

Research Article

Study of the Effect of Drying on the Pore Structure and Rate of Oxygen Consumption and Heat Release of Immersed Lignite

Wei Guo ¹, Chunhua Zhang ¹, Ziwen Dong ², and Yongliang Han³

¹College of Safety Science and Engineering, Liaoning Technical University, Fuxin, Liaoning 123000, China

²School of Safety Engineering, Ningbo University of Technology, Ningbo, Zhejiang 315211, China

³China Coal Technology & Engineering Group Xi'an Research Institute, Xi'an, Shanxi 710077, China

Correspondence should be addressed to Ziwen Dong; 1316859454@qq.com

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To reveal the influence mechanism of the drying process of immersed coal on the spontaneous combustion characteristics, we measured the pore structure of coal with low-field nuclear magnetic resonance and conducted the temperature-programmed spontaneous combustion experiment. The results showed that the pore volume content of the pores with pore sizes of less than 100 nm was significantly reduced. The diameter range of 100–1000 nm was gradually generated after 8 h of coal drying, and the proportion of large pores and cracks larger than 1000 nm increased significantly during the drying process of the immersed coal. The oxygen consumption rate and the heat release rate were significantly affected by the degree of dryness and the temperature of coal samples in the temperature-programmed process. The turning point temperature was 140°C; below the turning point temperature, the raw coal had the fastest oxygen consumption and maximum heat release. When the coal temperature was higher than 140°C, the oxygen consumption rate and the heat release rate of the coal sample that was dried for 24 h were higher than those for the raw coal, followed by the samples that were dried for 8 h and dried for 48 h. The main reason is that the water content of the coal sample decreases to 11.4% and the porosity increases to 16.5% after drying for 24 h, while the proportion of pores or cracks larger than 1000 nm increases significantly to 87.17%, the largest increase range and the pore connectivity will reach the maximum.

1. Introduction

China is rich in lignite resources, with a total resource of about 319.4 billion tons and proved reserves of 129.1 billion tons, accounting for 13% of proved coal reserves. Lignite in eastern Inner Mongolia accounts for 77.55% of the total lignite resources in China and 45.0% of the reserved coal reserves in Inner Mongolia. The mining, processing, utilization, storage, and transportation of lignite will be strengthened, and the problem of lignite spontaneous combustion will be highlighted [1, 2]. The physical and chemical structure of coal is easily changed by water immersion and air drying, and through these changes, the characteristics and process of spontaneous combustion of coal are affected. Compared with raw coal, when coal is immersed in water, the pores absorb water and expand, and the average pore size increases [3]. After the coal is soaked in water for a

certain period, obvious wet swelling and softening occur, forming a certain type of crushing, and some of the broken small particles fall off from the soaking coal [4–9]. The mineral dissolution results in a decrease in mineral content and an increase in macropore ratio and specific surface area [10]. After soaking in water, the volume of the original coal sample expands, the cracks and pores become more developed in the drying process, and the oxygen absorption is greater than that of the dry coal sample [11]. After soaking and drying, the pore volume and the specific surface area of the mesopore and micropore decrease, while the pore volume and the specific surface area of the mesopore and macropore increase, and the average pore size increases greatly, leading to the development of pore channels in coal, enhanced connectivity, enhanced oxygen adsorption and circulation capacity, enhanced oxidation capacity, and an increased risk of spontaneous combustion [12–14].

Water immersion can transfer the active sites, but proximation can activate the primary active sites [15]. Because of the generation and existence of humid heat, coal may easily undergo oxidation and spontaneous combustion at low temperatures [16]. Pan et al. [17] pointed out that the presence of water during the PR pyrolysis process showed the competitive characteristics of promoting and inhibiting the spontaneous combustion of the coal at the same time. In addition, the soaking process reduced the crossing point temperature of the soaked coal and increased the risk of spontaneous combustion for the soaked coal [18–20]. After coal is immersed in water, the type, generation rate, and temperature of gas generated during spontaneous combustion change [21–24]. After soaking and air drying, the aliphatic hydrocarbon chain is broken and the chain reaction process is accelerated, which can change the spontaneous combustion oxidation characteristics of coal and promote the oxidation and spontaneous combustion of coal [22, 25–27]. These studies have shown that after the coal is soaked in water and air-dried, the hydrocarbon aliphatic group increases, the oxygen-containing functional group increases, and the aromatic hydrocarbon group decreases, which accelerates the oxidation and spontaneous combustion of coal.

Different immersion and air-drying degrees, however, show different inhibition or promotion effects on spontaneous combustion, and the current reaction kinetics cannot fully and accurately explain this phenomenon [28–31]. Therefore, we proposed research to examine the influence of the drying degree of immersed coal on the spontaneous combustion characteristics of coal. Low-field nuclear magnetic resonance (NMR) technology was used to measure the change in the pore structure of saturated coal at different drying degrees. A temperature-programmed experiment was used to carry out the study of the macroscopic spontaneous combustion laws. Based on the comparison and analysis of the distribution and variation of the pore structure, oxygen consumption rate, and heat release rate of immersed coal in different air-drying conditions, we revealed the variation and mechanism of the spontaneous combustion characteristics of coal.

2. Materials and Methods

2.1. NMR Test Method for Coal Pore Structure. The samples of lignite used in this experiment were obtained from the Fengshuigou Coal Mine of the Pingzhuang Coal Company in Inner Mongolia, China. The moisture content of the raw coal was 12.35%, the ash content was 22.5%, the volatile content of the coal was 42.5%, and the fixed carbon content was 24.8%. The contents of C, H, N, S, and O in coal samples are 51.25%, 4.12%, 0.85%, 0.21%, and 18.87%, respectively. The hydrogen-carbon ratio $H/C = 0.9647$, and the oxygen-carbon ratio $O/C = 0.2761$.

The coal samples were cut into cuboids with sizes of 12–13 mm, placed in the vacuum saturation device of an NMI20-040 V-I system produced by Niumag Company, and then vacuumized after the sealing. After vacuum stabilization, 30 MPa high-pressure water saturation was carried out. After 12 h of high-pressure water immersion, the

samples were taken out, wrapped with raw tape, and placed in the NMI20-040 V-I system for NMR testing. The results revealed the pore structure of the raw coal. The treatment methods for coal samples with different degrees of drying after soaking in water were as follows: the cuboid coal samples were immersed in water for 14 d and then placed in a drying oven with constant temperature and humidity. The drying temperature was set at 30°C, the relative humidity was set at 60%, and the drying times were set at 8 h, 24 h, and 48 h, respectively. The coal sample used in this paper is lignite from Chifeng City, Inner Mongolia Autonomous Region. The experimental time is from June to July 2022, which is also the rainy season in summer in Chifeng City, Inner Mongolia. Both coal mines and coal storage and transportation sites will have the process of coal piles being soaked by water and then drying naturally, and there is a great risk of spontaneous combustion in this process. Therefore, during the experiment, this paper determined to carry out the research based on the common temperature of 30°C and humidity of 60% in Chifeng City, Inner Mongolia, in this season. After the corresponding drying time was reached, the same pore structure test as that of the raw coal was carried out, and the test result was the pore structure of the coal samples after soaking in water and drying at different times. The NMR test procedure for the coal pore structure is shown in Figure 1.

2.2. Temperature-Programmed Experiment. We established the temperature-programmed experimental system, as shown in Figure 2. The coal sample scanning parameters are given in Table 1, and the particle size composition of the samples is given in Table 2. The heating rate of the temperature-programmed furnace during the experiment was 1°C/min, and the airflow was 500 mL/min. Gas samples were collected and analyzed with gas chromatography as the temperature varied in an air atmosphere from 30°C to the maximum temperature of the coal samples at intervals of 10°C to determine the compositions and concentrations of the gas samples. The experiment was stopped when the temperatures of the coal samples in the air environment reached 170°C.

3. Experimental Results

3.1. Pore Structure Change Rule. Based on the low-field MRI results, Figure 3(a) shows that in the range of peaks on the left, the T_2 value corresponding to the peak value of the dry coal sample was basically stable. In the range of the right peak, compared with the raw coal, the width of the peak of the coal sample that was dried for 8 h increased significantly, showing an increase to the left and right sides, and the intensity of the peak also increased significantly. This result indicated that during the drying process of the saturated coal, the content of micropores and mesopores in the coal decreased somewhat, but the decrease in amplitude was not significant. The number of larger pores and cracks corresponding to the second peak value and the size of the pore diameter increased significantly—that is, the expansion and generation of larger pores and cracks in the drying process was the main cause of the change in the pore structure.



FIGURE 1: NMR test procedure for coal pore structure.

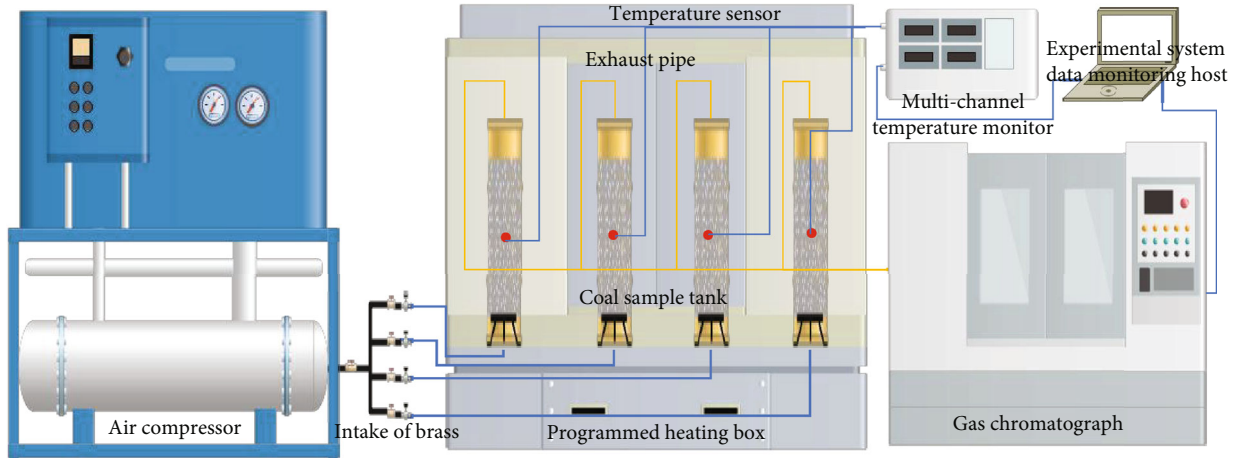


FIGURE 2: Temperature-programmed experimental system.

TABLE 1: Coal sample scanning parameters.

Coal sample processing method and number	m (g)	h (cm)	M_c (%)	V (cm ³)	n (1)
Raw coal (Rc)	1333	19.8	12.1	1172	0.41
ad-8 h	1328	22	14.0	1727	0.47
ad-24 h	1349	21	11.4	1649	0.42
ad-48 h	1247	19	8.4	1492	0.39

TABLE 2: Particle size composition of the sample.

Number	Rc	ad-8 h	ad-24h	ad-48h
Particle size range	%			
20–10 mm	12.3	14.1	6.9	7.1
10–5 mm	31.5	25.6	29.4	27.9
5–2.5 mm	17.2	21.4	22.0	20.8
2.5–1.0 mm	19.1	21.8	23.8	23.7
1.0–0.5 mm	8.8	8.3	8.8	10.1
0.5–0.25 mm	5.8	5.0	5.3	6.0
0.25–0.18 mm	1.4	1.0	1.0	1.2
0.18–0.15 mm	0.8	0.6	0.7	0.8
0.152–0.1 mm	1	0.8	0.8	1.0
<0.1 mm	2	1.4	1.4	1.5

Figure 3(b) shows that new pores with sizes of 6550–10680 nm were generated in coal samples 1-2, and 63900–28350 pores or cracks were generated in coal samples 1-3 after 8 h of drying. Similarly, the total integral area of the pores, namely the pore volume, did not increase significantly

in the range below 1000 nm, whereas the total amount of larger pores increased significantly, and large numbers of new pores and fissures appeared.

In Figure 4(a), the change of the peak on the left is basically similar to that of the other drying degrees. The

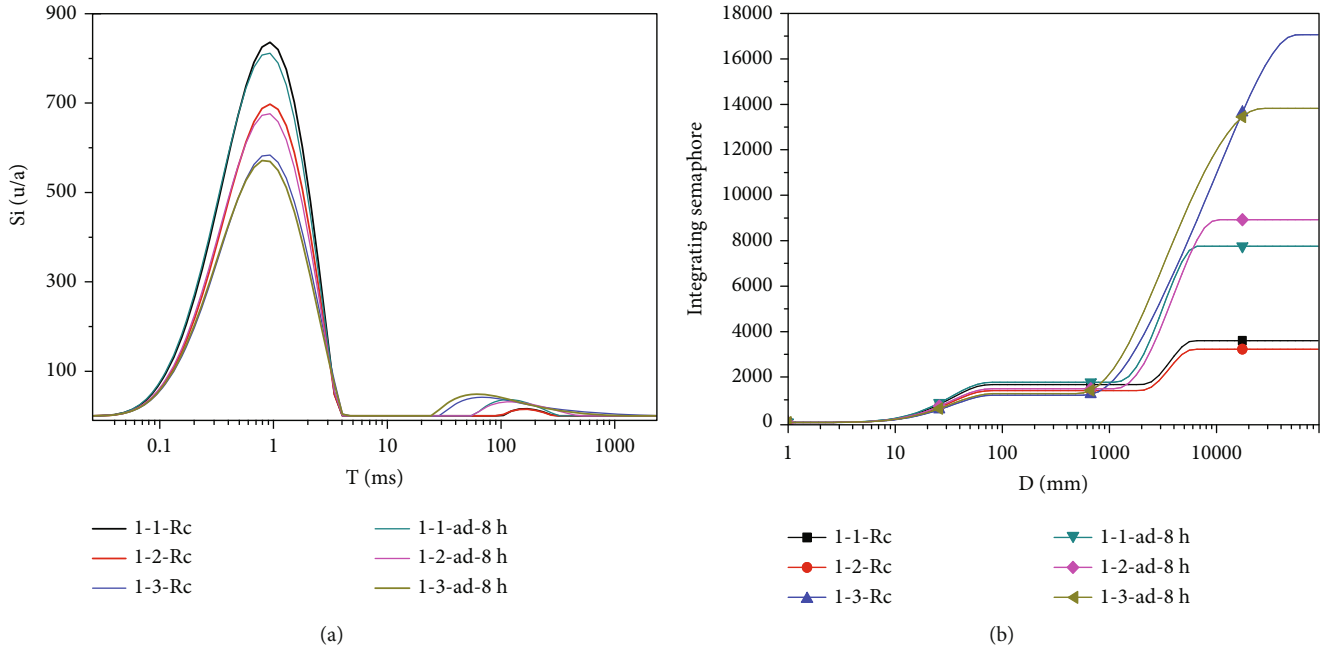


FIGURE 3: T_2 distribution and accumulated integral area of pores with different pore sizes for saturated coal after 8 h of drying. Note: sample number 1-1-Rc indicates the test result of the first sample in the first experimental group under raw coal conditions. 1-1-ad-8 h is the test result of the first sample in the first experimental group after drying for 8 h in the air environment after saturation. This figure, Figure 4, and Tables 1 and 2 are similar. (a) T_2 distribution. (b) Accumulated integral area of pores with different pore sizes.

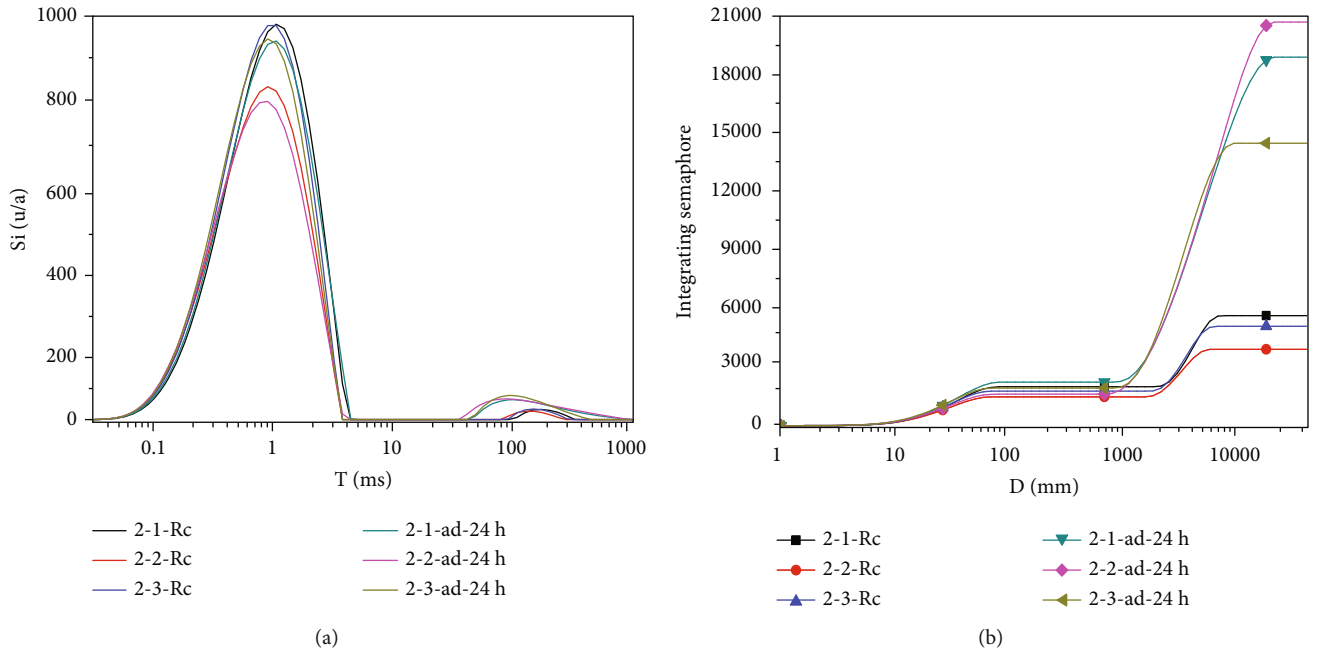


FIGURE 4: T_2 distribution and accumulated integral area of pores with different pore sizes of saturated coal after 24 h of drying. (a) T_2 distribution. (b) Accumulated integral area of pores with different pore sizes.

width and the peak value of the peak increased, but the increase was not significant, indicating that the pore size, content, and total amount of relatively small pores did not change significantly during the drying process. The distribution of the peak on the right side showed that

after 24h of drying, the peak extended significantly to the left and right sides, the peak width increased significantly, and the peak signal increased significantly, indicating that large pores and cracks occurred in the coal after 24h of drying.

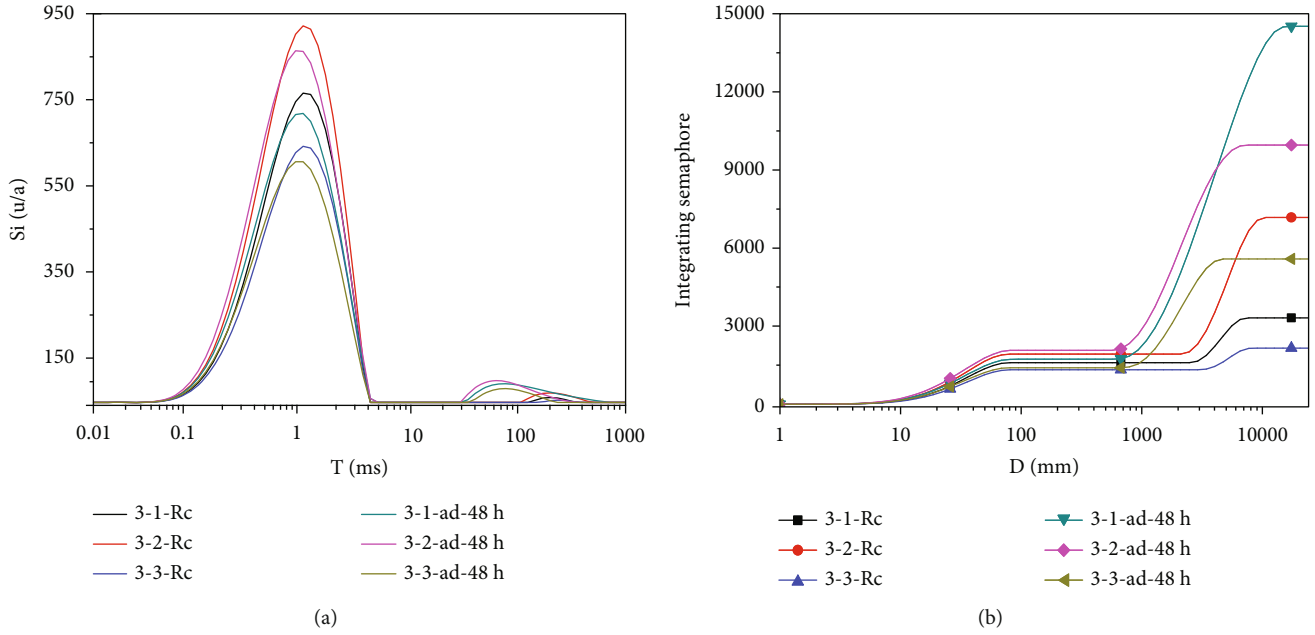


FIGURE 5: T_2 distribution and accumulated integral area of pores with different pore sizes for saturated coal after 48 h of drying. (a) T_2 distribution. (b) Accumulated integral area of pores with different pore sizes.

TABLE 3: Porosity changes of raw coal and saturated dry coal.

Raw coal	Porosity (%)	Saturated dry coal	Porosity (%)	Percentage increase in porosity (%)	Average porosity increases (%)
1-1-Rc	20.1	1-1-ad-8 h	22.0	9.7	
1-2-Rc	20.6	1-2-ad-8 h	22.7	10.3	9.4
1-3-Rc	21.0	1-3-ad-8 h	22.7	8.2	
2-1-Rc	19.1	2-1-ad-24 h	22.3	16.4	
2-2-Rc	18.1	2-2-ad-24 h	21.3	17.7	16.5
2-3-Rc	20.0	2-3-ad-24 h	23.1	15.4	
3-1-Rc	20.2	3-1-ad-48 h	24.8	22.4	
3-2-Rc	22.5	3-2-ad-48 h	26.8	19.1	20.1
3-3-Rc	21.4	3-3-ad-48 h	25.4	18.9	

Figure 4(b) shows the comparison of the pore structures of the coal sample and raw coal after drying for 24 h. Except for the pores below 1000 nm, which had changes like the other drying time changes, there were no significant changes. For coal sample 2-1, the pore sizes in the range of 7700–20470 nm were the newly generated large pores or cracks after drying for 24 h. Correspondingly, the size range for coal sample 2-2 was 4730–20470 nm, and that for coal sample 2-3 was 6550–9070 nm.

Figure 5(a) shows the pore scanning results for the coal before and after drying for 48 h. In the range of the left peak, the variation trend was basically consistent with those for drying for 8 h and 24 h. In the range of the left peak, the width and the peak value of the peak increased slightly, but the increase was not significant. Within the range of the right peak, however, taking coal sample 3-3 as an example, the left boundary of the right peak extended significantly to the left and to the right. The increase in the right peak

indicated that more and larger pores and cracks were generated or expanded. The significant increase in the left peak indicated that the pores within the range of the original right peak also increased.

The comparison between the coal samples that were dried for 48 h and raw coal is shown in Figure 5(b). For coal samples 3-1, larger pores or cracks with pore sizes of 7700–17400 nm were generated due to 48 h of drying. For the other coal samples, although there was no significant increase in the larger pore size, the total amount of pores corresponding to the original pore size increased significantly, which also indicated that drying resulted in a significant increase in the large pores in the coal.

Table 3 shows the porosity change and porosity increase percentage of the coal during the drying process. Compared with raw coal, the porosity of the coal increased after drying, and the longer the drying time was, the greater the porosity increase proportion of the coal was.

TABLE 4: Pore volume ratios for different pore sizes of raw coal and saturated coal after drying.

Raw coal	<10 nm	10–100 nm	100–1000 nm	>1000 nm	Saturated dry coal	<10 nm	10–100 nm	100–1000 nm	>1000 nm
	Pore volume ratio (%)					Pore volume ratio (%)			
1-1-Rc	4.4	41.8	0.0	53.8	1-1-ad-8 h	2.3	20.5	0.0	77.2
1-2-Rc	4.1	39.4	0.0	56.5	1-2-ad-8 h	1.7	15.0	0.0	83.4
1-3-Rc	0.7	6.4	3.9	89.0	1-3-ad-8 h	0.9	8.3	7.9	82.9
2-1-Rc	3.1	32.3	0.0	64.6	2-1-ad-24 h	1.1	10.8	1.1	87.1
2-2-Rc	4.6	33.3	0.0	62.1	2-2-ad-24 h	1.0	6.8	2.6	89.5
2-3-Rc	3.8	31.2	0.0	65.0	2-3-ad-24 h	1.6	11.7	1.8	84.9
3-1-Rc	3.9	44.1	0.0	52.0	3-1-ad-48 h	1.1	10.8	4.9	83.2
3-2-Rc	2.3	24.6	0.0	73.2	3-2-ad-48 h	2.1	18.7	10.8	68.4
3-3-Rc	5.1	56.3	0.0	38.6	3-3-ad-48 h	2.5	22.6	6.8	68.0

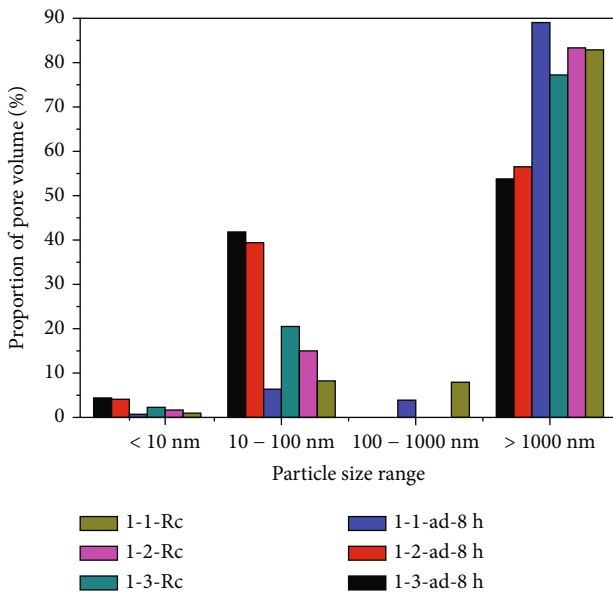


FIGURE 6: Pore volume ratios for different pore sizes of the saturated coal after drying for 8 h.

Table 4 shows the percentage of the pore volume in different pore size ranges during the drying process. As illustrated in Figures 6–8, the proportion of micropores in the coal samples used in this experiment was very low, and the total amount of micropores in the 18 tested coal samples decreased to 45% of the original coal on average, except that the proportion increased only slightly after the drying of samples 1-3 and 3-2. Two samples with low relative errors in the same group were averaged, and the proportion of micropores in the coal samples decreased to 53%, 61%, and 61% of that in raw coal after drying for 8 h, 24 h, and 48 h, respectively. Except for samples 1-3 and 3-2, the average results of the other 16 groups of coal samples showed that the content of mesopores was reduced to 49% after drying. However, large pores (100–1000 nm) and large pores or cracks (>1000 nm) were not detected in the raw coal, except for sample 1-3. After drying for 8 h, the existence of large pores or cracks could be detected in the individual coal sam-

ples of each group. When the drying time reached 24 h, the occurrence of macropores could be detected in all the dried coal samples, and the proportion of macropores increased gradually with the drying time. The proportions of macropores in the coal after drying for 8 h, 24 h, and 48 h were 1.2%, 1.8%, and 7.5% on average. Although the proportion of macropores in the pore system of the coal increased with the drying time, the proportion of large pores was not large. At the same time, large pores, and cracks larger than 1000 nm were detected in all the coal samples after drying and the proportions of large pores and cracks in the coal samples after drying for 8 h, 24 h, and 48 h reached 83%, 87%, and 73% on average, respectively.

Combined with the significant drying shrinkage of the coal and the drying and cracking phenomenon of the shallow surface, we found that the volume of high water-bearing coal in the drying process had significant drying shrinkage, and many large pores and cracks larger than 1000 nm, accounting for 73–87% of the total, appeared in the shallow surface. The generation of deep cracks in the block coal was much lower than that on the shallow surface. Therefore, the deep layer of coal was accompanied by significant drying shrinkage and significant reductions in the micropore, pore, and mesopore content. In summary, the porosity and the large pores and cracks of coal increased significantly during the drying process, resulting in an increase in large channels for oxygen flow and storage in the coal as well as enhanced connectivity.

3.2. Changes in Oxygen Consumption Rate and Exothermic Intensity. According to the temperature, oxygen concentration, carbon monoxide, and carbon dioxide concentration of coal samples during the temperature-programmed experiment, the oxygen consumption rates for different coal samples in the low-temperature oxidation stage were calculated as shown in Figure 9.

When the temperature of the coal sample was lower than 70°C, the oxygen consumption rate of the raw coal was higher than that of the coal sample dried for 48 h, which in turn was higher than that of the other dry coal samples soaked in water. The oxygen consumption rates of the coal samples dried for 8 h and 24 h were lower than those for

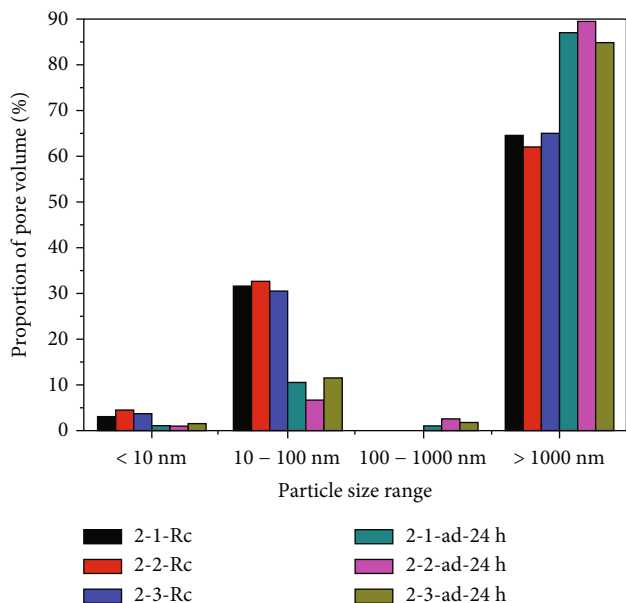


FIGURE 7: Pore volume ratios for different pore sizes of the saturated coal after drying for 24 h.

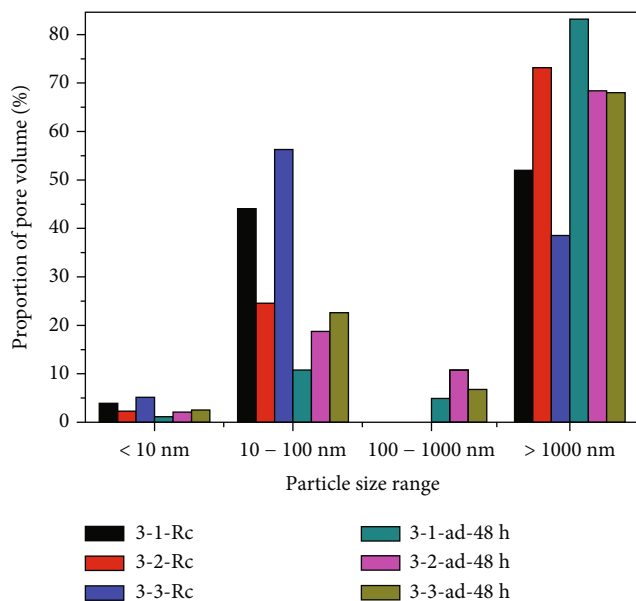


FIGURE 8: Pore volume ratios for different pore sizes of the saturated coal after drying for 48 h.

the raw coal and the coal samples dried for 48 h, but the difference between the rates was small.

When the temperature of the coal sample was in the range of 70–90°C, the oxygen consumption rate of the raw coal was the highest, followed by that of the coal samples dried for 48 h, then by that of the coal samples dried for 8 h, and the lowest oxygen consumption rate was that of the coal samples dried for 24 h after soaking in water. When the temperature of the coal sample was in the range of 90–100°C, the order of the oxygen consumption rates was raw

coal > coal sample dried for 8 h > coal sample dried for 48 h > coal sample dried for 24 h. When the coal temperature was within the range of 100–120°C, the order of the oxygen consumption rates was raw coal > coal sample dried for 8 h > coal sample dried for 24 h > coal sample dried for 48 h. When the coal temperature was in the range of 120–140°C, the order of the oxygen consumption rates was raw coal > coal sample dried for 24 h > coal sample dried for 8 h > coal sample dried for 48 h. When the temperature of the coal was in the range of 140–170°C, the order of the oxygen consumption rates was coal sample dried for 24 h > raw coal > coal sample dried for 8 h > coal sample dried for 48 h.

For the temperature-programmed condition, the spontaneous combustion risk for the coal samples could be directly determined using the oxygen consumption rate and the heat release intensity of the coal samples, and the coal samples with a high oxygen consumption rate and heat release intensity had a high spontaneous combustion risk. At different temperature stages, the sequence of spontaneous combustion risks for different coal samples was different, and there were significant differences in the spontaneous combustion risks due to different temperatures and different treatment methods for the coal samples. When the temperature of the coal sample was below 90°C, the spontaneous combustion risk for the coal sample dried for 48 h was close to that of the raw coal but higher than those of the coal samples dried for 8 h and 24 h. When the temperature of the coal sample reached 90–120°C, the raw coal had the highest spontaneous combustion risk, followed by the samples that were dried for 8 h, 24 h, and 48 h, which showed the highest spontaneous combustion risk of the raw coal. The longer the drying time after soaking in water was, the higher the spontaneous combustion risk of the coal sample was. When the temperature of the coal sample reached the range of 120–140°C, the spontaneous combustion risk of the raw coal was close to that of the coal sample dried for 24 h and higher than that of the coal sample dried for 8 h, and the lowest was that of the coal sample dried for 48 h. When the temperature of the coal sample was higher than 140°C, the spontaneous combustion risk of the coal sample dried for 24 h was the highest, followed by those for the raw coal, and then the coal sample dried for 8 h after soaking in water, and the lowest risk was that for the coal sample dried for 48 h.

Coal oxidizes with oxygen, releasing a large amount of heat. According to the experimental results, as shown in Figure 10, different treatment processes had a significant impact on the upper limit of the heat intensity released by coal oxidation. The change rule of the upper limit of the heat intensity with temperature was basically consistent with the change rule for the oxygen consumption rate with temperature. When the coal temperature was lower than 70°C, the upper limit of the heat release intensity of the coal sample was lower, but the upper limit of the heat release intensity of the raw coal sample was the highest in general, followed by the coal sample that was soaked and dried for 48 h. When the temperature of the coal sample was higher than 90°C, the upper limit of the heat release intensity of the coal sample began to increase significantly. In the range of 100–120°C, the upper limit of the heat release intensity of the raw coal

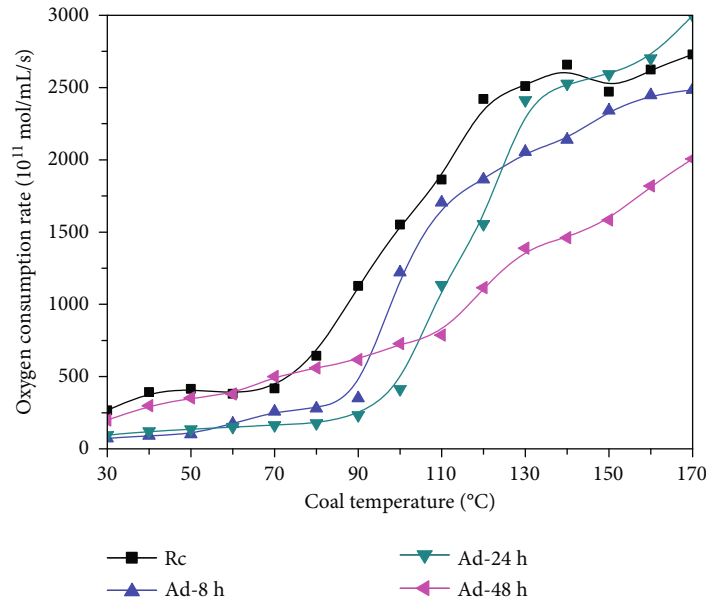


FIGURE 9: Oxygen consumption rate.

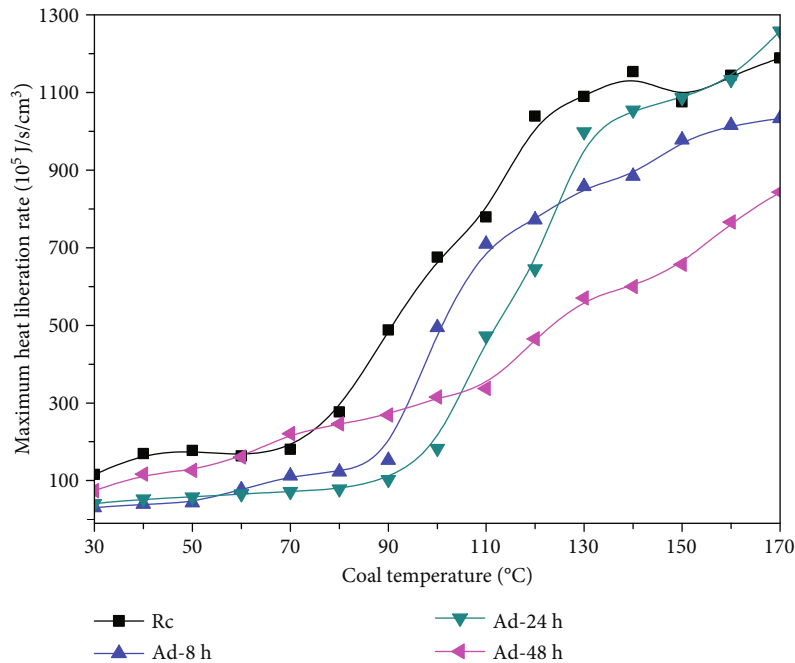


FIGURE 10: Upper limits of oxidation exothermic intensity.

was the highest, followed by that for the sample dried for 8 h after soaking in water, then by that for the sample dried for 24 h, and the lowest was that for the coal sample dried for 48 h. When the temperature of the coal sample was above 160°C, the upper limit of the heat release intensity of the coal sample dried for 24 h after soaking in water was the highest, higher than that of the raw coal. The coal sample dried for 8 h was next to the raw coal, and the coal sample dried for 48 h after soaking in water had the lowest upper limit.

The upper limit of the exothermic strength and the change of the oxygen consumption rate with the tempera-

ture fully indicated that the oxidation capacity and the exothermic strength of the coal decreased and increased in a certain temperature range after a certain length of time of air drying at room temperature. Generally, when the temperature of the coal sample reached approximately 120°C, the oxidation capacity and the heat release intensity of the coal sample after soaking and drying for 24 h gradually became close to those of the raw coal. When the temperature of the coal sample was above 140°C, the oxidation capacity and the heat release intensity of the coal sample after soaking and drying for 24 h gradually became higher than those of

the raw coal, and the difference between the coal sample and the raw coal after soaking and drying for 8 h began to narrow. This indicated that the spontaneous combustion risk of the coal samples dried for 24 h after soaking in water gradually increased when the coal temperature was higher than 120°C. When the temperature of the coal samples was higher than 140°C, the spontaneous combustion risk was significantly higher than that of raw coal, and the difference between the spontaneous combustion risk for the coal samples dried for 8 h and the raw coal gradually decreased. The effects of the drying after soaking on the spontaneous combustion risk of the coal were complex, and there were both promoting and inhibiting effects. The two effects were significantly affected by the degree of drying and the temperatures of the coal samples. The oxidation capacity, heat release intensity, and spontaneous combustion risk of the coal samples dried for 24 h at the high-temperature stage were significantly higher than those of the raw coal and the other coal samples with different drying treatments or without drying treatment after soaking.

4. Discussion

During the drying process of the coal (12–13 mm) after soaking in water, the porosity increased continuously, and the growth rate of the porosity increased continuously within the drying time range of 48 h. During the drying process, the pore volume of the micropores and small pores in the coal pore system was reduced to about half of that of the raw coal. In the pore system of the raw coal used in the experiment, there were very few large pores in the pore size range of 100–1000 nm. Then the proportion of large pores began to increase significantly after drying for 24 h, and the proportion increased significantly in the process of the extended drying time. More than half of the total pore volume in the raw coal was composed of larger pores or fractures with pore sizes larger than 1000 nm. With the development of the drying process, the pore size larger than 1000 nm accounted for a significant increase in the total pore volume, reaching an average of 73–87%. The decrease of the micropores and the pore volume with a pore diameter of less than 100 nm, the production of large pores at 100–1000 nm, the significant increase of the proportion of pores with pore sizes greater than 1000 nm in the total pore volume, and the significant increase of the total pore volume and porosity jointly determined the enhancement of the pore connectivity, porosity, and large cracks in the coal during the drying process. As a result, the air seepage resistance decreased and the contact surface between the air and coal increased significantly. In particular, the proportion of larger pores and cracks larger than 1000 nm in the coal samples dried for 24 h was the highest.

Considered from the perspective of the pore structure alone, the oxidation capacity and the spontaneous combustion risk of the coal samples dried for a certain period after soaking in water had to increase. The change in pore structure provided the structural basis for the change in the spontaneous combustion risk characteristics of the coal. The pore structure change rule and experiments for the different

temperature-programmed oxidation processing of the spontaneous combustion of the coal sample rule had strong consistency in that the pore structure had a significant effect on the spontaneous combustion of the low-temperature oxidation of the coal. The larger pores and fissures produced in the drying process and the increase in porosity could improve the risk of coal oxidation and spontaneous combustion to a certain extent.

The temperature-programmed spontaneous combustion experiment results for the coal samples with different particle sizes (less than 20 mm) showed that the oxygen consumption rate and heat release rate were significantly affected by the degree of dryness and the temperature of the coal samples in the temperature-programmed process. When the temperature of the coal was lower than 140°C, the oxygen consumption rate and the heat release rate of the dried coal were lower than those of the raw coal, and this stage of the drying after immersion showed oxidation inhibition. When the coal temperature was higher than 140°C, the oxygen consumption rate and the heat release rate of the coal sample that was dried for 24 h were higher than those of the raw coal. At that stage, drying for 24 h showed oxidative promotion, but the other dryness degree of the coal was still lower than that of the raw coal. For the three kinds of coal samples with different drying times, when the coal temperature was below 90°C, the highest oxygen consumption rate and the heat release rate were those for the coal sample that was dried for 48 h, which had little difference from the raw coal. However, there was no significant difference between the rates for the coal samples that were dried for 8 h and 24 h, which were lower than those for the coal samples that were dried for 48 h. In the range of 90–120°C, the maximum oxygen consumption rate and the heat release intensity were those for the coal samples dried for 8 h, followed by those for the samples dried for 24 h and 48 h. When the coal temperature was higher than 120°C, the maximum oxygen consumption rate and the heat release intensity were those for the coal samples dried for 24 h, followed by those for the samples dried for 8 h and 48 h.

The change in regularity of the pore structure for different degrees of drying after soaking in water showed that the pore structure composition of the dried coal was more easily oxidized after drying, the risk of spontaneous combustion increased, and the risk of spontaneous combustion for the dried coal might have been higher than that of raw coal. The risk of spontaneous combustion for the coal samples dried for 24 h was the highest. The macroscopic experimental results of the spontaneous combustion of the coal based on temperature-programmed heating, however, had some differences. The macroscopic temperature-programmed spontaneous combustion experiment showed that when the coal temperature was higher than 140°C, the microscopic experimental results were consistent with the macroscopic experimental results. When the coal temperature was lower than 140°C, the microscopic experimental conclusion could explain the sequence of the spontaneous combustion risk for three coal samples with different degrees of drying, but it was not enough to fully explain the relationship between raw coal and coal with different degrees of drying. This

indicated that the macroscopic spontaneous combustion of coal in the low-temperature oxidation stage was affected by the pore structure, functional group, and activation energy as well as by the macroscopic particle size, stacking state, macroscopic ventilation, and oxygen supply conditions of the coal. The spontaneous combustion of the coal was the result of multiscale and multifactor micro- and macrocomprehensive actions as well as both internal and external factors. Although the microscopic experimental results could reveal the mechanism of the spontaneous combustion of the coal to a certain extent, it was difficult to predict the process of the spontaneous combustion of the coal accurately. It was not suitable to judge the characteristics and the development process of the spontaneous combustion of the coal with the microscopic experimental results alone. To reveal the mechanism of spontaneous combustion and predict the process of spontaneous combustion, it was necessary to combine microscopic and macroscopic experiments.

5. Conclusion

Based on this research, the following conclusions can be drawn:

- (1) During the drying process of saturated coal, pores with pore sizes of less than 100 nm were significantly reduced to about half of that of raw coal. Pores with pore diameters in the range of 100–1000 nm were gradually generated after 8 h of coal drying, and the proportion increased gradually with the extension of the drying time. The proportion of large pores and cracks larger than 1000 nm increased significantly. The change in the coal pore structure caused by drying resulted in the gradual connection of the pores in the dried coal and the increase in porosity. Large pores or fissures arose and increased in large numbers. In particular, the proportion of larger pores and cracks larger than 1000 nm in the coal samples dried for 24 h was the highest
- (2) The oxygen consumption rate and the heat release rate were significantly affected by the degrees of dryness and the temperatures of the coal samples in the temperature-programmed process. When the temperature of the coal was lower than 140°C, the oxygen consumption rate and the heat release rate of the dried coal were lower than those of raw coal. When the coal temperature was higher than 140°C, the oxygen consumption rate and heat release rate of the coal sample that was dried for 24 h were higher than that of the raw coal. At that stage, drying for 24 h showed oxidative promotion, but the other dryness degree of the coal was still lower than that of the raw coal
- (3) The macroscopic spontaneous combustion of coal in the low-temperature oxidation stage was the result of multiscale and multifactor micro- and macrocomprehensive action as well as internal and external factors. Although the microscopic experimental

results could reveal the mechanism of the spontaneous combustion of coal to a certain extent, it was difficult to predict the process of the spontaneous combustion of coal accurately. Additionally, it was not suitable to judge the characteristics and the development process of the spontaneous combustion of the coal with the microscopic experimental results alone. To reveal the mechanism of spontaneous combustion and predict the process of spontaneous combustion, it was necessary to combine the microscopic and macroscopic experiments

Data Availability

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

Disclosure

This work “Study of the effect of drying on the pore structure and characteristics of spontaneous combustion of immersed Lignite” has been posted as a preprint on the Research Square by Springer Nature (<https://www.researchsquare.com/article/rs-1894214/v1> and MSID: ef65c892-30 dc-4 dc3-9887-4e68d2feded6).

Conflicts of Interest

The authors declare that they have no conflicts of interest or personal relationships that could have appeared to influence the work reported in this paper.

Authors' Contributions

Wei Guo performed the methodology, data curation, writing which is the original draft preparation, and writing which is the review and editing. Chunhua Zhang did the conceptualization, validation, funding acquisition, and writing which is the review and editing. Ziwen Dong worked on the software, funding acquisition, visualization, and writing which is the review and editing. Yongliang Han conducted the resources (provision of study materials and laboratory samples).

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