuation coefficient of gas drainage flow, the coal mine gas drainage specification (AQ1027-2006) divides the degree of difficulty of drainage into three categories, see table 3.

Table 3. Relationship between attenuation coefficient of gas drainage flow and degree of drainage difficulty

Categories	Easy to Drain Coal Seams	Drainable Coal Seams	Difficult to Drain Coal Seam
The attenuation coefficient of gas drainage flow d ⁻¹	<0.003	0.003~0.05	>0.05
Coal seam permeability coefficient m ³ /Mpa ² ·d	>10	10~0.1	<0.1

Gas drainage flow attenuation coefficient calculation formula [4]:

$$q_t = q_0 e^{-\beta' t} \quad (1)$$

where: q_t represents the gas flow volume of the drainage hole after t -days drainage; q_0 represents the initial gas flow volume of the gas drainage hole; t is the gas drainage time; β' represents the attenuation coefficient of the gas drainage flow of the drainage hole flow volume. According to formula (1), the average single-hole CH₄ drainage pure flow volume was fitted, and the average single-hole gas pure flow attenuation coefficient can be obtained. It can be seen from Figure 9 that the average single-hole gas flow attenuation coefficient in the original area was 0.052, and the average single-hole gas flow attenuation coefficient in the test area was 0.01, which was reduced by 80.76%. After the coal seams are injected with liquid CO₂, the coal seams that were difficult to drain are converted into drainable coal seams.

The above analysis shows that: The liquid CO₂ fracturing coal seam CH₄ displacement technology can effectively increase the CH₄ drainage concentration and pure flow, the maximum CH₄ drainage concentration is increased by 1.78 times, the maximum CH₄ drainage pure flow is increased by 1.4 times, the average single-hole gas drainage concentration was increased by 2.15 times, the drainage pure flow volume was increased by 2.27 times. After the coal seams are injected with liquid CO₂, the average single-hole gas flow attenuation coefficient was reduced by 80.76%, and the coal seam in the test area was changed from difficult extraction to extractable coal seam.

6. CONCLUSION

(1) The permeability improvement effect of liquid CO₂ on coal seams mainly includes phase change permeability improvement, negative temperature damage and extrusion stress damage. When liquid CO₂ of a certain pressure is injected into the coal mass, after heating and phase change, CO₂ diffuses and permeates into the interior of the coal matrices, with its strong adsorption capacity, it replaces and drives out the CH₄ in the coal seam.

(2) In the injection process, liquid CO₂ first fills in the cracking holes and visible cracks with larger opening degree, then enters the original weak plane areas, and finally penetrates into the coal matrix pores, the pressure at the orifice of the injection borehole presents fluctuation characteristics. When CO₂ flows into the coal mass cracks, and diffuses into the pores of the coal matrices, it's affected by flow resistance and adsorption, and the coal mass permeability in the fracture zone, plastic zone and elastic zone decreases in turn.

(3) In this coal seam liquid CO₂ injection permeability improvement and displacement test, the effective influence radius was more than 30 meters, the average single-hole gas drainage concentration was increased by 2.15 times, the

drainage pure flow volume was increased by 2.27 times, and the pure flow attenuation coefficient was reduced by 80.76%. The test indicated that the coal seam liquid CO₂ injection can improve the permeability and displacement of coal seam gas, thus improving the gas drainage effect of the coal seams.

ACKNOWLEDGMENTS

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