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Abstract

Coal is a porous medium. Due to the large number of pores in coal and the pore size on its surface, usually ranging from millimeter to nanometer, it is difficult to measure and analyze the microscopic pore structure of coal. In order to investigate the effect of the microscopic pore structure of coal on its spontaneous combustion tendency, coal samples from different coal mines of the Kailuan Group were selected as the research objects, and the data of the microscopic pore distribution of three different coal samples were measured by using mercury injection apparatus. The regression analysis of microscopic pore data of coal samples obtained in the mercury injection experiment shows that the correlation coefficients of the regression curves are all greater than 0.94 and the fitting degree is good, indicating that there is a good correlation between the pressure, mercury intake and pore size of the coal samples, indicating that the fractal dimension of pore distribution is very effective. The fractal dimension is generally between 2 and 3, indicating that the microscopic pores of coal samples have good fractal characteristics and meet the fractal theory to describe the distribution characteristics of microscopic pores in porous media. Through the simulation system of natural combustion of coal, the simulation experiment of temperature rise oxidation of different coal samples (gas coal, fat coal, and coke coal) was carried out, and the curve of the concentration of gas products CO and CO2 in the process of temperature rise and oxidation of coal samples was drawn in the experiment. The experimental results show the relationship between the distribution structure of coal pores and its spontaneous combustion tendency, and the coal with a good distribution dimension has a stronger combustion tendency

Keywords

spontaneous combustion tendency, mercury injection, fractal dimension, indicator gases

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Study on the Effect of Coal Microscopic Pore Structure to its Spontaneous Combustion Tendency

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Abstract

Coal is a porous medium. Due to the large number of pores in coal and the pore size on its surface, usually ranging from millimeter to nanometer, it is difficult to measure and analyze the microscopic pore structure of coal. In order to investigate the effect of the microscopic pore structure of coal on its spontaneous combustion tendency, coal samples from different coal mines of the Kailuan Group were selected as the research objects, and the data of the microscopic pore distribution of three different coal samples were measured by using mercury injection apparatus. The regression analysis of microscopic pore data of coal samples obtained in the mercury injection experiment shows that the correlation coefficients of the regression curves are all greater than 0.94 and the fitting degree is good, indicating that there is a good correlation between the pressure, mercury intake and pore size of the coal samples, indicating that the fractal dimension of pore distribution is very effective. The fractal dimension is generally between 2 and 3, indicating that the microscopic pores of coal samples have good fractal characteristics and meet the fractal theory to describe the distribution characteristics of microscopic pores in porous media. Through the simulation system of natural combustion of coal, the simulation experiment of temperature rise oxidation of different coal samples (gas coal, fat coal, and coke coal) was carried out, and the curve of the concentration of gas products CO and CO_2 in the process of temperature rise and oxidation of coal samples was drawn in the experiment. The experimental results show the relationship between the distribution structure of coal pores and its spontaneous combustion tendency, and the coal with a good distribution dimension has a stronger combustion tendency.

Keywords: spontaneous combustion tendency, mercury injection, fractal dimension, indicator gases

1. Introduction

In the past research on spontaneous combustion of coal, studies on coal spontaneous combustion index gas and microscopic physical and chemical changes have been carried out, respectively explaining coal's spontaneous combustion and oxidation process from macroscopic and microscopic perspectives [1–8]. Most of them consider the effect of the macroscopic structure of the coal body on the spontaneous combustion of coal, while the effect of the microscopic pore structure on the spontaneous combustion of coal is more negligible [9–14]. In addition, the research on the combination of the macroscopic appearance of spontaneous combustion of coal, that is, the generation of indicator gas and the change of microstructure, has also begun to be further studied. In the long process of geological evolution, a large number of plant remains were transformed into coal through complex biochemical, geochemical, and physicochemical reactions [15–19]. Under the influence of various metamorphism, there are differences in the overall morphology, porosity, fissure development, and physical structure of coal due to the differences in coal forming age, original material, reduction degree, and genetic type. From the perspective of the coal formation process, coal is a porous rock, namely a porous medium, with microporosity accounting for most coal surface area and dual structure of matrix pores and natural fractures [20–23]. When combustibles, oxygen, and ignition sources reach certain conditions, spontaneous combustion

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of coal will occur. Under this structure, it provides conditions for the formation and storage of gas (mention coal spontaneous combustion), and influences the possibility and consequence of spontaneous combustion of coal [24, 25]. Pore structure provides storage location for gas in coal mine. The pore range ranges from less than 1 nm to hundreds of nanometers [26–30]. When the storage reaches a specific concentration, it will explode when exposed to an open flame. The pore size distribution affects the difference in the amount of adsorbed oxygen, the rate of adsorbed oxygen, and the oxygen absorption and exothermic performance of coal samples, which in turn leads to the difference in the spontaneous combustion tendency of different coal samples. About half of the coal in China is prone to spontaneous combustion, which causes a severe waste of resources. According to the statistics of relevant departments, China loses about four billion yuan every year due to the spontaneous combustion of coal [31–35]. Therefore, it is essential to study the physical structure characteristics of coal samples to understand the spontaneous combustion reaction of coal. In the field, coal samples were taken from the Donghuantuo coal mine (gas coal), the Qianjiaying coal mine (fat coal) and the Tangshan coal mine (coke coal) of the Kailuan Group, respectively. The relationship between coal structure and spontaneous combustion tendency was comprehensively studied under the conditions of different temperatures of coal samples through the following experiments: mercury injection, coal heating, and oxidation [36, 37].

2. Materials and methods

2.1. Experimental methods

We go through temperature rising oxidation experiments of coal and mercury injection tests of coal. Firstly, all kinds of coal samples were crushed, and 2.000 g of coal particles of 60–80 mesh were screened out from the crushed cinder. These screened coal particles were distributed in an oxidation furnace with a heating rate of 1.5 °C/min to start the experiment. During the experiment, it was stipulated that the gas sample was extracted every time the temperature in the furnace increased by 10–15 °C, and the collected data were analyzed and processed. Before the mercury injection experiment, it should be noted that the coal sample to be measured should be fully dried in a thermostatic drying oven and vacuumized to eliminate the interference of moisture on the experimental results (see Figs. 1 and 2).

2.2. Experimental instrument

The mercury porosimetry instrument is an Autopore IV9500 type mercury porosimeter produced by Micromeritics. The low pressure analysis pressure is 3.45–310 kPa, and the measurable pore size range is 360–3.6 μm; the high pressure analysis pressure is 227.527 kPa, and the pore size analysis can reach 5.5 nm. The pore size of coal is generally uneven, which can be divided into three categories according to pore size:

- the large pore size is greater than 1000 nm;
- the middle pore size is 100–1000 nm;
- the transition pore size is 10–100 nm;
- and the micropore size is less than 10 nm.

2.3. Experimental system

The index gas of coal spontaneous combustion refers to a kind of gas that can predict and reflect the state of coal spontaneous combustion. The suitable index gas can be determined by the temperature rising oxidation experiment of the coal sample. Because the index gas concentration changes regularly with the temperature rising, the relationship between the microstructure of the coal sample and the tendency of spontaneous combustion can be judged by the change in gas concentration. The spontaneous combustion process of coal can be divided into three stages: slow oxidation stage, accelerated oxidation stage, and severe oxidation stage; those three different stages correspond to different kinds and concentrations of gas products [38,39].

3. Results and discussion

3.1. Temperature rising oxidation experiment result analysis

Figs. 3–11, Tables 1–3 show the mercury injection experimental regression curve and relevant parameters of three coal samples of the Donghuantuo Coal Mine (gas coal), the Qianjiaying Coal Mine (fat coal) and the Tangshan coal mine (coking coal) at 30, 70, and 120 °C, respectively.

Figs. 3–5 show that the correlation coefficients of mercury injection experimental regression curves of gas coal at 30, 70, and 120 °C are 0.9798, 0.9795, and 0.9934, respectively, which are more significant than 0.94, indicating good correlation.

(a) (b)

Fig. 1. (a) Pulverized coal; (b) Three kinds of coal samples.

(a) (b)

Fig. 2. (a) Autopore IV9500 mercury porosimeter; (b) KSS-5690A series gas chromatograph.

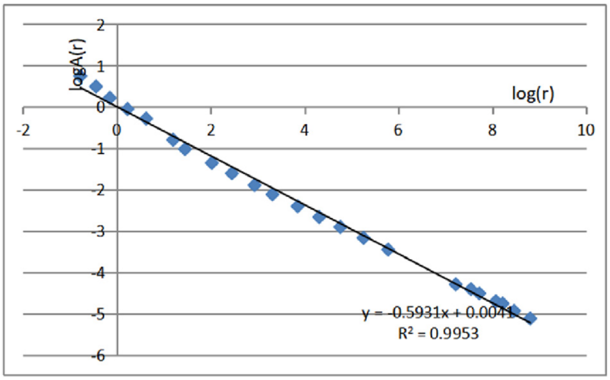

Fig. 3. Mercury injection experiment regression curve of 30 °C gas coal.

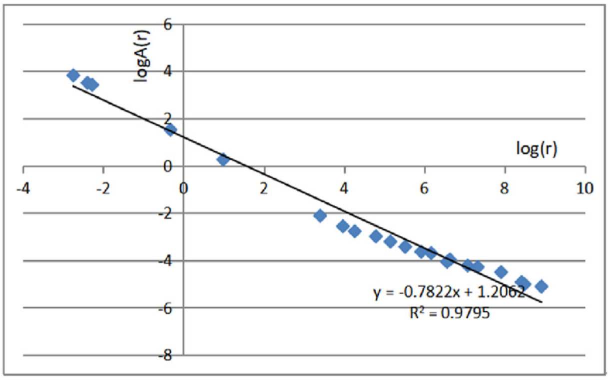

Fig. 4. Mercury injection experiment regression curve of 70 °C gas coal.

Figs. 6–8 show that the correlation coefficients of mercury injection experimental regression curves of fat coal at 30, 70, and 120 °C are 0.987, 0.9934, and 0.9832, respectively, which are more significant than 0.94, indicating good correlation.

Figs. 9–11 show that the correlation coefficients of the regression curves of mercury injection experiment of coking coal are 0.9782, 0.9805, and 0.9547, respectively, at 30, 70 and 120 °C, which shows that they have good correlation.

Table 4 shows the pore distribution of Donghuantuo Coal Mine (gas coal), Qianjiaying Coal Mine (fat coal), and Tangshan coal mine (coking coal) at 30, 70, and 120 °C.

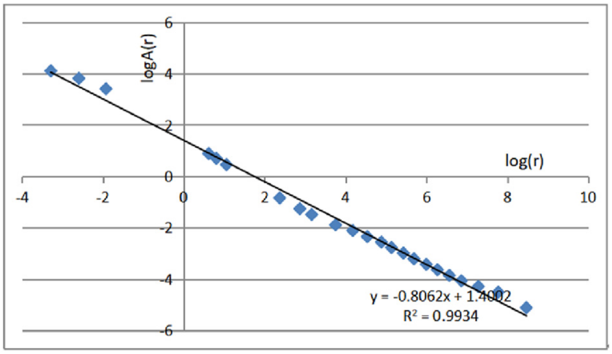

Fig. 5. Mercury injection experiment regression curve of 120 °C gas coal.

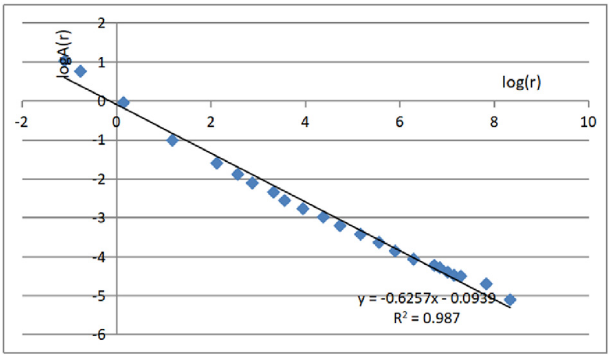

Fig. 6. Mercury injection experiment regression curve of 30 °C fat coal.

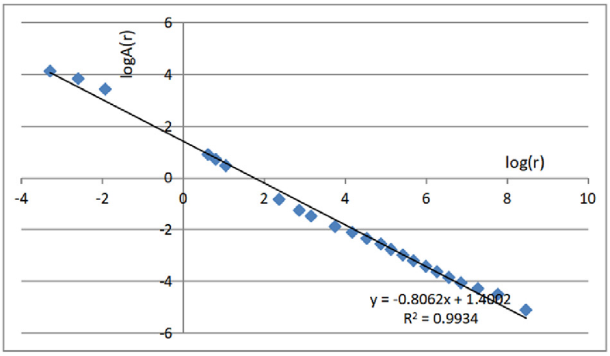

Fig. 7. Mercury injection experiment regression curve of 70 °C fat coal.

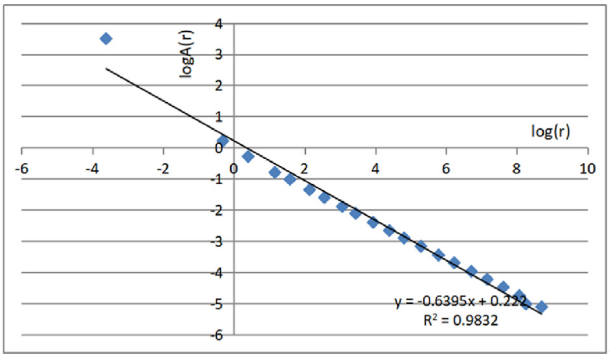

Fig. 8. Mercury injection experiment regression curve of 120 °C fat coal.

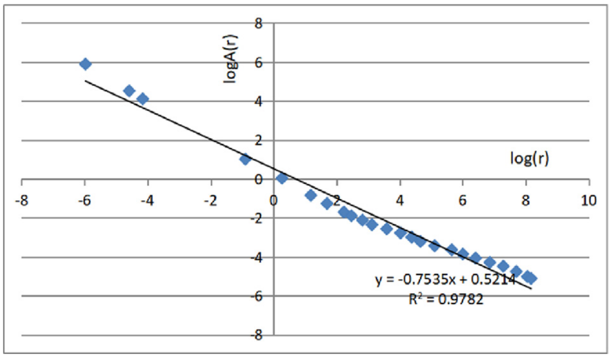

Fig. 9. Mercury injection experiment regression curve of 30 °C coking coal.

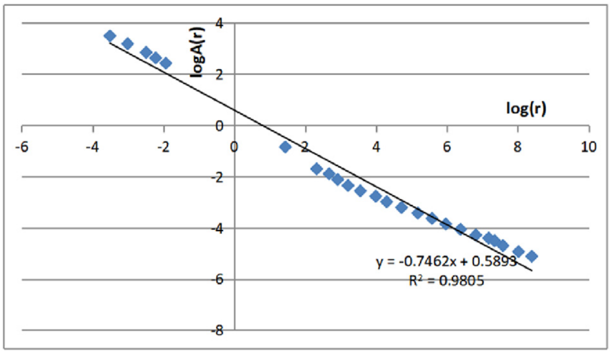

Fig. 10. Mercury injection experiment regression curve of 70 °C coking coal.

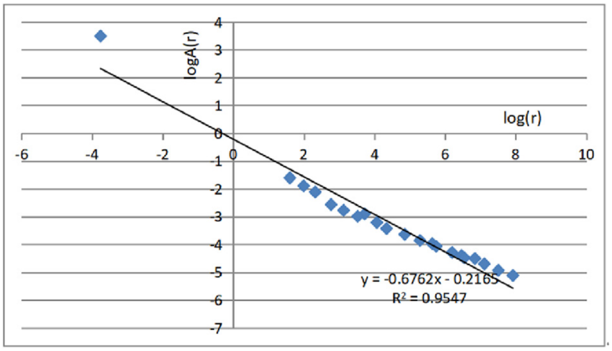

Fig. 11. Mercury injection experiment regression curve of 120 °C coking coal.

The temperature affects the fractal dimension change. In the temperature rise process, gas coal and fat coal are mainly macropore, transition pore, and micropore are the second, mesopore is the least; while coking coal is mainly transition pore, macropore and micropore are the second, mesopore is the least. With the increase in temperature, the pore distribution of all kinds of coal samples shows the trend of the number of macropores and mesopores decreasing first and then increasing, the number of micropores increasing first and then decreasing. The transition

Table 1. Gas coal regression curve parameters.

Temperature (°C)	Slope of straight line	Correlation coefficient	Fractal dimension
30	−0.5931	0.9953	2.4069
70	−0.7822	0.9795	2.2178
120	−0.8062	0.9934	2.1938

Table 2. Fat coal regression curve parameters.

Temperature (°C)	Slope of straight line	Correlation coefficient	Fractal dimension
30	−0.6257	0.9870	2.3743
70	−0.8062	0.9934	2.1938
120	−0.6395	0.9832	2.3605

Table 3. Coking coal regression curve parameters.

Temperature (°C)	Slope of straight line	Correlation coefficient	Fractal dimension
30	−0.7535	0.9782	2.2465
70	−0.7462	0.9805	2.2538
120	−0.6762	0.9547	2.3238

pores have not changed significantly. This is because during this first stage of low temperature oxidation, gases in the coal are heated and desorbed. The pressure in the closed hole in the coal is greater than the external atmospheric pressure, and the gas escapes to form nanoscale transition pores and micropores on the surface of the coal. In addition, the consumption of oxidized fine coal particles and impurities on the coal surface makes the coal surface tend to be smooth. In the second stage of low temperature oxidation, the gases that are easy to escape from the coal have been basically separated from the coal. As the temperature increases, the coal oxidation reaction rate increases, and the coal gas consumption increases. In the first stage, the nanopore transition to large holes, and the fractal dimension of coal increases.

The fractal dimension of three kinds of coal samples was analyzed by the linear regression method. According to the theory of Qu Shixian and Zhang Jianhua [40], the fractal dimension value in the three-dimensional Euclidean space is between 2 and 3. If the value of fractal dimension is close to 2, the smoother the pore surface is, the better the reservoir performance is; the closer the value of fractal dimension is to 3, the less smooth the pore surface is, the worse the reservoir performance is.

3.2. *Temperature rising oxidation experiment of coal data analysis*

Index gases carbon monoxide (CO) and carbon dioxide (CO_2) change with temperature as indicated below.

The critical temperature of CO occurrence is the temperature value when CO is detected after coal oxidation, and the change trend of this value generally increases with the deepening of coal metamorphism. The critical temperature of CO occurrence in different coal samples is 59 °C for gas coal, 66 °C for fat coal, and 81 °C for coking coal [41,42].

Figs. 12–14 show that most of the experimental coal samples detected CO and CO_2 at 50 °C and accompanied the whole temperature rising oxidation process of the experiment, and the CO_2 concentration was significantly higher than CO. When the experimental temperature reaches 50 °C, CO

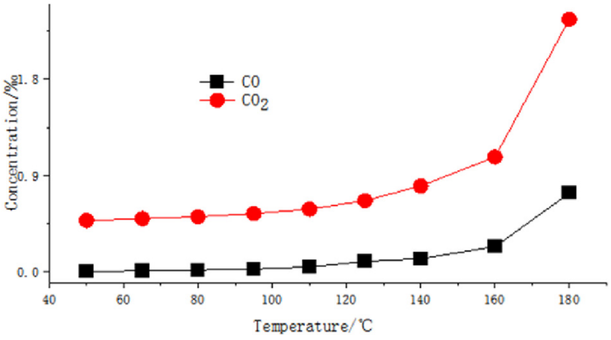

Fig. 12. Gas coal CO, CO_2 concentration changes curve with the temperature.

Table 4. Coal samples pore distribution.

Coal sample	Temperature (°C)	Pore distribution			
		Macropore (%)	Mesopores (%)	Transition hole (%)	micropore (%)
Gas coal	30	48.92	17.30	18.15	15.64
	70	35.95	8.15	31.34	24.56
	120	40.53	11.22	24.40	23.85
Fat coal	30	43.90	4.32	28.48	23.30
	70	38.56	3.98	26.37	31.09
	120	43.19	5.32	24.04	27.45
Coking coal	30	30.10	11.57	25.38	32.12
	70	24.77	10.00	40.90	24.33
	120	26.58	10.15	33.13	30.14

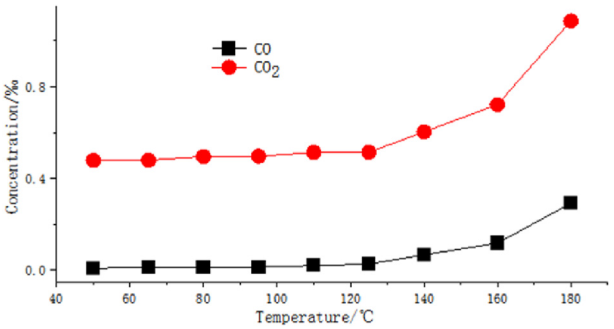

Fig. 13. Fat coal CO, CO_2 concentration changes curve with the temperature.

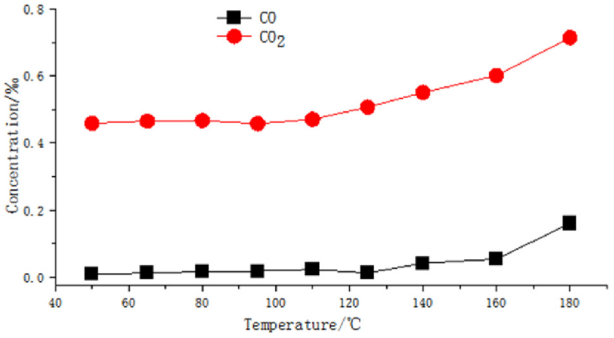

Fig. 14. Coking coal CO, CO_2 concentration changes curve with the temperature.

and CO_2 will be detected in the coal sample, which indicates that the coal sample has begun to oxidize.

With the increase in coal temperature, the CO production rate increases, and the CO output shows a strong regularity. During the whole oxidation process, the curve of CO concentration with temperature basically accords with the exponential distribution. When the coal temperature is between 50 and 150 °C, the concentration of CO and CO_2 has little change. The spontaneous combustion of coal is in the incubation period (Gradually reaching the ignition condition, also known as the slow oxidation stage). The concentration of CO began to increase at temperatures over 150 °C. Until the temperature reaches 180 °C, the concentration of CO and CO_2 increases sharply, which indicates that the coal oxidation reaction has entered the accelerated oxidation stage. If no measures are taken at this time, it will soon enter the intense oxidation stage, leading to the occurrence of coal spontaneous combustion.

When the coal temperature is between 50 and 120 °C, the concentration of CO and CO_2 does not change much, and the spontaneous combustion of coal is in the incubation period. When the temperature is 120 °C, compare three kinds of coal samples (The abbreviation for dimension is D) $D_{\text{gas coal}} < D_{\text{coking coal}} < D_{\text{fertilizer coal}}$. The fractal dimension of gas coal is close to 2, indicating that the pore surface of gas coal is smooth and the storage property of the reservoir is suitable. When the temperature is over 160 °C, the concentration of CO and CO_2 of gas coal changes significantly, and the oxidation speed is fast. However, the fractal dimension of fat coal and coking coal is similar, so the changing trend of oxidation rate is similar. The smaller the fractal dimension is, the easier it is to store and collect the gas. As the gas concentration increases, it is more likely to cause fire and explosion.

4. Conclusions

(1) The different pore distribution of the coal sample is obtained through the mercury injection experiment. The results show that the microscopic pores of coal samples with a pore diameter less than 100 nm and fractal dimension within 2–3 have good fractal characteristics, which can meet the fractal theory to describe the microscopic pore distribution characteristics of porous media.

(2) The simulation experiment of spontaneous combustion of coal found that the pore distribution structure of coal is closely related to its spontaneous combustion tendency. The results show that under the coal sample with a high degree of deterioration, the internal structure is more dense, and the storage capacity is better for CO, leading to the increased coal oxidation rate and causing spontaneous combustion.

(3) The study on the change of coal spontaneous combustion tendency of coal microscopic pore structure is helpful for us to take more effective measures for different coal combustion. The further study of coal rock structure and spontaneous combustion of coal is of far-reaching significance. The source of target gas and the change of coal structure can provide theoretical support for coal mining technology, which has far-reaching significance. The mystery of coal spontaneous combustion is revealed objectively and comprehensively, and the problem of mine fire caused by spontaneous combustion of coal is solved.

Conflicts of interest

The authors declare no conflict of interest.

Ethical statement

The authors state that the research was conducted according to ethical standards.

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