Mechanism Exploration and application on Improving Coal Permeability by Heat Treating

Liu Hao1,2, Zhang Kaiyi1,2

(1. HydroChina Guiyang Engineering Corporation Geotechnical Engineering Co., Ltd, Guizhou, 550081; 2. PowerChina Guiyang Engineering Corporation, Ltd, Guizhou, 550081)

Abstract: The relationship between coal permeability and temperature under 200 ℃ was studied using a coal-heating experiment, and the mechanism of coal permeability increase was analyzed using X-ray diffraction analysis and electron microscope scanning. The results show that coal rock’s permeability could be increased from 0.5 to 15 md after heat treating. These mechanisms may be explained as follows. First, the desorption of methane molecules and the dehydration of gelatinous microcomponents in the matrix, which shrinks the coal matrix. Second, dehydration of coal rock and volatile coal, which will flow out and form new pores. Third, coal rock encounters inhomogeneous deformation under thermal stress, producing a small crack, unevenly. Fourth, coal rock generates hydrocarbon gases after oxidation, forming tiny cracks and reducing the degree of coal rock compaction. Finally, the study proposes a new mode of coalbed methane (CBM) exploitation to enhance CBM recovery by injecting thermal CO₂. The above studies may be used to exploit CBM reservoirs with low permeability.

Keywords: coal rock; heating treatment; permeability; microcracks; mechanism; application

0 Introduction

Presently, coal bed methane (CBM) is a hot and difficult problem in the research and development of clean energy in China. The permeability of coal seams in China is low (generally not more than 1 md) (Ye Jianping, 1999), which restricts the efficient development of coalbed methane. The main technical measures of CBM stimulation, at home and abroad, are hydraulic fracturing, multi-element gas displacement, and directional-pinnate horizontal well technologies. However, there are some deficiencies in the application of these three technologies in China (Yang Xinle, 2009).

The permeability of coal can be increased by heat treatment, and the mechanism of this phenomenon is explored from the aspect of microstructure. Presently, many studies are available on the law of increasing permeability of heated coal and rock, at home and abroad. Wei Jianping and Sun Liutao (2017) studied the Change Law of coal permeability and the mechanism of increasing permeability under temperature shock. They found that the increased value of coal permeability, under thermal-shock treatment, was 3.6 times that under cold-shock treatment; compared with cold-shock treatment, the hot–cold treatment produces more microcracks and has a better antireflection effect. At the same time, they found that the anisotropy of coal properties and thermal stress produced by temperature shock are the main mechanisms of increasing the permeability of coal. Wang Ludi (2017) has studied the variation of permeability with temperature, in reference. The study shows that the critical threshold of permeability change is at 350 ℃, and the increment of permeability reaches its peak when the injection duration was 5 h, and then, heat injection has little effect on reservoir permeability. Yang Xinle (2009) has studied the mechanism of increasing production of coalbed methane (CBM) by heat injection in low permeability coalbed methane (CBM). When the CBM absorbs heat, the kinetic energy and activity of
gas molecules increase, several CBM is desorbed from the surface of the coal matrix, and the concentration of CBM increases. In addition, more coalbed gas diffuses outwards to the fracture through the bedrock and pore, in the fracture because the gas density and pressure gradient cause the gas seepage quantity to increase. The abovementioned literature analyzes the permeability increase in heated coal rock from a macroscopic aspect, however, the microstructural change of the coal, which is related to the permeability increase during the heating process is involved.

Based on the study of the relationship between the permeability of coal and temperature, the mechanism of increasing permeability of coal by heat is discussed from the aspect of microstructure, and a new exploitation model of coalbed methane by injecting hot CO₂ gas into coal seam is put forward. It is important to accelerate the development of low porosity and permeability coalbed gas reservoir.

1 Indoor experiment and observation

1.1 Mineral identification and analysis of coal samples

The coal samples from Taiyuan Formation No. 3 coal seam in the Jincheng Mining area were selected and identified using an X-ray diffractometer in the GPMR state key laboratory of the China University of Geosciences; it is thermally unstable.

| Table 1 mineral identification and thermal stability of coal samples |
|---------------------------------------------------------------|
| Sample number | Kaolinite/% | Quartz/% | Calcite/% | Dolomite/% | Amorphous substance/% |
|----------------|------------|----------|-----------|------------|-----------------------|
| Coal Sample 1(unheated) | 5 | 1 | 20 | 0 | 74 |
| Coal Sample 2(unheated) | 5 | 1 | 20 | 15 | 59 |
| Coal Sample 3(120℃ treatment) | 5 | 3 | 17 | 15 | 60 |
| Thermal stability (within 200℃) | stable | stable | stable | stable | unsteadiness |

1.2 Coal sample preparation

The three coal samples in Table 1 are taken from the same coal sample of Taiyuan Formation No. 3 coal seams in the Jincheng Mining area. The coal core with a diameter of 25 mm and length of 35 mm is drilled in the vertical direction of the coal and rock bedding with a vertical coring rig, and both ends of the coal core are cut and polished. During the experiment, an intelligent oven was used to heat the coal sample, and a JHGP gas permeability tester was used to measure the permeability of the coal. The heating temperature of coal in the experiment was within 200 °C. To avoid heat-treated coal samples absorbing moisture from the air, coal samples are sealed before and after the test.

1.3 Permeability test method and results

(1) Test the initial permeability of 1 # coal sample, then put the coal sample in the oven at 120 °C; after each heating for 2 h, test its permeability, and repeat the process for 16 h. The results are shown in Figure 1.

The permeability of coal samples heated at 120 °C increased from 0.4 to 16.3 md in the first 10 h and did not increase in the last 6 h with the increase in heating time (Fig. 1). The results show that the microcosmic change in the coal sample stopped after 10 h.
(2) test the initial permeability of 2# coal samples, then place the coal sample in the oven and heat it at 20°C as a gradient. After heating at each temperature point for 2 h, measure its permeability and quality until 180°C. The results are shown in Figure 2.

With the increase in heating temperature, the permeability of the coal samples increase from 0.5 md, at the initial stage, to 38.9 md at 180°C, and the quality of coal samples decreases. At 120°C, the permeability and mass have an inflection point, after which the permeability continues to increase, whereas the mass decreases.

Fig. 2 variation of permeability and quality of coal sample 2# during heating

1.4 Microstructure and observation of coal and rock

Coal is a porous material transformed from plant remains into a heterogeneous organic rock, and the cellular structure of plants has been preserved to varying degrees. The differences in plant primitive cell structure and preservation conditions cause the difference in pore types, sizes, and structures in coal reservoirs. In the process of coal metamorphism, the original pores are changed and new pores are formed. The micropores and microfractures in coal rocks are reservoirs of coalbed methane and the pathway of coalbed methane diffusion and migration. it was scanned using the environmental scanning electron microscope of the state key laboratory of GPMR. The result is shown in Figs 3 (I-V).
Structural vitrinite  Stroma vitrinite

I Magnify 25 times

Structural vitrinite  Stroma vitrinite

II Magnify 400 times

Structural vitrinite  Stroma vitrinite

III Magnify 1200 times
As shown in the SEM photo of No. 3 Jin-3 Coal in figure 3:

(1) There are several directional strip pores (see structural vitrinite in Figures I to IV) with sizes from 1 to 10 μm. These micropores are not only coalbed methane adsorption pores but also coalbed methane migration and diffusion pores. The structural vitrinite in Figure II contains a large fracture about 40 μm in size, which is perpendicular to the strip pore. It may be caused by some force during the formation of Coal Rock, and the large fractures of this size play an important role in the percolation of coalbed methane.

(2) The coal consists of two macerals, namely, structural and Matrix vitrinites. The structure vitrinite has a regular pore-fissure structure, whereas the Matrix vitrinite is irregular. The structural vitrinite is formed, partly, from the cell wall of plants, whereas the Matrix vitrinite is amorphous and unstructured.

(3) No matter the structure of vitrinite or the Matrix vitrinite, several mineral particles are in the pores or on the surface. These mineral particles originate from the coal-forming environment of plants and are composed of carbonate minerals.

2 Discussion on the mechanism of increasing permeability of coal and rock

Based on the results of X-ray diffraction and SEM analysis, we concluded that the increase in the permeability of coal after heat treatment is caused by the following four factors.

2.1 Coal Matrix shrinkage

According to Duthie’s law, the permeability of a coal sample depends on four factors: pressure difference at the two ends of the coal sample, seepage length, area of the seepage passage, and the viscosity of the fluid. During the heating process, the matrix of coal rock will shrink and the size of fractures will increase, thus, the permeability of coal rock will increase. Two main reasons for Matrix contraction:

(1) Contraction due to the release of methane molecules from the surface of the coal matrix (Chen Jingang, 2004). Due to the imbalance in gravitation, the surface of coal at the edge of the micropore has high free energy and strong adsorption to gaseous molecules. The activation energy of methane adsorbed on the surface of coal increases after heating. Once the activation energy exceeds the surface free energy of coal, methane can be desorbed from the bond of the coal matrix. Methane molecules are released from the micropore-fractured space and the colloidal particle structure of coal and the coal matrix shrinks, which leads to the relative crack opening of the coal.

(2) Shrinkage of coal matrix due to dehydration during heating (Pan Zhejun, 2010). The gelling component in the coal matrix has strong water absorption and retention capacity because of its well-developed pore structure and more hydrophilic groups. The results show that when the temperature rises to 100 °C–120°C, the adsorbed water will be separated from the gelated component of the Matrix. The Matrix will expand under the invasion of water. Once the water evaporates, the Matrix will dry and shrink.
2.2 Dehydration of coal and volatilization of volatile components

Adsorbed, interlayer, and structural water are in the rocks and minerals. The results show that the combination of adsorbed and interlayer water with minerals is looser and can be separated within 200 °C, while the temperature of the crystal lattice of structural water should reach 400 °C–800 °C (Liu Junrong, 2010).

The treatment temperature in this experiment is less than 200 °C. Thus, the reason for the decrease in the quality in this process is that the adsorbed water and the interlayer water in the coal rock are separated from the coal rock, however, the structural water remains in the coal rock due to the low temperature of coal rock treatment. The volume of various water molecules in coal is not negligible compared with the pore volume of coal. Therefore, with the increase in temperature, the water in pore passage evaporates and the volume of coalbed methane seepage passage increases, which is beneficial to the flow of gas, thus, the permeability of coal rock increases.

Another reason for the decreasing dry weight of coal is the change in some components in the minerals, such as thermal decomposition and volatilization. The calcite content of the coal sample treated at 120 °C decreased by 3% (Table 1), indicating that the calcite have decomposed into lime, carbon dioxide, and water. These components will volatilize from the coal rock in the form of gas, resulting in the formation of large and small voids; these voids increase the flow area of the seepage passage, thus, increasing the permeability of the coal rock.

2.3 Thermal stress causes tiny cracks in coal

When heating various rock masses, the core will produce greater thermal stress (Li Zenghua, 1996). Solid particles have certain internal stresses; when the thermal stress produced during heating exceeds its internal stress, it will destroy the original stress equilibrium state of the solid, leading to stress redistribution to create some structural changes.

This change is such that the coefficient of thermal expansion of each component of coal and rock will be different. The thermal expansion anisotropy causes the particle to expand and increase in volume. This change in volume and the difference in the thermal expansion will cause the local stress concentration in the coal and rock. Therefore, when the thermal stress produced by the temperature exceeds the tensile stress between and within the coal particles, the coal rocks are destroyed and new microcracks are produced. The change in the structure will destroy the original micронetwork structure, form a new fracture network, increase the connectivity of the original fracture and fracture structure network, and thus, improve the permeability of coal.

2.4 The molecular chains of coal are broken, and the gaseous products are formed and dispersed

It can be seen from Table 1 that one of the important components of coal is amorphous. This amorphous material is a variety of tymacromolecules, the structure of aromatic rings, and has a variety of side chains. The molecular chain of coal is easy to break under the action of external factors, such as geostress, mechanical action, and heat. The molecular chain rupture involves the rupture of a covalent bond in the chain, in which several free radicals are produced. In the presence of oxygen and heat, the broken chain molecules in the benzene ring can be decomposed or oxidized into various gaseous products (Yan Ronglin, 1995). Therefore, under the heat, the coal molecular chain breaks, and many carbon and hydrogen atoms escape from the coal sample in the form of gas, resulting in several new cracks in the coal. At the same time, the density of the coal rock decreases, thus, increasing the permeability of coal rock.

3 Discussion on eor model of thermal CO₂ injection in coal seam

3.1 Selection of well type and process of heat enhanced permeability

In the middle and later period of CBM exploitation, the output of CBM declines. Based on the above results, if a hot gas is injected into a wellhead of a U-shaped well (Fig. 4), the gas enters the wellbore under
pressure and penetrates the coal and rock of the sidewall, simultaneously, the heat by the well can be exchanged with the coal rock around the well, thus, improving the permeability of coal rock. Once the permeability of coal and rock around the wellbore increases, the CBM, which cannot be desorbed due to the low permeability will be desorbed into the wellbore, thus, increasing the CBM production. The hot gas will escape to the other wellhead of U-shaped well, and then to the exit wellhead.

![Fig. 4 schematic of eor by injecting hot gas into U-shaped well](image)

**3.2 Selective analysis of hot gases**

The analysis shows that there are four heat-carrying gases in the process: water vapor, N₂, CH₄, and CO₂. Water Vapor is easy to be produced, however, it is easy to be damaged by water lock when it condenses in the pores and fissures of coal or rock after heat exchange. The temperature of the other three gases can rise to higher temperatures, the adsorption effect of coal on these three gases is CO₂, CH₄, N₂ from large to small (Tang Shuheng, 2001). The better the adsorption effect of coal to gas, the greater the amount of gas entering the coal seam in the shaft wall, the more sufficient the heat exchange is inevitable. CO₂ can be adsorbed with lime solution after being discharged from the wellhead, which can be recycled. In addition, CO₂ injection into the coal seam to exploit CBM can help to reduce the greenhouse effect. Therefore, CO₂ is the first choice for hot gas injection.

**4 conclusions and recommendations**

(1) The relationship between the permeability of coal and temperature is studied using laboratory experiments. The results show that the permeability of coal can be increased by heating.

(2) The release of free water, adsorbed water, and volatiles in coal rock will make the pores of coal rock open and increase the permeability of coal rock. In addition, when the temperature rises to 120 °C, the free and the adsorbed water in the coal and rock volatilize.

(3) The methane molecules in the pores of the coal matrix will break away from the surface gravity of the coal matrix after being heated, and the adsorbed water in the coal matrix will volatilize after being heated, leading to the dry shrinkage of the coal matrix; both of these factors will cause the shrinkage of the coal matrix, thus, increasing the permeability of coal.

(4) In the process of heating, the covalent bond of coal molecules will break and produce many kinds of gas products, which will produce cracks and reduce the compactness of coal and improve the permeability of coal and rock.

(5) According to the rule that the permeability of coal rock increases under thermal action, a tentative plan
of injecting thermal CO₂ into U-shaped coal well to improve the extraction ratio of coalbed gas is put forward, theoretically, and its feasibility is discussed.

(6) It is difficult to observe the development of microcracks in coal rocks, after heating, due to the limitation of experimental conditions. The effect of stress should be considered in the study.

References
[1] Ye Jianping, Shi Baosheng, Zhang Chuncai. Permeability of coal reservoirs in China and its main influencing factors [J]. Journal of coal, 1999,24(2):118-122.
[2] Yang Xinle. Study on mechanism of increasing production of coalbed methane by heat injection in low permeability coal reservoir [D]. Fuxin: Liaoning Technical University, 2009.
[3] Wei Jianping, Sun Liutao, Wang Dengke. Permeability change law and anti-permeability mechanism of coal under temperature shock [J]. Journal of coal, 2017,42(8):1919-1925.
[4] Reed Wang. Study on permeability law of low permeability coal-bed gas by thermal injection[J]. Coal Mine Safety, 2017,4(48) : 5-8.
[5] Wang G X, MASSAROTTO P, Rudolph V. An improved performance model of coal for coalbed methane recovery and Co 2 geosequestration [J]. International Journal of Coal Geology, 2009,77,127-136.
[6] Liu Junrong, Wu Xiaodong. Preliminary study on mechanism of thermal permeability enhancement of Rock [J].Petroleum drilling and production process, 2003,25(5) : 43-49.
[7] Chen Jingang, Shixiong, etc. Experimental study on internal control factors of coal matrix shrinkage [J] . Coal Geology and exploration, 2004,32(5) : 26-28.
[8] Pan Zhejun, CONNEL L D, Camilleri M. Laboratory characterization of coal reservoir persistence for primary and enhanced coalbed methane recovery [J] . International Journal of Coal Geology, 2010,82,252-261.
[9] Qiu Xiaoling. On the law of water change in coal and rock composition [J] . Coal, 2003,12(6) : 48-49.
[10] Yan Ronglin, Qian Guoyin. Molecular structure of coal and gaseous products of coal spontaneous combustion [J] . Journal of coal, 1995,20(6) : 58-65.
[11] Li Zeng-hua. Radical mechanism of coal spontaneous combustion. Journal of China University of Mining and technology, 1996,25(3) : 111-114.
[12] Liu Junrong, Wu Xiaodong. Microcosmic experimental study on heat-treated rocks [J] . Proceedings of the Southwest Petroleum University: Natural Science, 2008,30(4) : 15-18.
[13] Guo Shucai. Coking Technology [M] . BEIJING: Metallurgical Technology Press, 1961.
[14] Tang Shuheng. Characteristics of coal reservoirs and adsorption-desorption characteristics of multi-component gases in Jincheng Area [D] . Xuzhou: China University of Mining, 2001.