Study on geochemistry discriminate method of gas emission in goaf

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Abstract: In order to effectively distinguish the source of gas gush in the goaf of the coal mine, Sihe coal mine was chosen as the studied object. Carbon isotope of methane, ethane, carbon dioxide and hydrogen isotope of methane in gas from the desorbed gas of the working coal seam and the near coal seam, and gas in the goaf were tested, by systemic sampling coal samples and gas samples of the goaf. Combined with gas component from the goaf, the working coal seam and the near coal seam, the proportion of gas gush in the goaf were determined by the numerical calculation on separate source of isotopes. The results show that the difference of gas component, stable carbon and hydrogen isotopes of gas from the different coal seams is obvious. And the methane concentration in gas increase along with the increase of coal seam depth, carbon and hydrogen isotopes in gas become heavier along with the deepen of burial depth of coal seam depth. The results of the numerical calculation on separate source indicate that the proportion of the gushed gas in the lagged traverse No.1 from the desorbed gas of coal seam No.3~No.8-2 is 38%, 37%, 12%, 8%, 5%, the proportion of the gushed gas in the lagged traverse No.2 from the desorbed gas of coal seam No.3-~No.8-2 is 22%, 36%, 24%, 12%, 6%.

1. Introduction

Goaf gas extraction is an important measure to prevent and control the overrun of mine gas. Gas sources must be integrated to reveal the specific source of gas, in order to effectively improve the efficiency of gas extraction, and to take reasonable extraction process and parameters. Domestic and foreign scholars generally formed a unified understanding of the ways of gas source. They generally believed that the gas emission sources in the coal mining face mainly include the gas poured out of the mining coal wall, mining down coal gas, the gas left out of the goaf and the gas poured out of upper and lower adjacent layers[1-4]. In relation to the gas emission sources, scholars at home and abroad analyzed the gas emission sources on the basis of gas concentration. This kind of study usually adopt similar physical simulation, numerical simulation and theoretical analysis, and analyze the gas flow rule to determine the distribution of the gas concentration. Thus it is concluded that the gas emission sources[3]. In this study, the vast majority of scholars established the gas flow model by the CFD software. Then simulated the stope gas migration rule by using Fluent software for numerical solution of gas flow rule means. Simulated the change rule of the gas concentration to obtain the distribution rules of gas concentration[4-10]. In the research of the gas emission prediction, there are several kinds of calculating method such as the calculation method of point source, estimated the gas emission on the...
basis of the change of the return air gas emission amount before and after the old roof caving, mapped method and so on[11]. That mainly used mine statistic source prediction method. According to the different sources of gas emission, used mine statistical method to study the law of the gas emission, respectively. Thus predicted the volume of the gas emission from different source. Finally, calculated the total volume of the gas emission. In combination with traditional gas concentration analysis, at the same time domestic researchers carried out the work using the carbon isotope geochemical means for the abnormal of coal mine gas[12]. For the gas source in goaf, some researchers gathered the gas sample in rock tunnel and the adjacent coal seam, and analyzed the carbon isotope to determine the gas source in the coal seam[13-14]. So it is difficult to distinguish between different sources on the basis of gas composition in the coal seam.

In conclusion, past researches had not really determined the gas emission sources of coal seam, and did not form an effective method of gas emission source identification. In this study take the No.3 coal mined-out area of the main working bed in Sihe mine on as the object of the research. Collected the coal and goaf gas sample, then tested and analyzed the components of desorbed gas and goaf gas and the stable isotope composition in the main working bed and adjacent seam. Mathematical calculation is used to determine the proportions of the goaf gas components from different sources. Preliminary established the gas emission prediction method based on the comprehensive discriminate of the goaf gas source. This method can provide technical and theoretical basis for the mine to take reasonable effective extraction process and parameters.

2. Methodology

2.1 Sampling and analysis
This study collected 40 coal samples from shallow to deep of Sihe coal mine in Jincheng of Shanxi province. There are 12 samples from No.3 coal seam, 4 samples from No.5 coal seam, 6 samples from No.7 coal seam, each 4 samples from No.8-1 and No.8-2 coal seam, 4 samples from No.9 coal seam and 6 samples from No.15 coal seam. The sampling method of making desorbed gas in desorption experiment was performed according to Coalbed methane(CBM) resources exploration technical specification (GB/T 29119-2012) . The weight of the coal samples which were collected to desorbed coalbed methane is about 200~300g. Loaded the coal samples to the vacuum tank and seal quickly. In order not to distort the composition of coalbed methane, filled the vacuum tank with coal samples as much as possible. The gas samples which were used to analyze the source of goaf are from lagged traverse No.1 and lagged traverse No.2, respectively.

2.2 Test method
Taking the gas sample collected from the field and the desorbed gas sample collected by desorption experiment as the object of laboratory experiment. Experiment is divided into two parts: ① The gas component concentration test instrument is GC-7800 gas chromatograph, the experimental process was conducted in accordance with the national standard GB/T 19559-2008. ② The gas stable carbon isotope test instrument is Delta Plus stable isotope gas mass spectrometer. The test method performed in accordance with the national standard GB/T 18340-2001. The methane stable hydrogen isotope test instrument is gas chromatography-isotope spectrometer (GC/TC/Delta Plus XL) . The test method is gas chromatography-high temperature pyrolysis-mass spectrometry analysis method online.

2.3. Analytical method
\[ \delta_{\text{max}} = \frac{V_A \delta_A + V_B \delta_B}{V_A + V_B} \]  (1)
In Eq.1, \( \delta_A \) was defined as the \( \delta^{13} \)C in methane of the first gas source, \( V_A \) was defined as the volume. \( \delta_B \) was defined as the \( \delta^{13} \)C in methane of the second gas source, \( V_B \) was defined as the volume. \( \delta_{\text{max}} \) was defined as the carbon isotope of methane in the mixed gas. The derivation process is illustrated by the case of methane, it is also established for any other kind of isotope values. Therefore, the values of
carbon isotope in the mixed gas could calculate by Eq.1. The error will not exceed the allowed error (0.5‰).

Based on the above formula, if the number of gas source in the mixed gas is n, the calculation formula of stable isotope in the mixed gas is Eq.2:

$$\delta_{\text{mix}} = \frac{V_1 \delta_1 + V_2 \delta_2 + ... + V_n \delta_n}{V_1 + V_2 + ... + V_n}$$

In Eq.2, $\delta_{\text{mix}}$ was defined as the carbon isotope values in the mixed gas. $V_1, V_2, ..., V_n$ were defined as the volume values of a component in gas from 1 to n gas source, respectively. $\delta_1$ to $\delta_n$ were defined as the isotope values of a marker in gas from 1 to n gas source, respectively.

In Eq.2, when the number of gas source in the goaf mixed gas is 5, the calculation model is Eq.3:

$$\delta_{\text{mix}} = \frac{V_1 \delta_1 + V_2 \delta_2 + ... + V_5 \delta_5}{V_1 + V_2 + ... + V_5}$$

Among them, $V_1, V_2, ..., V_5$ were defined as a component volume flow rate of the gas from 1 to 5 gas source, respectively. $\delta_1$ to $\delta_5$ were defined as a marker isotope characteristic value of the gas from 1 to 5 gas source, respectively. In order to simplify the calculation, set the total volume of the gas from different gas sources for unit volume. $V_1, V_2, ..., V_5$ were defined as the volume ratio of the gas from 1 to 5 gas source in the gas of the goaf (%), and $V_1 + V_2 + ... + V_5 = 1$. A marker isotope characteristic value of different gas sources used the median value of the isotope in Figure 1, the volume characteristic value used the average value.

3 Analysis of gas source in the goaf

3.1 Experimental results

The data analyses showed that the methane content from the desorbed gas of No.3 coal seam in the study area is about 42.68%~93.68% with 78.80% on average, ethane content is about 0.003%~0.024% with 0.013% on average, nitrogen content is about 4.71%~54.93% with 16.10% on average, carbon dioxide content is about 0.56%~11.37% with 5.08% on average. The methane content from the desorbed gas of No.5 coal seam in the study area is about 79.68%~84.14% with 83.24% on average, ethane content is about 0.013%~0.025% with 0.018% on average, nitrogen content is about 11.32~17.23% with 14.12% on average, carbon dioxide content is about 0.36%~4.90% with 2.62% on average. The methane content from the desorbed gas of No.7 coal seam in the study area is about 76.86%~96.56% with 88.16% on average, ethane content is about 0.011%~0.034% with 0.024% on average, nitrogen content is about 2.06%~22.52% with 10.23% on average, carbon dioxide content is about 0.73%~3.25% with 1.59% on average. The methane content from the desorbed gas of No.8-1 coal seam in the study area is about 89.67%~97.88% with 93.78% on average, ethane content is about 0.015%~0.033% with 0.027% on average, nitrogen content is about 2.54%~9.19% with 5.14% on average, carbon dioxide content is about 0.31%~1.69% with 1.05% on average. The methane content from the desorbed gas of No.8-2 coal seam in the study area is about 94.38%~97.33% with 95.58% on average, ethane content is about 0.018%~0.038% with 0.030% on average, nitrogen content is about 1.57%~5.25% with 3.48% on average, carbon dioxide content is about 0.34%~1.13% with 0.92% on average. The methane content from the desorbed gas of No.9 coal seam in the study area is about 96.24%~97.90% with 97.02% on average, ethane content is about 0.016%~0.044% with 0.032% on average, nitrogen content is about 1.68%~3.28% with 2.32% on average, carbon dioxide content is about 0.41%~1.47% with 0.63% on average. The methane content from the desorbed gas of No.15 coal seam in the study area is about 96.87%~98.73% with 97.82% on average, ethane content is about 0.018%~0.047% with 0.035% on average, nitrogen content is about 1.04%~3.10% with 1.70% on average, carbon dioxide content is about 0.08%~1.16% with 0.45% on average.

The carbon isotope of methane ($\delta^{13}C_{CH_4}$) from the desorbed gas of No.3 coal seam in the study area is about -29.87‰~39.16‰ with -34.22‰ on average, carbon isotope of ethane ($\delta^{13}C_{C_2H_6}$) is about -10.27‰~18.10‰ with -13.76‰ on average, carbon isotope of carbon dioxide ($\delta^{13}CO_2$) is
about -9.97‰~16.07‰ with -12.61‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -167.46‰~186.52‰ with -175.05‰ on average. The carbon isotope of methane ($\delta^{13}$C$_{CH_4}$) from the desorbed gas of No.5 coal seam in the study area is about -31.65‰~33.78‰ with -32.94‰ on average, carbon isotope of ethane ($\delta^{13}$C$_{C_2H_6}$) is about -11.03‰~15.18‰ with -12.84‰ on average, carbon isotope of carbon dioxide ($\delta^{13}$CO$_2$) is about -10.47‰~13.86‰ with -11.84‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -161.59‰~184.66‰ with -172.71‰ on average. The carbon isotope of methane ($\delta^{13}$C$_{CH_4}$) from the desorbed gas of No.7 coal seam in the study area is about -27.62‰~38.37‰ with -33.21‰ on average, carbon isotope of ethane ($\delta^{13}$C$_{C_2H_6}$) is about -9.87‰~13.02‰ with -11.30‰ on average, carbon isotope of carbon dioxide ($\delta^{13}$CO$_2$) is about -9.31‰~13.49‰ with -11.00‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -157.38‰~172.38‰ with -137.27‰ on average. The carbon isotope of methane ($\delta^{13}$C$_{CH_4}$) from the desorbed gas of No.8-1 coal seam in the study area is about -27.76‰~33.89‰ with -30.35‰ on average, carbon isotope of ethane ($\delta^{13}$C$_{C_2H_6}$) is about -8.16‰~12.70‰ with -10.45‰ on average, carbon isotope of carbon dioxide ($\delta^{13}$CO$_2$) is about -8.81‰~14.84‰ with -10.47‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -156.22‰~174.02‰ with -164.66‰ on average. The carbon isotope of methane ($\delta^{13}$C$_{CH_4}$) from the desorbed gas of No.8-2 coal seam in the study area is about -27.49‰~32.36‰ with -29.94‰ on average, carbon isotope of ethane ($\delta^{13}$C$_{C_2H_6}$) is about -7.89‰~12.76‰ with -10.14‰ on average, carbon isotope of carbon dioxide ($\delta^{13}$CO$_2$) is about -6.68‰~12.98‰ with -10.21‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -152.51‰~173.61‰ with -163.00‰ on average. The carbon isotope of methane ($\delta^{13}$C$_{CH_4}$) from the desorbed gas of No.9 coal seam in the study area is about -27.50‰~32.47‰ with -29.51‰ on average, carbon isotope of ethane ($\delta^{13}$C$_{C_2H_6}$) is about -7.90‰~12.44‰ with -9.66‰ on average, carbon isotope of carbon dioxide ($\delta^{13}$CO$_2$) is about -7.69‰~12.49‰ with -9.98‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -145.97‰~171.50‰ with -158.48‰ on average. The carbon isotope of methane ($\delta^{13}$C$_{CH_4}$) from the desorbed gas of No.15 coal seam in the study area is about -28.60‰~31.73‰ with -28.88‰ on average, carbon isotope of ethane ($\delta^{13}$C$_{C_2H_6}$) is about -7.80‰~11.10‰ with -9.28‰ on average, carbon isotope of carbon dioxide ($\delta^{13}$CO$_2$) is about -7.88‰~15.90‰ with -9.79‰ on average, hydrogen isotope of methane ($\delta^{13}$CH$_4$) is about -142.64‰~163.30‰ with -152.69‰ on average.

Table 1. Average composition and stable isotope of desorbed gas and gas from coal seam and goaf in study area

| Sampling point | Average gas composition (%) | Average carbon isotope values (%) | Average hydrogen isotope values(‰) |
|----------------|------------------------------|----------------------------------|-----------------------------------|
| Coal seam NO.3| CH$_4$ 78.80  C$_2$H$_6$ 0.01  N$_2$ 16.10  CO$_2$ 5.08 | $\delta^{13}$C$_{CH_4}$ -34.22  $\delta^{13}$C$_{C_2H_6}$ -13.76  $\delta^{13}$CO$_2$ -12.61 | $\delta^{13}$CH$_4$ -175.05 |
| Coal seam NO.5| CH$_4$ 83.24  C$_2$H$_6$ 0.02  N$_2$ 14.13  CO$_2$ 2.62 | $\delta^{13}$C$_{CH_4}$ -32.94  $\delta^{13}$C$_{C_2H_6}$ -12.84  $\delta^{13}$CO$_2$ -11.84 | $\delta^{13}$CH$_4$ -172.70 |
| Coal seam NO.7| CH$_4$ 88.16  C$_2$H$_6$ 0.02  N$_2$ 10.23  CO$_2$ 1.59 | $\delta^{13}$C$_{CH_4}$ -31.21  $\delta^{13}$C$_{C_2H_6}$ -11.30  $\delta^{13}$CO$_2$ -10.99 | $\delta^{13}$CH$_4$ -167.27 |
| Coal seam NO.8-1| CH$_4$ 93.78  C$_2$H$_6$ 0.03  N$_2$ 5.14  CO$_2$ 1.05 | $\delta^{13}$C$_{CH_4}$ -30.35  $\delta^{13}$C$_{C_2H_6}$ -10.45  $\delta^{13}$CO$_2$ -10.47 | $\delta^{13}$CH$_4$ -164.65 |
| Coal seam NO.8-2| CH$_4$ 95.58  C$_2$H$_6$ 0.03  N$_2$ 3.48  CO$_2$ 0.92 | $\delta^{13}$C$_{CH_4}$ -29.94  $\delta^{13}$C$_{C_2H_6}$ -10.14  $\delta^{13}$CO$_2$ -10.21 | $\delta^{13}$CH$_4$ -163.00 |
3.2. Analysis on gas source in goaf by numerical calculation

Isotope values of different maker in mixed gas is closely related to the blending ratio of each gas source\cite{15-17}. Started from the definition of carbon isotope according to the conservation of mass and volume ratio, Xianzhi Gao, etc. and Zongqi Wan, etc. derived a formula to calculate the carbon isotope value in the mixed gas\cite{14,17}, such as Eq.1.
Note: a, b, c, d, e, f were defined as $\delta^{13}$CH$_4$ vs $\delta^{13}$C$_2$H$_6$, $\delta^{13}$CH vs $\delta^{13}$CO$_2$, $\delta^{13}$CH vs $\delta^{13}$C$_2$H$_6$ vs $\delta^{13}$CO$_2$, $\delta^{13}$C$_2$H$_6$ vs $\delta^{13}$CH, $\delta^{13}$CO$_2$ vs $\delta^{13}$CH$_4$, $\delta^{13}$H vs $\delta^{13}$C, isotope units are ‰ respectively.

**Fig. 1.** Standard chart of stable carbon and hydrogen isotope in gas from desorbed gas of different coal seam and lagged traverse

Take lagged traverse No.1 in the goaf as an example, combine with the gas component concentration and the hydrocarbon isotope date in the coal seam of lagged traverse No.1, No.3 to No.8-2 coal seam gas component concentration and gas carbon and hydrogen isotopes eigenvalue. The $\delta^{13}$CH$_4$, $\delta^{13}$C$_2$H$_6$, $\delta^{13}$CO$_2$, $\delta^{13}$CH$_4$ of the mixed gas in goaf expressed by Eq. 4 to 7, respectively.

\[
\begin{align*}
-32.78\% &= [V_1 \times 78.80\% \times (-34.22\%) + V_2 \times 83.24\% \times (-32.94\%) + V_3 \times 88.16\% \times (-31.21\%) + V_4 \times 93.22\% \times (-29.94\%)] / [(V_1 + \ldots + V_5) \times 83.60\%] \\
-12.36\% &= [V_1 \times 0.01\% \times (-13.76\%) + V_2 \times 0.02\% \times (-12.84\%) + V_3 \times 0.02\% \times (-11.30\%) + V_4 \times 0.03\% \times (-10.45\%) + V_5 \times 0.03\% \times (-10.14\%)] / [(V_1 + \ldots + V_5) \times 9.02\%] \\
-12.20\% &= [V_1 \times 5.08\% \times (-12.61\%) + V_2 \times 2.61\% \times (-11.30\%) + V_3 \times 1.59\% \times (-10.99\%) + V_4 \times 1.10\% \times (-10.97\%) + V_5 \times 0.92\% \times (-10.21\%)] / [(V_1 + \ldots + V_5) \times 3.22\%] \\
-171.58\% &= [V_1 \times 83.20\% \times (-175.05\%) + V_2 \times 83.24\% \times (-172.70\%) + V_3 \times 88.16\% \times (-163.77\%) + V_4 \times 93.78\% \times (-164.65\%) + V_5 \times 95.85\% \times (-163.00\%)] / [(V_1 + \ldots + V_5) \times 83.60\%]
\end{align*}
\]

By the above, set the total volume of the gas from different gas sources for unit volume 1, can use Eq. 8:

\[
V_1 + V_2 + V_3 + V_4 + V_5 = 1
\]

According to Eq. 4 to Eq. 8 established a quinary linear equation group, simplified the equation group and got the following equations:

\[
\begin{align*}
0 &= 43.22V_1 - 1.22V_2 - 11.54V_3 - 106.21V_4 - 121.78V_5 \\
0 &= 0.041372V_1 - 0.00728V_2 - 0.04168V_3 - 0.05317V_4 - 0.07545V_5 \\
0 &= -24.84V_1 + 8.26333V_2 + 21.81288V_3 + 28.32362V_4 + 29.93562V_5 \\
0 &= 550.23V_1 - 30.57V_2 - 402.05V_3 - 1097.36V_4 - 1234.79V_5 \\
1 &= V_1 + V_2 + V_3 + V_4 + V_5
\end{align*}
\]

The above equations can be solved by the MATLAB software. First, artificial building coefficient matrix A and B. Then coefficient matrix A and B into MATLAB, figure out V$_1$, V$_2$, V$_3$, V$_4$ and V$_5$, the operation process and the result is shown in figure 3:

\[
A = \begin{bmatrix}
43.22 & -1.22 & -11.54 & 106.21 & 121.78 \\
0.041372 & -0.00728 & -0.04168 & -0.05317 & -0.07545 \\
-24.84 & 8.26 & 21.81 & 28.32 & 29.94 \\
550.23 & 30.57 & -402.05 & -1097.36 & -1234.79 \\
1 & 1 & 1 & 1 & 1
\end{bmatrix}
\]

\[
B = \begin{bmatrix}
0; \\
0; \\
0; \\
1
\end{bmatrix}
\]
The operation results of lagged traverse No.1 by MATLAB

Figure out $V_1$, $V_2$, $V_3$, $V_4$ and $V_5$ were 0.38, 0.37, 0.12, 0.08 and 0.05, respectively. Got the proportion of the gas from No.3 to No.8-2 coal seam desorbed gas were 38%, 37%, 12%, 8% and 5%, respectively. Repeated the calculation process and substituted the gas component and gas hydrocarbon isotope values of lagged traverse No.2 into the computational model (refer to Eq.3). Then set up simultaneous equations and simplified coefficient equation group. Used MATLAB to solve and got the proportion of the gas from No.3, No.5, No.7, No.8-1 and No.8-2 coal seam desorbed gas were 22%, 36%, 24%, 12% and 6%, respectively.

4 Conclusions

(1) The desorbed gas components and stable carbon isotope from the same coal seam exist certain differences in Sihe coal mine. There are significant differences in the desorbed gas components and stable carbon isotope from different coal seam. The gas methane concentration increased along with the deepening of coal seam depth, and the gas hydrocarbon isotope along with the deepening of coal seam depth showed the characteristic of eccentric weight.

(2) The mathematical calculations suggested that the proportion of the gas in lagged traverse No.1 from No.3 to No.8-2 coal seam desorbed gas were 38%, 37%, 12%, 8% and 5%, respectively. The proportion of the gas in lagged traverse No.2 from No.3, No.5, No.7, No.8-1 and No.8-2 coal seam desorbed gas were 22%, 36%, 24%, 12% and 6%, respectively.

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