Measurement of gas diffusion coefficient and analysis of influencing factors for Shaanxi Debao coalbed methane reservoir in China

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Received: 22 August 2020 / Accepted: 26 November 2020 / Published online: 4 January 2021
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Abstract
Shanxi Debao has affluent coalbed methane (CBM) reservoir in China. Understanding its characteristics, especially the diffusion characteristics of natural gas in the coal seam, is the key data of CBM development in this block. In this paper, a method of measuring the gas diffusion coefficient of coal and rock by the adsorption method is proposed by using the experimental method. The calculation model for calculating the gas diffusion coefficient of coal and rock is established according to the adsorption amount, and the diffusion correlation coefficient under different pressures, different water saturation and the different temperatures is established. The results show that it is feasible to calculate the diffusion coefficient of the gas in coal and rock according to the adsorption capacity of coal and rock. With the increase in the pressure, the diffusion coefficient increases, the water saturation increases. While the diffusion coefficient decreases, the temperature increases, and the diffusion coefficient increases. The results calculated by the model could provide guidance for the future development of Debao coalbed methane reservoir in Shanxi Province, China.

Keywords Coalbed methane reservoir · Diffusion coefficient · Pressure · Temperature · Saturation

Introduction
The data of the coalbed methane reservoir in the Debao area, Shanxi Province, China, are very rich and are preparing for large-scale development (Yu 1992; Wang et al. 1994). However, at present, the diffusion mechanism of gas in coalbed methane reservoir is not clear enough, and it is necessary to understand the characteristics of gas diffusion, which is the basis of scientific development of coalbed methane reservoir (Yu et al. 1986, 1989; Zhou and Sun 1965; Yu 1985), while the research of coalbed methane reservoir diffusion is the current research hot spot.

Scanning electron microscope (SEM) and low-temperature nitrogen adsorption test were applied to systematically study the pore structure characteristics of coal and rock, and the influence of tectonic action on pore system and methane diffusion model was analyzed by Jiang et al. (2016). In 2016, the experimental object is located in the south of Huhe lake depression in Hailaer basin. It concluded that in the initial stage of gas production, molecular diffusion plays an important role, while with the decrease in the pressure, the diffusion mode of methane molecules in coal and rock in the middle part of trough is mainly transitional diffusion, and the diffusion model of methane molecules in coal and rock in anticline changes little; in 2016, Sun et al. (2016) evaluated the reliability of extended Langmuir, IAS and 2D PR-EOS multi-component gas adsorption models based on experimental data, and established Maxwell–Stefan double-diffusion model to simulate the gas diffusion process. Finally, the double-diffusion model is coupled with the gas water two-phase multi-component seepage model of the coal reservoir, and the IMPES method is used to solve the problem. The results show that the predicted results of 2D PR-EOS model are better than those of extended Langmuir model and IAS model, while at the beginning of gas injection, the multi-component gas adsorption and methane desorption rate in the matrix is faster, and then gradually slow down; thus, the double-diffusion model is established.

The influence of different gas pressures, coal rank and moisture on methane diffusion coefficient was tested and analyzed by Zhao et al. (2016) by using a flaky coal matrix.
as a detection sample. It is concluded that the flaky samples keep the special spatial structure of the coal matrix, and the methane diffusion coefficient of coal matrix (order of magnitude \(10^{-11} - 10^{-9} \text{ m}^2/\text{s}\)) is more reasonable. When the temperature is constant, the methane diffusion coefficient of the coal matrix increases with the increase in the gas pressure, whether it is dry samples, saturated water samples, or low-, medium-, and high-coal-rank samples. The adsorption capacity of coal with a different metamorphic degree is different. With the increase in the coal metamorphism, the adsorption capacity of coal rock is enhanced, but the diffusion coefficient of the coal matrix decreases first and then increases with the increase in the coal metamorphism. The methane diffusion coefficient of saturated samples is lower than that of dry samples, and the diffusion rate of dry samples is faster, so the diffusion coefficient of the coal matrix is obtained.

Zhao et al. (2016) summarized the general understanding and the latest progress of permeability dynamic changes, the problems existing in the research process and the development trend in the future were pointed out. It is concluded that the effective stress, coal matrix shrinkage and Klinkenberg effect are the main factors affecting CBM well drainage and production, resulting in an asymmetric “U” type change trend in the future were pointed out. It is concluded that in the process of drainage and production of high-rank coalbed methane wells, the coal seam permeability presents the characteristics of decrease recovery increase; the effective stress effect and matrix shrinkage effect directly control the change characteristics of coal seam permeability. The adsorption and desorption characteristics fundamentally control the action time and intensity of matrix shrinkage effect, and the dynamic change characteristics of coal permeability are obtained.

In 2014, through the application of molecular dynamics and rock mechanics theory, the diffusion and seepage mechanism of high-order coalbed methane was analyzed by Li et al. (2014). Using Simed software, the initial drainage strength of high-order coalbed methane wells with different coal structures is studied by using the S-D permeability model with constant permeability, stress sensitivity and considering the change of coal matrix compressibility. Taking the coalbed methane reservoir in the south of Qinshui Basin as an example, the conclusion is drawn: With the development of drainage and production, the diffusion coefficient will gradually increase, while the fracture conductivity will decrease due to effective stress and coal powder plugging; permeability is the key factor affecting the gas well productivity in the study area (Zhou and Lin 1999; Cao et al. 2004; Zhang 1998), and the gas production effect with high permeability is good; the structural coal is sensitive to the initial liquid drop rate, but not to the higher conductivity coefficient; the primary and fractured coal is not sensitive to the initial liquid drop rate. However, it is sensitive to the conductivity coefficient; the low-permeability CBM well should adopt a lower initial liquid drop rate; the high-permeability CBM well can adopt a higher initial drainage strength to continuously discharge water and pulverized coal (Sang et al. 2005; Liu et al. 2000; Wang 2005; Fu et al. 2001). Based on the above research, this paper studies the characteristics of Debao coalbed methane reservoir in the Shanxi Province of
China. Through the experimental study of coal samples, this paper proposed the method of applying experimental adsorption research to obtain the diffusion coefficient of coal, rock and gas fields under different pressures, temperatures and water saturation. The gas movement regulation of Debao coalbed methane reservoir has laid the foundation.

**Study on measurement method of the gas diffusion coefficient of coalbed methane reservoir**

Generally, the diffusion coefficient in coal seam cannot be directly measured; therefore, the method of testing adsorption is proposed to test, and then, the diffusion coefficient of the coalbed methane reservoir is calculated according to the adsorption amount.

**Experimental process**

The flowchart of the experiment is shown in Fig. 1, and the equipment used is shown in Fig. 2.

The coal sample diffusion coefficient test device is shown in Fig. 1, which including coal sample, flange, sealed diffusion chamber, constant pressure device, gas source, measurement device and vacuum pumping device. The process of measuring the diffusion coefficient is as follows.

The wire mesh wrapped coal sample is suspended inside the sealed diffusion chamber to ensure that all surfaces of the coal matrix are in full contact with the coalbed methane in the sealed diffusion chamber, and then, the coal sample is fully degassed by vacuuming. The sample cylinder is sealed by flange.

A constant pressure device is used to inject methane with a certain pressure into the diffusion chamber. When the valve is closed, the pressure sensor is used to continuously detect and record the pressure change in the diffusion chamber with time until the coal is fully saturated, while the pressure remain unchanged. Before the test of pressure drop, if the pressure in the sealed diffusion chamber is lower than the rated pressure, coalbed methane will be injected into the sealed diffusion chamber through the sealed pipe to make the gas pressure in the sealed diffusion chamber reach the rated pressure.

The diffusion coefficient of coalbed methane in coal matrix is calculated by the flat diffusion model according to the rated temperature, rated pressure and the amount of...
coalbed methane injected into the sealed diffusion chamber at any time by using the data acquisition card and computer. According to the measured experimental data, the diffusion coefficient of coalbed methane is calculated by Fick’s law. This method can use the same coal as underground coal seam as the detection sample and maintain the special spatial structure of coal sample. At the same time, the vacuum test can be carried out under the condition of no other gas interference, so that the adsorption and diffusion of coalbed methane are more sufficient, and the detection results of diffusion coefficient are closer to the diffusion coefficient of coalbed methane in the real block coal of underground coal seam.

**Coal sample preparation**

Before measuring the diffusion coefficient of coal sample, the coal sample of raw coal is a regular cuboid sample which is cut into the regular cuboid sample by slicing machine, then refined by automatic grinding machine and finally cleaned by ultrasonic cleaning machine; then, the volume of sample is calculated by drainage method, and the thickness of the sample is measured by Vernier caliper; then, the coal sample is dried in the constant temperature box, and the basic material diffusion coefficient can be tested after the sample is completely dried. In order to compare the difference of diffusion coefficient of coal and rock samples under dried and saturated conditions, after the diffusion coefficient of dry coal sample is measured, the sample is saturated with distilled water, and then, the matrix diffusion coefficient under saturated water condition is measured. Figure 3 demonstrates the coal sample.

**Method of calculating the diffusion coefficient**

A parallel thin plate (coal sample sheet) with a thickness of $L$ is placed in a closed container filled with a certain volume of solution. According to Fick’s law, the diffusion of the solution in the thin plate is described by Eq. (1).

$$\frac{\partial c}{\partial t} = D \frac{\partial^2 c}{\partial x^2}$$  \hspace{1cm} (1)

In formula:
- $c$ Gas concentration (mg m$^{-3}$);
- $x$ Diffusion distance (m);
- $t$ Diffusion time (s);
- $D$ Diffusion coefficient (m$^2$ s$^{-1}$).

$x = 0$ is the surface of coal sample and the coal sample has no adsorption gas.

The initial condition of Eq. (2) is as follows:

$$c(x, 0) = 0, \quad 0 < x < Lc(x, 0)$$ \hspace{1cm} (2)

In the formula:
- $L$ Sample length (m).

When the adsorption reaches equilibrium (the rate of gas entering the diffusion chamber from the coal surface is equal to the rate of gas entering the coal sample from the diffusion chamber), the boundary conditions of Eq. (3) are obtained

$$\begin{cases}
\frac{\partial c}{\partial t} = 0 & x = \frac{1}{2}L, \quad t < 0 \\
\frac{\partial^2 c}{\partial x^2} = D \frac{\partial c}{\partial x}, & x = 0, \quad t > 0 \\
\alpha = \frac{V_g}{aL}
\end{cases}$$ \hspace{1cm} (3)

In the formula:
- $a$ Ratio of gas volume in sample cylinder to sample volume;
- $V_g$ Total volume of gas in sample cylinder (m$^3$);
- $A$ the area of the largest surface of the sample, while the sample volume is $V_{sa} = AL$, in square meters (m$^2$);
- $\alpha$ the partition coefficient, usually is 1.

Compose equations (Eq 1)–(Eq 3) under the initial conditions (Eq 2) and boundary conditions (Eq 3), the solution of equation (Eq 1) is

$$1 - \frac{q_t}{q_\infty} = \sum_{n=1}^{\infty} \frac{2a(1+a)}{1+a+a^2q_n^2} \exp \left( -\frac{4a^2q_n^2Dt}{L^2} \right)$$ \hspace{1cm} (4)

In the formula:
- $q_t$ the adsorption capacity of coal to gas at time $t$, in cubic meters per ton (m$^3$ t$^{-1}$);
- $q_\infty$ the saturated adsorption capacity of coal, in cubic meters per ton (m$^3$ t$^{-1}$);
- $q_n$ the continuous nonzero positive roots of equation $\tan(q) + aq = 0$.

For gases

$$1 - \frac{q_t}{q_\infty} = \frac{p - p_\infty}{p_2 - p_\infty}$$ \hspace{1cm} (5)

In the formula:
- $p$ the gas pressure at time $t$ (MPa);
- $p_2$ the initial gas pressure (MPa);
the initial gas pressure (MPa).

Substitute Eq. (5) into Eq. (4), then:

\[
p - p_\infty = \frac{2a(1 + a)}{1 + a + a^2q_1^2} \exp \left( -\frac{4q_1^2Dt}{L^2} \right)
\]

In the formula:

\[
q_1 \text{ the first nonzero positive root of equation } \tan(q) + aq = 0.
\]

Take the natural logarithm on both sides of Eq. (6), then:

\[
\ln(p - p_\infty) = \ln \left( \frac{2a(1 + a)}{1 + a + a^2q_1^2}(p_2 - p_\infty) \right) - \frac{4q_1^2Dt}{L^2}
\]

To

\[
\ln(p - p_\infty) = \ln \left( \frac{2a(1 + a)}{1 + a + a^2q_1^2}(p_2 - p_\infty) \right) - \frac{4q_1^2Dt}{L^2}
\]

The equation \(\ln(p - p_\infty)\) and \(t\) were fitted linearly; then, the diffusion coefficient \(D\) of methane in coal can be calculated by the slope \(K\):

\[
D = -\frac{KL^2}{4q_1^2}
\]

Test results and analysis

This paper mainly analyzes the influence of testing pressure, testing temperature and water saturation of coal on gas diffusion coefficient in Debao coalbed methane reservoir of Shanxi Province.

Measurement of the diffusion coefficient under different pressures

In this experiment, the diffusion coefficient of four coal samples from Baode coalbed methane reservoir (HB-5\#, HY-11\#, HJ-3\#, HW-11\#) under 6 pressure points (room temperature 20 °C) was measured. The experimental results are shown in Figs. 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26 and 27, where the abscissa is time \(t\) and the ordinate is \(\log(p - p_\infty)\).

Based on the above data and Eq. 8, the diffusion coefficient of coalbed methane under different pressures of different coal samples in Baode coalbed methane reservoir can be calculated. Table 1 shows the comparison of the experimental results of four coal samples under different pressures.

According to Table 1, the comparison chart of the diffusion coefficient of four coal samples under different injection pressures can be drawn, as shown in Fig. 28. The variation law of diffusion coefficient of 3\#, 5\# and 11\# coal seams in the Hancheng area can be obtained through comparative analysis.

Figure 28 demonstrates that the diffusion coefficients of HB-5\#, HY-11\#, HJ-3\# and HW-11\# are positively correlated.
which is the diffusion coefficient of the coal matrix increases with the increase in the experimental pressure. The adsorption capacity of coal increases with the increase in the pressure, but it barley increases when the coal sample reaches the adsorption saturation. According to the experimental data, the diffusion coefficients of coal $3^\#$, $5^\#$ and $11^\#$ are similar, which is related to the same rank of coal samples $3^\#$, $5^\#$ and $11^\#$.

Measurement of diffusion coefficient under different saturation

Four coal samples were placed into the sample cylinder and measure their diffusion coefficient (room temperature 20 °C). Then the diffusion coefficient is compared with that of dry coal.

The experimental results are shown in Figs. 29, 30, 31, 32 and Table 2.
According to the above data, the gas diffusion coefficient of coal samples under 100% saturation can be obtained by calculating the above data according to formula Eq 8, as shown in Table 2.

Through the comparison between Tables 1 and 2, it can be seen that the diffusion coefficient of the saturated coal sample is smaller than that of the dry coal sample. The reason is that the polar functional groups on the coal surface...
have good wettability to water, and the adsorption force of coal matrix on methane molecules is very small, which makes methane molecules not easy to be adsorbed, so it is not easy to form a large concentration gradient, so the gas diffusion coefficient in the sample is low and the diffusion rate is slow.
Measurement of the diffusion coefficient at different temperatures

Generally, the temperature is an important factor affecting the gas diffusion coefficient, so it is necessary to study the gas diffusion coefficient at different temperatures. Put the coal sample into the sample cylinder, and measure the diffusion coefficient after 2 to 4 days of constant temperature.

In this experiment, HY-11# coal was selected to carry out the experiment at the experimental temperature of 40 °C. The experimental results are shown in Fig. 33.

The diffusion coefficient of the HY-11# coal sample is $3.54 \times 10^{-9}$ m²/s at 3.8 MPa. Compared with the diffusion coefficient of this coal sample under dry and saturated conditions, the diffusion coefficient at 40 °C is greater than that at 20 °C. This is due to the increase in temperature and the acceleration of gas molecular movement.

Figure 34 demonstrates the diffusion coefficient. The experimental results show that the diffusion coefficient of the saturated coal sample is smaller than that of the dry coal sample, and the higher the coal grade is, the smaller the difference value is. The different microstructure of pore leads to the different gas diffusion coefficient. With the increase in the coal rank, the total pore volume first decreases and then rises slowly, and the diffusion coefficient changes accordingly. The measurement results of the specific surface area also confirm this change. With the increase in the proportion of mesoporous and micropore, the adsorption diffusion rate gradually slows down. It is generally considered that in the early stage of CBM exploitation, CBM flows in fracturing fractures and large fractures, so the diffusion coefficient has little effect on the initial production of CBM. However, in the later stage of CBM exploitation, with the production of pulverized coal and the reduction in bottom hole pressure, the pressure-sensitive effect leads to the decrease in the coalbed permeability, and the size of the diffusion coefficient directly affects the production of CBM in the later stage.

**Conclusion**

(1) By using the flaky coal as the sample for detection, the unique spatial structure of coal sample is maintained. At the same time, the test is carried out under safe and non-interference conditions, which makes the adsorption and diffusion of coalbed methane more sufficient, and the results are closer to the diffusion coefficient of coalbed methane in the real massive coal matrix of underground coal seam. The adsorption method proposed in this paper to study the diffusion coefficient of the gas in coal and rock is feasible. The formula for calculating the diffusion coefficient is simple and convenient.

(2) The diffusion coefficient of Debao coalbed methane reservoir is affected by many factors, such as pressure, temperature and water saturation. The influence of each factor is different. For pressure, the diffusion coefficient of the gas in coal and rock increases with the increase in the pressure.

(3) The diffusion coefficients of dry coal samples and coal samples with 100% saturated were compared by the experimental method. The experimental results show that the diffusion coefficient of saturated coal samples in Baode
Coalbed methane reservoir is smaller than that of dry coal samples.

(4) The results show that the higher the temperature is, the greater the diffusion coefficient is. This is because the higher the temperature is, the faster the gas molecules move, and the diffusion coefficient increases.

Table 1: Comparison of diffusion coefficients of 4 coal samples

| Sample         | Pressure (MPa) | Linear fitting equation | Diffusion coefficient (10⁻¹⁰m²/s) |
|----------------|----------------|-------------------------|-----------------------------------|
| HB-5# (vertical) | 0.62           | y = -1.74404 − 0.000196222x | 1.12733                           |
|                | 1.11           | y = -1.49312 − 0.000329467x | 1.89284                           |
|                | 1.53           | y = -1.3938 − 0.000567951x | 3.26297                           |
|                | 2.07           | y = -1.79502 − 0.000853289x | 4.90228                           |
|                | 2.82           | y = -1.72329 − 0.000881925x | 5.06681                           |
|                | 2.95           | y = -1.38738 − 0.000941373x | 5.40835                           |
| HY-11# (vertical) | 0.60           | y = -1.96009 − 0.000413791x | 1.55046                           |
|                | 1.35           | y = -2.02282 − 0.000199759x | 2.19675                           |
|                | 1.53           | y = -1.874 − 0.000217779x  | 2.39492                           |
|                | 2.49           | y = -1.94644 − 0.000469446x | 5.1625                           |
|                | 2.77           | y = -1.15623 − 0.000511605x | 5.62612                           |
|                | 3.62           | y = -1.47602 − 0.000706456x | 7.76891                           |
| HJ-3# (parallel) | 0.44           | y = -2.641690.000937119x   | 1.53948                           |
|                | 1.83           | y = -2.1694 − 0.000736197x  | 2.99411                           |
|                | 1.86           | y = -1.80518 − 0.000205857x | 3.38177                           |
|                | 2.97           | y = -1.43779 − 0.000355939x | 5.8473                           |
|                | 3.38           | y = -1.44755 − 0.000467439x | 7.67894                           |
| HW-11#         | 0.52           | y = -1.87895 − 0.00016654x  | 1.82588                           |
|                | 1.21           | y = -1.96258 − 0.000436394x | 2.15736                           |
|                | 2.06           | y = -0.84237 − 0.000224631x | 2.46275                           |
|                | 1.91           | y = -1.18463 − 0.000206801x | 2.26727                           |
|                | 2.77           | y = -1.04987 − 0.000231239x | 2.5352                           |
|                | 3.40           | y = -0.634468 − 0.000242302x| 2.65649                           |
|                | 4.16           | y = -0.885991 − 0.000382191x| 4.19017                           |

Fig. 28: Comparison of results of 4 coal samples

Fig. 29: HB-5# results of diffusion coefficient at 3.72 MPa
Table 2: Experimental results of 4 coal samples under 100% saturation

| Coal sample | Injection pressure | Linear fitting equation | Diffusion coefficient ($10^{-10} \text{m}^2/\text{s}$) |
|-------------|--------------------|--------------------------|-----------------------------------------------|
| XS-5#       | 3.72               | $y = -1.00357 - 0.00035919x$ | 2.06259                                       |
| YC-11#      | 3.38               | $y = -1.50683 - 0.000189549x$ | 2.08448                                       |
| SJ-3#       | 4.13               | $y = -1.54661 - 0.000222499x$ | 3.65514                                       |
| NG-11#      | 2.99               | $y = -1.50579 - 0.000179828x$ | 1.97155                                       |

Fig. 30 HY-11# results of diffusion coefficient at 3.38 MPa

Fig. 31 HJ-3# results of diffusion coefficient at 4.13 MPa

Fig. 32 HW-11# results of diffusion coefficient at 2.99 MPa

Fig. 33 HY-11# results of diffusion coefficient at 3.79 MPa

Fig. 34 Diffusion coefficient of saturated coal sample and air dry coal sample
Funding  The authors received no specific funding for this work.

Data availability  The data used to support the findings of the study are included within the article.

Compliance with ethical standards

Conflict of interest  The authors declared that there is no financial and personal relationships with other people or organizations that can inappropriately influence our work, there is no professional or other personal interest of any nature or kind in any product, service and/or company that could be construed as influencing the position presented in, or the review of, the manuscript entitled, “Measurement of gas diffusion coefficient and analysis of influencing factors for coalbed methane reservoir.”

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