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Coupled Mathematical Model for Prediction of Coal Seam CH_4 Gas Diffusion Coefficient

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Abstract

Considering the lack of simple and effective test method or prediction model for coal seam CH_4 gas diffusion coefficient, this paper analyses the factors influencing the diffusion coefficient based on the physical simulation experiment of CH_4 diffusion in reservoir condition, selects crustal stress, ground temperature, reservoir pressure and direction of diffusion path as qualitative and quantitative variables, and builds a coupled mathematical model for coal seam CH_4 diffusion using the Quantification Theory I. Through theoretical and empirical analysis, it is concluded that the proposed model features high accuracy and fast speed, and sheds new light on the prediction of coal seam CH_4 gas diffusion coefficient.

Key words

Diffusion coefficient, Quantification theory, Coupled mathematical model, Prediction.

1. Introduction

The diffusion coefficient is extensively applied to explore the diffusion features of coal seam CH_4 gas. The important and practical parameter is mainly identified through desorption method and block-sample gas chromatographic method [1-2]. Nevertheless, the effect of desorption method is limited to the characterization of the diffusion features of coal dusts [3-4]. The block-sample gas chromatographic method overcomes the limitation and expands the scope of characterization to coal pillars. However, the method is too difficult to implement in actual practice.

Based on the physical simulation experiment of CH_4 diffusion in reservoir condition, this paper analyses the factors influencing the CH_4 diffusion coefficient, selects the parameters and the "Mathematical Model for Gas Geology" software for modeling, and constructs a coupled mathematical model for coal seam CH_4 diffusion using the Quantification Theory I. The accuracy of the model was proved through precision investigation.

2. Construction Principles of the Model

The model was constructed on the following principles: On the basis of the physical simulation experiment of CH_4 diffusion in reservoir condition, the variation law and main influencing factors of coal seam CH_4 gas diffusion should be analysed through multi-factor physical simulation experiment on CH_4 gas diffusion with variable conditions; With the variable law and influencing factors, the multivariable mathematical model of coal seam CH_4 gas diffusion should be established using a certain mathematical method.

The Quantification Theory I was adopted for construction of the model. As it name suggests, it is one of the many methods of the quantification theory. Essentially, the Quantification Theory I is a multivariate statistical analysis method capable of processing both quantitative and qualitative variables simultaneously [5]. As a result, the method has been a popular tool for predicting dependent variable based on independent variables that are qualitative or both qualitative and quantitative.

Some of the factors related to CH_4 gas diffusion are difficult to quantify during the analysis. For instance, the division of coal seam CH_4 gas diffusion path into vertical and parallel diffusions is qualitative in nature and involves no quantity. Such variables are known as qualitative variables [6-8]. These variables should not be ignored if they are main influencing factors of coal seam CH_4 diffusion. Nevertheless, the traditional multivariate statistical methods like multivariate regression can only predict dependent variable based on quantitative variables. Therefore, the Quantification Theory I is a suitable method for building the model [9-10].

(1) Introduction to the Quantification Theory I

As a common way to predict quantitative dependent variable, the Quantification Theory I works well when independent variables are qualitative or both qualitative and quantitative.

(2) Mathematical model of Quantification Theory I

① Mathematical model for qualitative independent variables

Assuming that the interaction between the dependent variable and each item or category abides by the linear model below:

$$y_i = \sum_{j=1}^{m} \sum_{k=1}^{r_j} \delta_i(j,k) b_{jk} + \varepsilon_i, \quad i = 1,2,\ldots,n \tag{1}$$

Find the coefficient b_{jk} by the least squares principle to minimize the deviation of the predicted value from the measured value, that is, to minimize:

$$q = \sum_{i=1}^{n} \varepsilon_i^2 = \sum_{i=1}^{n} \left[y_i - \sum_{j=1}^{m} \sum_{k=1}^{r_j} \delta_i(j,k) b_{jk} \right]^2 \tag{2}$$

Find the partial derivative of q for buv and set it to 0, and we have:

$$\frac{\partial q}{\partial b_{uv}} = -2 \sum_{i=1}^{n} \left[y_i - \sum_{j=1}^{m} \sum_{k=1}^{r_j} \delta_i(j,k) b_{jk} \right] \delta_i(u,v) = 0 \tag{3}$$
$$u = 1,2,\ldots,m; v = 1,2,\ldots,r_u$$

Denote the b_{jk} corresponding to the minimum q as $\hat{b}_{jk}$. Then, $\hat{b}_{jk}$ should satisfy the above formula, that is:

$$\sum_{j=1}^{m} \sum_{k=1}^{r_j} \left[\sum_{i=1}^{n} \delta_i(j,k) \delta_i(u,v) \right] \hat{b}_{jk} = \sum_{i=1}^{n} \delta_i(u,v) y_i \tag{4}$$
$$u = 1,2,\ldots,m; v = 1,2,\ldots,r_u$$

Rewrite the above formula as a matrix:

$$X'X\hat{B}=X'Y \tag{5}$$

where $Y' = (y_1, y_2, \ldots, y_n)$;

$$\hat{B}' = (\hat{b}_{11}, \ldots, \hat{b}_{1r_1}, \hat{b}_{21}, \ldots, \hat{b}_{2r_2}, \ldots, \hat{b}_{m1}, \ldots \hat{b}_{mr_m}) \tag{6}$$

The equation system is called the normal equation (system).

After solving $\hat{b}_{jk}$ via the normal equation, the prediction equation is obtained as follows:

$$\hat{y} = \sum_{j=1}^{m}\sum_{k=1}^{r_j}\delta(j,k)\hat{b}_{jk} \tag{7}$$

where $\delta(j, k)$ is the reaction of any sample to the k-th category of the j-th item. For any sample, it is possible to compute $\hat{y}$, the predicted value of the dependent variable y, with the above formula based on its reaction $\delta(j, k)$.

For the established prediction equation, the significance should be verified in the F-test, the contribution of each item to the prediction should be tested by calculating the partial correlation coefficient, and the prediction accuracy should be checked by computing the residual standard deviation. Through the F-test, the items in the prediction equation may be properly added or reduced to keep only the items with significant impact on the dependent variable, thus creating the optimal prediction equation.

② Solution of the normal equation system

The normal equation system $X'X\hat{B}=X'Y$ is proved to contain $\sum_{j=1}^{m} r_j - m + 1$ linearly independent equations at the most, indicating that *X'X* is a rank deficient coefficient matrix consisting of $\sum_{j=1}^{m} r_j - m + 1$ ranks at the most. Thus, the equations have infinite number of solutions. When the number of samples n is large enough, there are always $\sum_{j=1}^{m} r_j - m + 1$ ranks of *X'X*. In this case, we can delete the equations in the first category of the j-th item, and set $\hat{b}_{j1} = 0$. After the deletion, the coefficient matrix of the equation system has full rank, making it possible to find the unique solution to the rest $\hat{b}_{jk}$. The normal equation solving process prevents the loss of generality, and minimizes the value of q.

③ Mathematical model for qualitative and quantitative independent variables

Suppose there are h quantitative and m qualitative independent variables. The values of the h quantitative variables in the i-th sample are expressed as $x_i(u)$ (u=1, 2, …, h; i=1, 2, …, n). The number of qualitative independent variables corresponds to the number of items. The j-th item contains rj categories. For the m qualitative independent variables, the degree of reaction to the i-th sample is expressed as (j =1, 2, …, m; k=1, 2, r_j; i=1, 2, …, n). Besides, the value of the dependent variable is expressed as y_i(i=1, 2, …, n).

Suppose that the following linear model applies to the reaction between the dependent variable and each quantitative independent variable, item and category:

$$y_i = \sum_{u=1}^{h} b_u x_i(u) + \sum_{j=1}^{m}\sum_{k=1}^{r_j} \delta_i(j,k) b_{jk} + \varepsilon_i, \quad i = 1,2,\dots,n \tag{8}$$

where b_u (u=1, 2, …, h) and b_{jk} (j=1, 2, …, m; k=1, 2, …, rj) are unknown coefficients; ε_i(i=1,2, …,n) is a random error. Through a derivation process similar to the case of only qualitative independent variables, the least squares estimates of b_u and b_{jk} are derived as $\hat{b}_u$ (u=1, 2, …, h) and $\hat{b}_{jk}$ (j=1, 2 , …, m; k=1, 2, …, rj), which satisfy the normal equation:

$$X'X\hat{B} = X'Y \tag{9}$$

where $X = \begin{bmatrix} x_1(1) & \dots & x_1(h) & \delta_1(1,1) & \dots & \delta_1(1,r_1) & \delta_1(2,1) & \dots & \delta_1(m,r_m) \\ x_2(1) & \dots & x_2(h) & \delta_2(1,1) & \dots & \delta_2(1,r_1) & \delta_2(2,1) & \dots & \delta_2(m,r_m) \\ \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots & \dots \\ x_n(1) & \dots & x_n(h) & \delta_n(1,1) & \dots & \delta_n(1,r_1) & \delta_n(2,1) & \dots & \delta_n(m,r_m) \end{bmatrix}$

$$\hat{B}' = (\hat{b}_1,\dots,\hat{b}_h,\hat{b}_{11},\dots,\hat{b}_{1r_1},\hat{b}_{21},\dots,\hat{b}_{mr_m})$$
$$Y' = (y_1, y_2, \dots, y_n)$$

The above equation can be solved under the condition of $\hat{b}_{j1} = 0$ (j=2, …, m). Thus, the prediction equation is obtained as:

$$\hat{y} = \sum_{u=1}^{h} \hat{b}_u \, x(u) + \sum_{j=1}^{m} \sum_{k=1}^{r_j} \delta(j,k) \hat{b}_{jk} \tag{10}$$

(3) The significance test and prediction accuracy of the prediction equation

The established prediction equation should be statistically tested to verify its prediction effect. The test indices include F-statistic, and the multiple correlation coefficient R.

$$F = \frac{\sum_{i=1}^{n} (\hat{y}_i - \bar{y})^2 / m}{\sum_{i=1}^{n} (y_i - \hat{y}_i)^2 / (n - m - 1)} \tag{11}$$

where m is the first degree of freedom and n-m-1 seconds is the second degree of freedom. According to the F-distribution table at a certain significance level, the y is linearly correlated with m items if the F-statistic surpasses the threshold, and the prediction equation is significant. Otherwise, the equation is not significant.

$$R = \frac{d_{\hat{y}}}{d_y} = \sqrt{\frac{\sum_{i=1}^{n} (\hat{y}_i - \bar{y})^2}{\sum_{i=1}^{n} (y_i - \bar{y})^2}} \tag{12}$$

The closer the value of R to 1, the greater the correlation between y and m items, and the more significant the equation.

3. Physical Simulation Experiment of CH_4 Diffusion in Reservoir Condition

3.1 Experimental System and Philosophy

The experimental system mainly consists of a TK-1 natural gas diffusion measuring system and a GC-2014 gas chromatograph analyzer. As the core device of the experiment, the measuring system includes a core holder, an annular pressure system, a gas supply system, a vacuum-pumping system, and a constant temperature control system. The core holder provides a space to simulate the stratum environment of the test core; the annular pressure system applies necessary pressure on the core; the constant temperature control system controls the temperature of the core in a proper range; the gas supply system provides the two gas chambers with CH_4 and N_2, respectively; the vacuum-pumping system creates a vacuum environment before the gases are introduced to the two gas chambers (Figure 1).

Fig.1. Physical graph of the experimental device

The experimental philosophy is based on the free diffusion of gas through the coal sample under the concentration gradient. There are two diffusion chambers, one at each end of the coal sample. One of the chambers should be filled with CH_4, and the other should be filled with N_2. There should not be any pressure difference between the gases of the two ends. Under the rated temperature difference, the concentration of each gas changes with time. The diffusion coefficient of CH_4 in coal seam can be obtained by measuring the concentration of each gas in the two diffusion chambers at different times.

3.2 Coal Seam Collection and Preparation

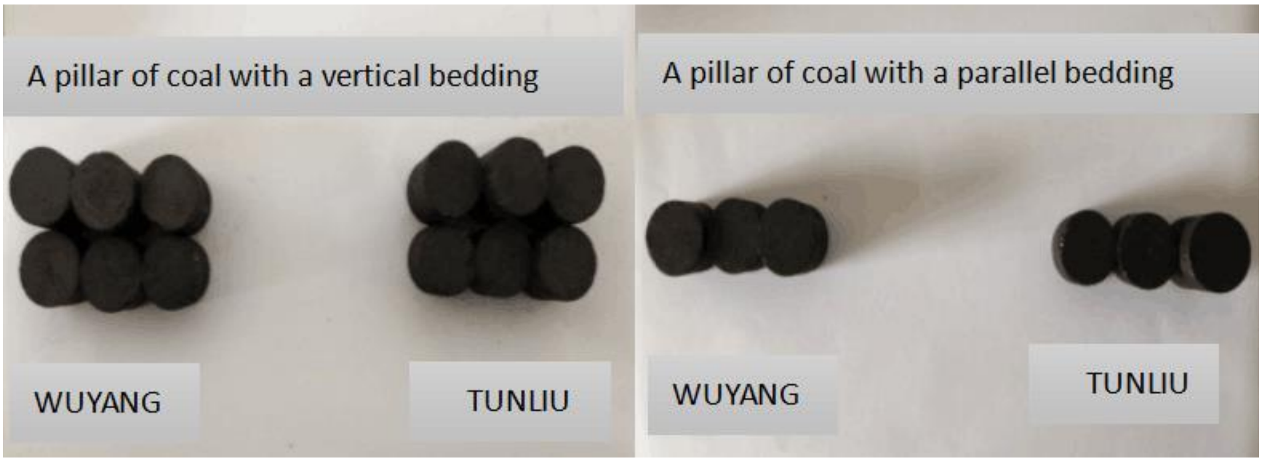

Fig.2. Raw coal samples

Coal block taken in the direction vertical to the bedding from Wuyang; Coal block taken in the direction parallel to the bedding form Tunliu

The coal samples were meager coal ($R_{o,max}$=2.10%) collected from coal seam 3# in Wuyang Mine and Tunliu Mine, Lu'an Mining Area. According to the national standard, the test samples were taken from the wax-sealed raw coal with a Φ25mm×50mm core barrel in the directions vertical and parallel to the bedding; the upper and lower ends were polished with a cutting machine (smoothness≤0.02%) and made parallel to each other, so that the two ends are under uniform stress during the loading process. Figure 2 show the coal samples.

3.3 Experimental Plan

Aside from the bedding features of the coal seam, the external factors of coal seam temperature, reservoir pressure and crustal stress also directly bears on the diffusion performance of coal seam CH_4 gas [11-13]. In this experiment, the external factors were approximated by the confining pressure, temperature and gas pressure. The experiment conditions (Table 1) were determined based on the actual conditions of coal seam 3# in Wuyang Mine and Tunliu Mine [14] and in consideration of the experiment purposes and the pressure conditions of the coal samples.

3.4 Calculation of Diffusion Coefficient

The diffusion coefficient (D/cm^2/s) [15] is an important yardstick of the diffusion capacity of gas in the coal seam. If the diffusion rate per unit area is proportional to the concentration gradient, the diffusion rate is time-invariant and dependent only on distance. This scenario, known as the (quasi) steady-state diffusion, follows the Fick's first law [16]. If the diffusion flux of coalbed CH_4 varies with both time and distance, the scenario is called the unsteady diffusion and is explained by Fick's second law [17]. Thus, the diffusion coefficient of the CH_4 gas in the experiment should be calculated by Fick's second law (Equation 13):

$$D=\frac{\ln(\Delta C_0/\Delta C_i)}{B(t_i-t_0)} \tag{13}$$

where $\Delta C_i=C_{1i}-C_{2i}$; $B=A(1/V_1+1/V_2)/L$; D is the diffusion coefficient of CH_4 gas in the coal samples (cm^2/s); ΔC_0 is the concentration difference of CH_4 gas in the two diffusion chambers at the initial moment (%); ΔC_i is the concentration difference of CH_4 gas in the two diffusion chambers at moment i (%); t_i is moment i (s); t_0 is the initial moment(s); C_{1i} is the concentration of CH_4 gas in the CH_4 diffusion chamber at moment i (%); C_{2i} is the concentration of CH_4 gas in the N_2 diffusion chamber at moment i (%); A is the sectional area of the coal sample (cm^2); L is the length of the coal sample (cm); V_1 and V_2 are the volumes of the CH_4 and the N_2 diffusion chambers, respectively (cm^3).

Table 1 lists the calculated CH_4 diffusion coefficients under different conditions. It can be seen that the CH_4 diffusion coefficients measured by the block-sample gas chromatographic method concentrate in the order of magnitudes between 10^{-7} and 10^{-8}.

Tab.1. Raw coal CH_4 diffusion experiment under different conditions

Mine	Sample No.	Conditions			CH_4 chamber concentration		N_2 chamber concentration		CH_4 diffusion coefficient D/(cm^2/s)
		Confining pressure/MPa	Temperature/℃	Gas pressure/MPa	CH_4/%	N_2/%	CH_4/%	N_2/%	
Wu yang Mine	CZ1	21.00	24.00	6.60	97.68	2.32	0.77	99.23	2.03E-08
	CZ1	21.00	24.00	8.60	98.18	1.82	1.26	98.74	1.38E-08
	CZ1	21.00	24.00	10.60	99.18	0.82	0.80	99.20	1.16E-08
	CZ1	21.00	24.00	12.60	98.98	1.02	0.65	99.35	1.06E-08
	CZ2	18.50	21.00	5.80	99.87	0.13	0.10	99.90	1.14E-08
	CZ2	15.00	21.00	5.80	99.55	0.45	0.066	99.93	2.64E-08
	CZ2	11.50	21.00	5.80	99.23	0.77	0.26	99.74	4.56E-08
	CZ2	8.00	21.00	5.80	99.20	0.80	0.15	99.85	8.14E-08
	CZ3	16.00	18.00	4.90	99.90	0.10	0.02	99.98	1.13E-08
	CZ3	16.00	28.00	4.90	99.92	0.08	0.07	99.93	1.37E-08
	CZ3	16.00	38.00	4.90	99.85	0.15	0.04	99.96	1.65E-08
	CZ3	16.00	48.00	4.90	99.82	0.18	0.08	99.92	2.13E-08
	PX1	21.00	24.00	6.60	96.86	3.14	3.98	96.02	3.88E-07
	PX2	18.50	21.00	5.80	95.51	4.49	5.75	94.25	4.88E-07
	PX3	16.00	18.00	4.90	97.42	2.58	3.91	96.09	6.41E-07
Tunliu Mine	CZ4	21.00	24.00	6.60	99.81	0.19	0.09	99.90	1.99E-08
	CZ4	21.00	24.00	8.60	99.89	0.11	0.07	99.92	1.37E-08
	CZ4	21.00	24.00	10.60	99.84	0.16	0.03	99.97	1.13E-08
	CZ4	21.00	24.00	12.60	99.88	0.12	0.05	99.95	1.08E-08
	CZ5	18.50	21.00	5.80	99.86	0.14	0.06	99.94	1.13E-08
	CZ5	15.00	21.00	5.80	99.85	0.14	0.30	99.70	2.77E-08
	CZ5	11.50	21.00	5.80	99.70	0.30	0.59	99.41	3.99E-08
	CZ5	8.00	21.00	5.80	99.65	0.35	0.56	99.44	6.24E-08
	CZ6	16.00	18.00	4.90	99.91	0.09	0.05	99.95	0.98E-08
	CZ6	16.00	28.00	4.90	99.89	0.11	0.05	99.95	1.27E-08
	CZ6	16.00	38.00	4.90	99.87	0.13	0.06	99.94	1.51E-08
	CZ6	16.00	48.00	4.90	99.84	0.16	0.14	99.86	2.13E-08
	PX4	21.00	24.00	6.60	98.39	1.61	3.03	96.97	2.99E-07
	PX5	18.50	21.00	5.80	97.87	2.13	5.80	94.19	4.56E-07
	PX6	16.00	18.00	4.90	96.37	3.63	5.97	94.03	6.08E-07

Note: CZ indicates that the sample was taken in the direction vertical to the bedding; PX indicates that the sample was taken in the direction parallel to the bedding.

4. Model Establishment

(1) Selection and valuation of variables

The model was established by the Quantification Theory I with CH_4 gas diffusion coefficient as dependent variable, and the influencing factors as independent variables.

① Dependent variable

The CH_4 gas diffusion coefficient measured under different reservoir conditions.

② Independent variables

The independent variables are classified into qualitative and quantitative variables by nature. The former category includes the vertical path and the parallel path, while the latter includes confining pressure, temperature and gas pressure. The qualitative variables were valued as binary-state variables, that is, the presence/absence of a certain attribute was valued as 1/0.

Table 1 displays the original data and variable values for modeling.

Tab.2. Original data and data values for modelling

Serial No.	Sample No.	Conditions			Diffusion path		CH_4 gas diffusion coefficient (cm^2/s)
		Confining pressure (MPa)	Temperature (℃)	Gas pressure (MPa)	Vertical path	Parallel path	
1	CZ1	21	24	6.6	1	0	2.03E-08
2	CZ1	21	24	8.6	1	0	1.38E-08
3	CZ1	21	24	10.6	1	0	1.16E-08
4	CZ1	21	24	12.6	1	0	1.06E-08
5	CZ2	18.5	21	5.8	1	0	1.14E-08
6	CZ2	15	21	5.8	1	0	2.64E-08
7	CZ2	11.5	21	5.8	1	0	4.56E-08
8	CZ2	8	21	5.8	1	0	8.14E-08
9	CZ3	16	18	4.9	1	0	1.13E-08
10	CZ3	16	28	4.9	1	0	1.37E-08
11	CZ3	16	38	4.9	1	0	1.65E-08
12	CZ3	16	48	4.9	1	0	2.13E-08
13	PX1	21	24	6.6	0	1	3.88E-07
14	PX2	18.5	21	5.8	0	1	4.88E-07
15	PX3	16	18	4.9	0	1	6.41E-07
16	CZ4	21	24	6.6	1	0	1.99E-08
17	CZ4	21	24	8.6	1	0	1.37E-08
18	CZ4	21	24	10.6	1	0	1.13E-08
19	CZ4	21	24	12.6	1	0	1.08E-08
20	CZ5	18.5	21	5.8	1	0	1.13E-08
21	CZ5	15	21	5.8	1	0	2.77E-08
22	CZ5	11.5	21	5.8	1	0	3.99E-08
23	CZ5	8	21	5.8	1	0	6.24E-08

24	CZ6	16	18	4.9	1	0	0.98E-08
25	CZ6	16	28	4.9	1	0	1.27E-08
26	CZ6	16	38	4.9	1	0	1.51E-08
27	CZ6	16	48	4.9	1	0	2.13E-08
28	PX4	21	24	6.6	0	1	2.99E-07
29	PX5	18.5	21	5.8	0	1	4.56E-07
30	PX6	16	18	4.9	0	1	6.08E-07

(2) Establishment of the prediction equation

Following the principles of Quantification Theory I, the model was established with the selected dependent and independent variables and in accordance with the basic data in Table 2.

To test the significance of the equation, the F-statistic should be calculated by the variance analysis method, and compared to the F distribution table. The equation is proved significant if the F-statistics surpasses the threshold.

The significance of each independent variable in the equation should also be tested. The partial correlation coefficient method is commonly used out of the various test methods. In this method, the partial correlation coefficient of the dependent variable should be calculated relative to each independent variable. The greater the coefficient, the closer the dependent variable is to the dependent variable. Meanwhile, the t_i-statistic should be obtained based on the partial correlation coefficients, and compared to the t distribution table at a certain significance level. If t_i is greater than or equal to the t threshold, the i-th independent variable is significant to the equation. The prediction equation should contain only the independent variables significantly correlated with the dependent variable.

The “Mathematical Model for Gas Geology” software was adopted to cope with the large amount of calculation during the model construction based on Quantification Theory I. The software offers a rapid, efficient, accurate and flexible way to perform the calculation on computers.

The model was created as follows based on the Mathematical Model for Gas Geology” software:

$$y= \left| -0.0000000075\, dl\,(1) -0.0000000010 dl\,(2) +0.0000000016 dl\,(3) +1.6242115424\, dx\,(1,\ 1) +6.2960782662 dx\,(1,\ 2) \right| \tag{14}$$

where y is the CH_4 gas diffusion coefficient (cm^2/s); $dl\,(1)$ is the confining pressure (MPa); $dl(2)$ is the quantitative variable of temperature (℃); $dl(3)$ is the quantitative variable of gas pressure

(MPa); *dx(1,1)* is the qualitative variable of diffusion path (vertical to the bedding); *dx(1,2)* is the qualitative variable of diffusion path (parallel to the bedding).

The F-statistic of the prediction equation was 92.088875, far greater than the threshold $F_{4,30}^{0.01}$=4.02 at the significance level of 0.01; the multiple correlation coefficient R= 0.967701 is close to 1; the residual standard deviation S= 0.527800 is close to 0. The results demonstrate the significance and high accuracy of the equation (model).

Conclusion

Through the analysis of experiment results, this paper selects crustal stress, ground temperature, reservoir pressure and direction of diffusion path as the main influencing factors on CH_4 gas diffusion, and, on this basis, establishes the model using the Quantification Theory I. Both theoretical and empirical analysis manifest that the proposed model features high accuracy and fast speed.

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