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Research on the Main Controlling Factors of CO_2 Stability in Closed Coal Goafs Based on Gray Theory

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Abstract: The use of closed goafs to store CO_2 is one of the important ways to capture, utilize and store carbon, and play an important role in helping to achieve the "dual carbon" goal. Aiming at the problem of identifying the main controlling factors of CO_2 sequestration stability in closed coal mine goafs, the stability influencing factors are systematically selected from three aspects: hydrogeological conditions, goaf occurrence characteristics and cap conditions, and the CO_2 sequestration per unit area is used as a reference sequence, and the gray correlation analysis method is comprehensively used to carry out research. The results show that the gray correlation of the influencing factors of CO_2 stability in goaf is as follows: cover thickness, burial depth, cap continuity, lithology of overlying rock layer, number of cap assemblies, porosity, groundwater salinity, ground temperature gradient, goaf collapse degree and permeability. According to the ranking results, it is concluded that the two factors with the highest correlation degree and stronger correlation between cover thickness and burial depth are the main controlling factors affecting the stability of CO_2 sequestration in the goaf.

Keywords: Goaf; Carbon dioxide storage; Main controlling factors; Gray correlation analysis

1. Introduction

The emergence of carbon capture, storage, and utilization (CCUS) technology provides a feasible technical path for CO_2 recovery and recycling [1], among which CO_2 storage is one of the important links to help achieve the "dual carbon" goal. As of 2020, there are about 12,000 closed or abandoned mines in the country, and it is expected to reach 15,000 by 2030. There are still a large number of rock pores and fissures in the goaf left after the mine closure, which provides a technical way for the geological sequestration of carbon dioxide [2]. Carrying out research on the reuse of closed resources and sequestering CO_2 in goafs is conducive to the in-depth development of closed mine resources [3], and goafs, as an unconventional geological sequestration, have great storage potential and utilization value [4].

In terms of the influencing factors of sequestration feasibility, scholars have mainly studied from two perspectives: Guo[5] et al. analyzed the influence mechanism of porosity, CO_2 injection rate, CO_2 sequestration depth and other factors on surface deformation using FLAC^{3D} simulation; Ding Yang [6] took the goaf of a coal mine in northern Shaanxi as an example to simulate the leakage diffusion law and concentration distribution of CO_2 leakage under different leakage velocities and wind speeds. Li Shugang [7] used the self-developed geological storage experimental platform to reveal the coupling relationship between CO_2–formation water interface effect and dissolved mass transfer. Meng Henglei [8] analyzed the influence of the original geological structures such as faults, joints, and fractures in the early stage, the elevator shafts, roadways and other connecting channels formed by human factors in the later stage, and the fracture zones formed by mining activities on the sealing of CO_2 sequestration in the goaf. Lu [9] took CO_2 geological storage in Tianjin as an example to study the effects of cap, site earthquake, hydrogeological conditions, and ground geological conditions on the stability of carbon dioxide geological storage. Qian [10] mainly used nine factors such as geological structure, caprock lithology, permeability, and stability coefficient as stability evaluation indicators when selecting the influencing factors of sequestration stability.

In order to clarify the mechanism of CO_2 storage in the closed goaf of coal mines and obtain the main influencing factors of CO_2 storage stability in goafs, the gray correlation method is used to screen the main controlling factors based on the gray theory.

2. Materials and Methods

Research on the influencing factors of CO_2 stability in goaf storage

In the process of CO_2 sequestration in the goaf, the injected CO_2 will increase the pressure in the goaf and gradually diffuse and migrate, and under the action of pressure and buoyancy, the injected carbon dioxide may migrate to shallow groundwater or the atmosphere along the leakage channel, causing leakage. This causes surface deformation and affects the stability of storage. Therefore, combined with the existing research data, the stability influencing factors are selected from three aspects: hydrogeological conditions, goaf occurrence characteristics and cap conditions, which are used to evaluate the stability of CO_2 sequestration in goafs, and the main controlling factors affecting the stability of CO_2 sequestration in goafs are determined.

Hydrogeological conditions

In the process of CO_2 sequestration in goafs, CO_2 will react with groundwater, affecting the stability of sequestration, and different hydrological conditions will also have different effects on CO_2 sequestration effects. In addition, hydrogeological conditions have a controlling effect on the dissolution and sequestration of CO_2 and mineralization. In the short term, solubility is an important parameter for estimating dissolution and storage, and low temperature, high pressure and low salinity can more effectively strengthen the dissolution and storage efficiency and improve the CO_2 storage. When the reservoir pressure is constant, there is a negative correlation between temperature and CO_2 solubility, and in the long run, appropriate high salinity can form more new mineral precipitation, which is more beneficial to the mineralization and sequestration of CO_2. This study focuses on stability, and prioritizes factors that have been proven to have a significant impact on CO_2 sequestration and can be quantified or available in the coal mine closure scenario. At the same time, with reference to the specification of "Specification for Leakage Risk Assessment of Carbon Dioxide Geological Utilization and Storage Projects", the ground temperature gradient and salinity are selected as representative indicators.

(1) Geothermal gradient

When the reservoir pressure is constant, there is a negative correlation between temperature and CO_2 solubility, and a higher geothermal gradient may lead to an increase in the temperature in the goaf, which in turn affects the solubility of carbon dioxide, thereby reducing the density and viscosity of carbon dioxide, and affecting the safety of sequestration [11].

(2) Groundwater salinity

Studies have shown that formation water salinity has a significant effect on the interfacial tension and solubility of CO_2 [7]. High salinity increases interfacial tension, reduces CO_2 solubility, and affects the sequestration effect. However, in the long run, the appropriate high salinity is rich in calcium and magnesium plasma, which can promote carbonate mineral precipitation, enhance mineralization capture, and achieve stable CO_2 sequestration. According to previous studies, the influencing factors affecting the storage stability of goafs in hydrogeological conditions are quantitatively graded, see Table 1.

Table 1: Quantitative grading table of hydrogeological conditions

	Very stable	**Stability**	**More stable**	**unstable**
Geothermal gradient X_1（℃/100 m）	2	2-3	3-4	4
Groundwater salinity X_2（g/L）	30-50	10-30	3-10	50

Goaf Occurrence Characteristics

As a CO_2 storage site, after the early mining activities, the original stress of the goaf is destroyed, the fracture evolution and seepage law are changed, and the migration process after carbon dioxide injection is related to these factors.

(1) Bury deep

In CO_2 sequestration, it is usually injected into deep burial in a supercritical state, which ensures the stability after injection and increases the sequestration density. Theoretically, CO_2 should be injected into the depth of the ground below 800 m in a supercritical state, and with the increase of burial depth, the deformation resistance of the overlying rock layer is enhanced, the surface displacement is reduced, the deformation duration is shortened [12-14], and the storage is more stable.

(2) Goaf collapse degree index

The degree of goaf collapse is determined by the maximum subsidence value, mining thickness, and coal seam inclination angle [10], and when the maximum subsidence value is equivalent to the mining value, the goaf collapse degree index is the sinking coefficient. The lower the sinking index, the lower the roof, the less large-scale collapse of the roof, the structure is intact, which can effectively block the upward migration of carbon dioxide, reduce the risk of leakage, and improve the stability of sealing. The low subsidence index also indicates that the surrounding rock in the goaf is relatively stable, the deformation is small, the coal-rock structure is relatively intact, and the water environment is relatively stable, which is conducive to enhancing the stability of carbon dioxide sequestration.

(3) Permeability

When the burial depth and cover thickness are in the stable range, the permeability directly determines the sequestration stability of CO_2. According to Darcy's law, the worse the permeability of the reservoir, the greater the flow resistance that CO_2 needs to overcome, and the weaker its free migration ability [15]. It can be seen that the permeability conditions in the goaf have an important impact on the CO_2 sequestration effect, and for the reservoir, the worse the permeability, the stronger the flow power of the sequestered CO_2 is required to flow freely. Therefore, the permeability conditions of goaf play an important role in the sequestration of carbon dioxide.

(4) Porosity

High porosity means that there is more pore space in the coal rock, and the permeability of the coal rock is relatively good, and it can also reduce the pressure accumulation generated during the carbon dioxide injection process and avoid the damage of the surrounding rock in the goaf caused by excessive pressure. The saturated adsorption capacity of coal for carbon dioxide increases with the increase of specific surface area, and high porosity is often accompanied by the increase of specific surface area, which is conducive to the adsorption and storage of carbon dioxide. Too low porosity will limit the pore space in the goaf, limit the sequestration capacity, and also increase the pressure during the injection process, which may lead to local stress concentration, affect the stability of the goaf, and then reduce the stability of the goaf. The quantitative grading is shown in Table 2.

Table 2: Quantitative grading table of occurrence characteristics in goafs

	Very stable	Stability	More stable	unstable
Buried deep X_3（m）	＞3500	1500-3500	800-1500	＜800
Penetration X_4（md）	0.1	0.1-1	1-10	10
Porosity X_5（%）	20	15	10	5
Goaf Collapse Degree Index X_6	0.20	0.40	0.60	0.94

Cap conditions

For the closure of coal mine goafs for CO_2 sequestration, good cap conditions are the prerequisites for ensuring the long-term stability of CO_2 sequestration in goafs, and one or even more sets of stable caps can effectively block CO_2 upward escape [16].

(1) Lithology of the upper overburden

The upper overburden is the key sealing barrier for CO_2 sequestration in goafs, and its lithological characteristics directly affect the stability of the storage system. Generally speaking, the larger the porosity of the cover layer, the worse the sealing performance. Compared with high-porosity sandstone, low-porosity limestone can provide more stable storage conditions when used as a capstone.

(2) Cover thickness

The thickness of the cover is the main evaluation index of sealing, and when the cover is thin, it is often unstable, which is not conducive to large-scale carbon dioxide geological sequestration. Previous experiments have obtained empirical data that the coal seam roof with a thickness of 20~300 m can be used as an effective sealing thickness[17]. In this study, the grading criteria of cover thickness index are quantitatively graded with reference to relevant data.

(3) The number of cover combinations

The CO_2 sequestration in the goaf must have good cover conditions to prevent direct CO_2 leakage. In general, a single geological cap that meets the requirements of containment capacity can effectively sequester CO_2. If there is a multi-layer closed combination on the direct cover layer to form a multi-level cap structure, the overlying multi-level cap structure can achieve secondary interception or well sealing after the failure of the direct cover to prevent carbon dioxide from leaking further upwards after breaking through the direct cover. Therefore, the spatial combination of the cover layer is closely related to the sealing ability of the cap, and the better the quality and quantity of the secondary cap, the stronger the plugging ability, the higher the long-term safety, and the lower the risk of CO_2 geological sequestration.

(4) Continuity of the cover

The continuity of the cap layer directly affects the stability of large-scale CO_2 sequestration. The larger the thickness of the cover layer, the wider the distribution area, and the better the horizontal distribution continuity, the higher the sealing stability. When CO_2 is stored in the goaf, it is necessary to prevent unenclosed boreholes left over from coal exploration, and to avoid the development of faults and fractures in the cap. The cover is complete and there are no faults to score 1 point, the cap is generally intact and there are a small number of cracks to score 0.8 points, the presence of small faults is 0.6 points, and the integrity of the cap is poor, and the existence of a large number of faults is 0.3 points.

Table 3: Quantitative grading table for cover conditions

	Very stable	Stability	More stable	unstable
Lithology of the overlying rock layer X_7	2	1.5	0.75	0.5
The cumulative thickness of the cover layer X_8 (m)	300	150-300	100-150	20
Number of cover combinations X_9	3	2	1	0
Cover continuity X_{10}	1	0.8	0.6	0.3

Research on the main controlling factors of CO2 storage in goaf based on gray theory

In order to clarify the influence of each factor on the stability of CO_2 sequestration, based on the gray system theory, the influence of each factor on the stability of CO_2 sequestration was quantitatively analyzed by the gray correlation analysis method. Gray correlation analysis is to calculate the main relationship between the target factor and the corresponding factor, find out the factors that have the greatest influence on the target factor, quantify the degree of relationship between them, obtain the correlation value and ranking, and quantitatively explain the interconnection between the changing factors and the corresponding factors[18]. Through gray correlation analysis, it is helpful to quantitatively explain the importance of different influencing factors to the stability of CO_2 sequestration in closed goafs.

Determine the sequence matrix

When performing gray correlation analysis, first determine the reference feature sequence and the comparison sequence:

The reference sequence is:

$$X_0=\{x_0(1), x_0(2), \ldots, x_0(k), \ldots, x_0(n)\}$$

The comparison sequence is:

$$X_i=\{x_i(1), x_i(2), \ldots, x_i(k), \ldots, x_i(n)\}$$

Where $x_0(k)$ is the reference sequence is data at the kth trial, $x_i(k)$ is the ith comparison sequence was at the kth trial.

In goaf CO_2 storage projects, it is necessary to pay attention not only to the total amount of carbon dioxide sequestration, but also to consider the sequestration volume per unit area, because this can more intuitively reflect the utilization efficiency of the storage space and the uniformity of the sequestration effect. Taking the storage volume per unit area as a reference sequence, the stability and reliability of the sealing project can be evaluated more comprehensively, and the sealing effect is better when it reaches 150×104 t/km^2.

Therefore, this paper quantifies and grades the CO_2 sequestration per unit area as the reference sequence X_0, and selects ten factors affecting the stability of CO_2 sequestration in goaf from the aspects of hydrogeological conditions, goaf itself factors and cover conditions, and quantifies and classifies them as a comparison sequence to obtain the corresponding sequence matrix. See Table 4.

Table 4: Sequence matrix

	Y_1	Y_2	Y_3	Y_4
X_0	150	50-150	10-50	10
X_1	2	2-3	3-4	4
X_2	30-50	10-30	0-10	50
X_3	3500	1500-3500	800-1500	800
X_4	0.1	0.1-1	1-10	10
X_5	20	15	10	5
X_6	0.20	0.40	0.60	0.94
X_7	2	1.5	0.75	0.5
X_8	300	150-300	100-150	20
X_9	3	2	1	0
X_{10}	1	0.8	0.6	0.3

Calculation of gray correlation

(1) Data preprocessing

Since there are interval values in the data, gray correlation analysis requires a specific numerical series and cannot be used directly, so the interval values need to be processed to ensure that the data can be quantified and processed. Therefore, it is necessary to extract the values suitable for gray correlation analysis from the interval to ensure the uniformity of the data and facilitate subsequent use. For the comparison series containing interval values, the interval midpoint method is used for processing, and the midpoint value of each interval is taken as the representative value of the reference series, and the processed results are shown in Table 5.

Table 5: Data preprocessing sequence matrix

	Y_1	Y_2	Y_3	Y_4
X_0	150	100	30	10
X_1	2	2.5	3.5	4
X_2	40	20	5	50
X_3	3500	2500	1150	800
X_4	0.1	0.55	5.5	10
X_5	20	15	10	5
X_6	0.20	0.40	0.60	0.94
X_7	2	1.5	0.75	0.5
X_8	300	225	125	20
X_9	3	2	1	0
X_{10}	1	0.8	0.6	0.3

(2) Dimensionless processing

The relative index data of each influencing factor are different, and the numerical differences are large, making it difficult to directly compare them. In order to facilitate calculation and dimensionless processing of data, gray correlation analysis often uses the initial value method or mean method to standardize the data, eliminate dimensional influence, and make the factors comparable.

The initial value method first takes a non-empty number, and then divides it by this value with other data, which is suitable for analysis of data with strong trend or regularity. The mean method represents the general level of the set of data by summing and dividing by the number of data to obtain the average value of the data. A single value (mean) can quickly summarize the central trend of a set of data, which can resist the influence of individual outliers to a certain extent. For a large amount of data, the fluctuation of individual data has relatively little impact on the mean, so that the mean can reflect the overall characteristics of the data relatively stably, and the entire set of data values is required to be positive. Due to the large fluctuation of data in the initial sequence matrix, the data are dimensionless by the mean method, and the dimensionless processing method of the mean method is to divide the data values in the whole sequence by the average value of the sequence to eliminate the dimensional influence, and the processing results are shown in Table 6.

Table 6: Dimensionless processing of data tables

	Y_1	Y_2	Y_3	Y_4
X_0	2.069	1.379	0.414	0.138
X_1	0.667	0.833	1.167	1.333
X_2	1.391	0.696	0.174	1.739
X_3	0.025	0.136	1.362	2.477
X_4	1.600	1.200	0.800	0.400
X_5	0.374	0.748	1.121	1.757
X_6	1.684	1.263	0.632	0.421
X_7	1.829	1.371	0.571	0.229
X_8	1.791	1.343	0.746	0.119
X_9	2.000	1.333	0.667	0.000
X_{10}	1.600	1.200	0.800	0.400

(3) Calculation of data correlation coefficients

The so-called degree of correlation is actually a measure of the shape difference between the curves composed of various data, and the calculation of the correlation coefficients in the data is mainly determined by the difference between these curves.

For the reference series X0, there are n comparison series $X_1, X_2, ..., X_n$;

For indicator k, the difference between the comparison sequence xi and the reference sequence x0 needs to be calculated:

$$\Delta_i(k) = | x_i(k) - x_0(k) | \tag{1}$$

The correlation coefficients of the data are:

$$\xi_i(k) = \frac{\Delta(\min) + \rho\Delta(\max)}{\Delta_i(k) + \rho\Delta(\max)} \tag{2}$$

(4) Calculation of correlation degree

In order to measure the correlation between series as a whole, it is necessary to time average the correlation coefficients to obtain the overall correlation. The calculation of gray correlation is mainly to use weighted average to process the correlation coefficient:

$$r_i = \frac{1}{m}\sum_{k=1}^{m}\xi_i(k) \tag{3}$$

where r_i is the correlation of the comparison series, the closer the r_i value is to 1, the better the correlation with the comparison sequence.

3. Results & Discussion

The main control factor is determined

The correlation coefficients obtained by bringing the dimensionless data into the formula are shown in Table 7.

Table 7: Correlation coefficient results

X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}
0.458	0.637	0.798	0.366	0.719	0.411	0.758	0.813	0.799	0.798
0.686	0.635	0.914	0.488	0.873	0.654	0.916	0.977	0.768	0.833
0.612	0.835	0.885	0.556	0.757	0.627	0.849	0.784	0.856	0.757
0.498	0.425	0823	0.336	0.822	0.422	0.811	0.991	0.757	0.822

The correlation degree calculation and weighting process of the 10 evaluation objects were finally obtained, and the results are shown in Table 8.

Table 8: Correlation results

X_1	X_2	X_3	X_4	X_5	X_6	X_7	X_8	X_9	X_{10}
0.564	0.633	0.871	0.436	0.793	0.529	0.833	0.905	0.795	0.845

In the determination of the main control factor, it is mainly selected by the ranking of the correlation degree, the correlation degree represents the similarity between the comparison series and the reference sequence, the correlation value is between 0~1, the larger the value, the closer the relationship between the comparison sequence and the reference sequence, the stronger the correlation, and the higher its evaluation. Combined with the obtained correlation results, all evaluation items are sorted to obtain each ranking. The ranking of correlation obtained from this is shown in Table 9.

Table 9 Relevance sorting

X_8	X_3	X_{10}	X_7	X_9	X_5	X_2	X_1	X_6	X_4
0.905	0.871	0.845	0.833	0.795	0.793	0.633	0.564	0.529	0.436

From the ranking of correlation degrees in Table 9, it can be seen that the correlation degree of cover thickness X_8 is the highest, with a correlation degree of 0.905, which is significantly higher than that of other influencing factors, indicating that the cover thickness plays an important role in the stability of CO_2 sequestration in the goaf. followed by burial depth X_3, with a correlation degree of 0.871, which is also at a highly correlated level, indicating that the two have a decisive influence on the amount of CO_2 sequestration per unit area, and the relationship with the sequestration amount is also closer and stronger. Therefore, it is shown that cover thickness and burial depth play an important role in stabilizing and reducing the risk of leakage, and the above two factors are taken as the main controlling factors affecting the stability of CO_2 sequestration in goafs.

From the screening results of the main control factors, it can be seen that in the study of the influencing factors of CO_2 sequestration stability in goafs, the cap conditions occupy an important position, and the closure of the cap is a prerequisite for CO_2 sequestration in goafs. As a natural barrier to block the upward migration of CO_2, the thickness of the cover layer directly determines the length of the CO_2 transport path, capillary sealing pressure and mechanical integrity, and is the physical basis for maintaining the pressure balance of the storage system and preventing vertical leakage. The burial depth indirectly affects the injection efficiency, migration and long-term storage mechanism through the change of temperature and pressure environment of the reservoir layer and the change of CO_2 thermophysical properties.

4. Conclusion

(1) The influence of various factors on the stability of CO_2 sequestration in goaf is discussed from three aspects: hydrogeological conditions, goaf occurrence characteristics and cap conditions, and a quantitative grading table of influencing factors is obtained for the construction of sequence matrix in gray correlation analysis.

(2) The correlation ranking between each influencing factor and CO_2 sequestration per unit area was obtained by the calculation of gray correlation: cover thickness> burial depth> cap continuity> lithology of overlying rock layers> number of cap combinations> porosity> groundwater salinity> ground temperature gradient> goaf collapse degree> permeability.

(3) According to the ranking results, the two factors with the highest correlation degree and stronger correlation between cover thickness and burial depth were screened out as the main controlling factors affecting the stability of CO_2 sequestration in the goaf.

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