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Full Length Article

Simulation of dynamic changes of methane state based on NMR during coalbed methane output

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HIGHLIGHTS

- Adsorption and desorption of dried coal follow a logarithmic relation with time.
- Adsorption is from larger pores to smaller pores in the adsorption pores.
- The adsorption rate during the initial stage does not correlate to the pressure.
- Adsorption process includes three stages and reversibility is affected by water.
- The bottom hole pressure difference plays a dual role during the production of CBM.

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ABSTRACT

With anthracite coal of the Qinshui basin as the study object, the effect of water and the displacement pressure on the state of coalbed methane (CBM) during production was studied by combining core flooding with nuclear magnetic resonance (NMR). The adsorption of methane is from larger pores to smaller pores in the adsorption pores. Adsorption and desorption of a dried coal sample follows a logarithmic behavior with time. The process of adsorption is divided into three stages. The adsorption rate increases rapidly in the first stage. The adsorption rate in the initial stage does not correlate to pressure. Meanwhile, the water in the coal reservoir not only hinders the migration of CBM but also reduces the rate of adsorption and desorption. Furthermore, the reversibility of adsorption and desorption is affected. The bottom hole pressure difference plays a dual role during the production of CBM. It can promote the migration of CBM, while hinder its desorption. The control of the bottom hole pressure difference and displacement pressure is very important during CBM continuous output and enhanced CBM recovery. The reasonable range is influenced by the critical desorption pressure, type of reservoir porosity and permeability.

1. Introduction

CBM has been playing an increasingly important role in natural gas. Many countries are developing CBM [1]. At present, America has obtained significant success in developing CBM from lignite [2]. Meanwhile, China has achieved success in developing CBM from anthracite in recent years. The research of occurrence and migration of CBM in coal reservoir has enormous significance for the production of CBM [3]. Most of CBM is adsorbed by the micropores [4]. In general, water needs to be drained from the reservoir before CBM desorption. Subsequently, CBM needs to migrate out of the micropores to be drained [5,6]. CBM begins to diffuse from the matrix to the cleats because of the difference in gas concentrations [7]. Finally, CBM flows to the well through the cleat system due to the pressure difference [8].

Many studies [2,9–15] have reported the effect of water on the gas adsorption capacity using a high pressure water injection and isothermal adsorption. These studies concluded that the methane adsorption capacity is inhibited with increasing moisture content. However, the adsorption capacity holds constant once the moisture content exceeds a certain critical value [7,10,12]. Moreover, water in coal also plays a significant role in the production of CBM [8,16]. The migration ratio of methane in coal decreases with increasing moisture content [7,17]. Although the effects of water and other factors on adsorption, desorption and migration have

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been studied extensively, the dynamic change in the state of CBM during its production is difficult to observe by conventional methods. Low-field nuclear magnetic resonance technology has an advantage in solving this problem due to its non-destructive characteristic [18–21].

There have been a few displacement experiment studies on methane and the interaction between methane and carbon dioxide in rocks, nanoporous materials and other porous media by NMR technology [20,22–24]. However, these materials are different from coal. Only a limited number of experiment studies have been reported on methane and water in coal by NMR technology. The study by Guo et al. [25] illustrated the state of water and methane in coal, and Yao et al. [24] further studied the state of methane in powdered coal. In their research, adsorbed methane, bound methane in porous medium and free methane were identified. On the one hand, the lump coal sample was different from the powdered coal sample with regard to the storage of methane. On the other hand, their research mainly focused on the change in the state of methane with pressure. In fact, the state of methane changes with time during CBM production. Moreover, the state of methane is also affected by water and the bottom hole pressure difference. To solve these problems, a combination of core flooding and low-field ^{1}H NMR technology was proposed, which allows the dynamic change in the state of methane during CBM production to be observed. Moreover, the factors that affect the state of CBM can also be studied.

2. Materials and methods

2.1. Coal samples

The sample employed in this study was obtained from No. 3 coal bed of the Yameidaning mine, south of Shanxi Province, China. Proximate analysis and vitrinite reflectance for the coal obtained are summarized in Table 1. The coal sample is 2.5 cm in diameter and 4.7 cm in length. It was dried in an oven at 110 °C for 5 h to remove pre-existing moisture.

2.2. Experimental setup

NMR experiments were conducted with a 12.798 MHz Macro MR-150 spectrometer, manufactured by Niumag Corporation Ltd. Figs. 1 and 2 show the schematic diagram of the NMR experiments, which consists of an NMR measurement instrument and a displacement device. The displacement device includes a displacement pump, a confining pressure pump and a non-magnetic core holder. To remove air from the sample and monitor the confining pressure and displacement pressure, a vacuum pump and two pressure sensors were mounted.

2.3. NMR relaxation properties of methane in coal

Hydrogen protons are polarized when the sample is stimulated under a magnetic field [26]. Then the relaxation time (T_1 or T_2) is measured [26]. T_1 and T_2 contain the same information. However, the measuring time of T_1 is longer than that of T_2. Therefore, T_2 is often used in many studies. The equation for T_2 is as follows [24,27]:

$$\frac{1}{T_2} = \frac{1}{T_{2S}} + \frac{1}{T_{2B}} + \frac{1}{T_{2D}} \quad (1)$$

where T_{2S} is the surface relaxation, T_{2B} is the bulk relaxation and T_{2D} is the diffuse relaxation. For a homogeneous internal field gradient, T_{2D} can be ignored [24]. Since the pores in the coal sample are mainly micropores, bulk relaxation can also be ignored. Eq. (1) then becomes:

$$\frac{1}{T_2} = \frac{1}{T_{2S}} = \rho \frac{S}{V} \quad (2)$$

where ρ is the surface relaxation rate and S/V is the specific surface area of the pores. As the specific surface area increases, T_2 becomes shorter. At the same time, the specific surface area of coal is mainly controlled by micropores. Therefore, the smaller the pore is, the shorter the transverse relaxation time will be.

2.4. Experimental methodology

The dried coal sample was placed in the non-magnetic core holder. The test results show that there was no nuclear magnetic signal for the dried coal sample (Fig. 3). Subsequently, the experimental device was evacuated for 8 h. Finally, the vacuum pump was removed and a tube was connected to valve 2. To observe the gas flow rate, the other end of the tube was immersed in water.

(1) The confining pressure and displacement pressure were set as 6 MPa and 2 MPa, respectively. Gas passed through the sample and was immediately released after opening valve 2, indicating that methane had filled the entire pipeline. Then, the displacement pressure was successively adjusted to 1 MPa, 1.5 MPa and 2 MPa. After that, the coal sample was desorbed for approximately 12 h.
(2) The displacement pressure was adjusted to 1.5 MPa; then, the coal sample was adsorbed for some time until its signal amplitude matched the signal amplitude of the initial adsorption. Afterwards, the setup was changed from gas displacement (Fig. 1) to water displacement (Fig. 2). Heavy water was used to replace ordinary water due to its non-magnetic characteristic. To simulate the reservoir condition, water was made to flow to the coal sample at a displacement pressure of 1.6 MPa. Then, the displacement pressure was adjusted to 2.5 MPa and methane was released slowly after opening valve 2. The displacement pressure was adjusted to 2.0 MPa after 8 h and then gradually decreased. The displacement pressure was reduced to 0.6 MPa after 13 h. Meanwhile, methane could not be observed from the gas outlet. However, methane was released slowly when the displacement pressure was adjusted to 1.5 MPa.
(3) The coal sample that was displaced by water was adsorbed by methane again, from which the impact of water on the adsorption capacity of coal could be observed. The displacement pressure was adjusted gradually from low to high. The methane was released slowly when the displacement pressure was adjusted to 2.5 MPa. Then, the displacement pressure was adjusted to 1.5 MPa. After that, the coal sample began to desorb after 3 h. The desorption lasted for approximately 10.5 h.

The changes of methane in the experiments were observed in T_2 spectrum, which was based on the parameters given in Table 2.

Table 1
Proximate analysis and vitrinite reflectance of the coal sample.

M_{ar} (%)	A_d (%)	V_{daf} (%)	$R_{o,max}$ (%)
3.37	12.25	5.5	2.89

M_{ar} is the moisture content when received, A_d is the ash yield after drying, V_{daf} is the volatile matter content of dry and ash free sample, and $R_{o,max}$ is the vitrinite reflectance.