

Experimental Study on Spontaneous Combustion Characteristics of Large Coal Particles after Soaking

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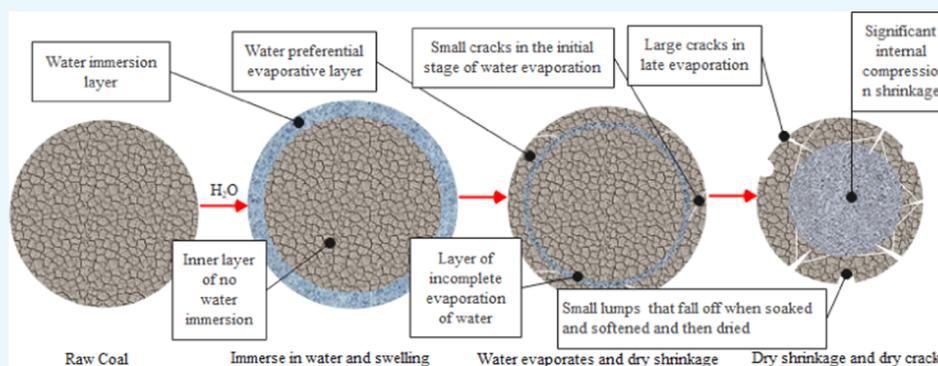
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ABSTRACT: The spontaneous combustion of coal is affected by many factors, among which the influence of water is significant and complicated. To explore the influence of water on the spontaneous combustion characteristics of goaf residual coal, coal samples with similar particle size distributions to those of goaf residual coal were prepared. After the coal samples were immersed in water for 7–21 days and the external flowing water was drained, spontaneous combustion experiments were carried out using a temperature-programmed method. The results showed that soaking in water could promote and inhibit the spontaneous oxidative combustion of large coal particles in different temperature ranges. When the coal temperature was below 50 °C, water immersion had a significant inhibition effect on coal oxidation and spontaneous combustion. When the temperature of coal was 50–110 °C, soaking in water for 7 days could promote the oxidation and spontaneous combustion of coal. However, soaking for 14 and 21 days had a significant inhibition effect in this temperature range. When the coal temperature was higher than 110 °C, water immersion had a significant inhibition effect on the coal. Moreover, a prolonged immersion time significantly enhanced the inhibition effect. When the immersion time was less than 21 days, the spontaneous combustion of large coal particles by short-term soaking was mainly inhibited.

1. INTRODUCTION

Coal spontaneous combustion (CSC) is a well-known phenomenon worldwide that can cause coal mine fires.¹ Coal mine fires are one of the most serious disasters faced by the coal industry all over the world, where coal is mined using the underground method. Take China, for example, more than 90% of the coal mine fires are caused by CSC.^{2,3} This significantly affects and restricts the safe production of coal mines, threatens the lives and health of miners and causes greenhouse gas emissions and environmental pollution.^{4–7} In addition, CSC also burns large amounts of coal resources and indirectly causes the destruction of surface vegetation, soil desertification, surface collapse, and disorderly discharge of dust particles. Thus, they have become a serious international environmental disaster.^{8–13} Water mainly affects coal spontaneous combustion by altering the physical and chemical structures of coal and changing the characteristics and processes of coal spontaneous combustion. Song, et al.^{14,15}

used coal particles with diameters less than 3 mm to conduct pore structure experiments. The results showed that compared with raw coal, the average pore size of the soaked coal increased, the total pore volume decreased, and the specific surface area decreased. The research reported by Dong et al.,¹⁶ Wen et al.,¹⁷ and Fry et al.¹⁸ showed that when coal was immersed in water for a certain period, the coal underwent significant wet swelling and softening, and part of the broken coal fell off from the coal. Zheng et al.¹⁹ used coal particles with diameters less than 0.425 mm to study the effects of the

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pore and chemical structures on the spontaneous combustion of coal. Qin et al.²⁰ studied the effect of the soaking process on the spontaneous combustion characteristics of long-flame coal with coal particle sizes less than 3 mm. The results showed that after soaking for a long time, the coal had a more developed surface pore structure, and the average pore size, mesoporous volume, and micropore volume increased. Li et al.²¹ studied the effect of the pore structure on the spontaneous combustion characteristics of submerged coal with particle sizes less than 0.109 mm. After soaking in water, the volume of the raw coal sample expanded, cracks and pores became more developed, and oxygen absorption was greater after water was removed by drying. The above results indicate that after coal is immersed in water for a long time, the pore volumes and specific surface areas of the mesopores and micropores decrease, while those of mesopores and macropores increase, additionally, average pore diameters increase substantially. This resulted in the development of pore channels in coal, enhanced connectivity, enhanced oxygen adsorption and circulation capacities, enhanced oxidation, and increased risk of spontaneous combustion.

Coal is a porous medium, and it has moisture due to van der Waals forces and hydrogen bonding.²² At low temperatures, only a few functional groups in coal can react with oxygen and chemisorb.^{23,24} When coal is immersed in water, the active structure or functional groups will be changed, which will change the characteristics and processes of coal spontaneous combustion. Qiao et al.²⁵ selected coal samples with particle sizes of 100–250 meshes to carry out infrared spectroscopy and thermogravimetry experiments. The results showed that the number of active groups and oxidation activity along with the amount of heat release of the soaked coal was higher than those of raw coal. Zheng et al.¹⁹ pointed out that as the number of active functional groups and the oxidation heat release of the water-immersed and air-dried coal increase, the activation energy decreases, and the spontaneous combustion tendency is enhanced. Qin et al.,²⁶ Deng et al.,²⁷ Yuan et al.,²⁸ Lu et al.,²⁹ Song et al.,³⁰ and Kan¹⁵ concluded through experiments and literature analysis that after the coal was immersed in water, the hydrocarbon–aliphatic group content increased along with that of oxygen-containing functional groups, while the content of aromatic hydrocarbon groups decreased. All of these changes accelerated the process of coal oxidation and spontaneous combustion. More accurate infrared tests on coal samples with particle sizes of 0.28–0.20 mm showed that the number of –OH and C=O groups and the side chain lengths increased with increasing water soaking duration.^{31,32}

Furthermore, the period of immersion and air-drying could change the spontaneous combustion oxidation characteristics of coal.^{33–35} Lu et al.³⁶ used raw coal and coal samples soaked in water for seven years. The results suggested that the coal accumulated many active sites during long-term water leaching and could be oxidized at low temperatures. In addition, the soaking process increased the expected gas yield of the soaked coal, reduced the crossing point temperature, and increased the spontaneous combustion risk of immersed coal.^{37–40} Various gases, such as CO, CO₂, CH₄, C₂H₆, C₂H₄, C₂H₂, C₃H₈, etc., will be generated in the process of coal spontaneous combustion. The type, generation rate, and generation temperature of gases indirectly reflect the process of coal spontaneous combustion, and the law of gas generation will change after soaking in water.^{41–44} However, gas release

behaviors during the spontaneous combustion of coal with different degrees of water immersion cannot be simply analyzed because the gas production rate is constantly changing at different temperature stages.^{45–47}

Based on the above factors and the stress distributions of the residual coal in goafs of underground coal mines, large lump coal samples were crushed by axial compression in this study. Crushed coal samples with different particle sizes were selected as the experimental materials, and the coal samples were soaked in water. Finally, coal samples soaked for different periods were drained of water, and temperature-programmed spontaneous combustion experiments were carried out. The temperature and gas concentration were measured, and the oxygen consumption rate, gas production rate, and heat release rate were calculated. Accordingly, the influence of water immersion on the spontaneous combustion of coal in goafs was analyzed. This method can approximate the crushing process of mined-out residual coal under stress, and the particle size distributions of the coal samples were closer to those of real abandoned coal in goafs. The experimental results can more effectively and accurately reflect the macroscopic spontaneous combustion characteristics of goaf residual coal after immersion.

2. RESULTS AND DISCUSSION

2.1. Analysis of Oxidizing Gas Concentration. In the temperature-programmed experiments of coal, the residual

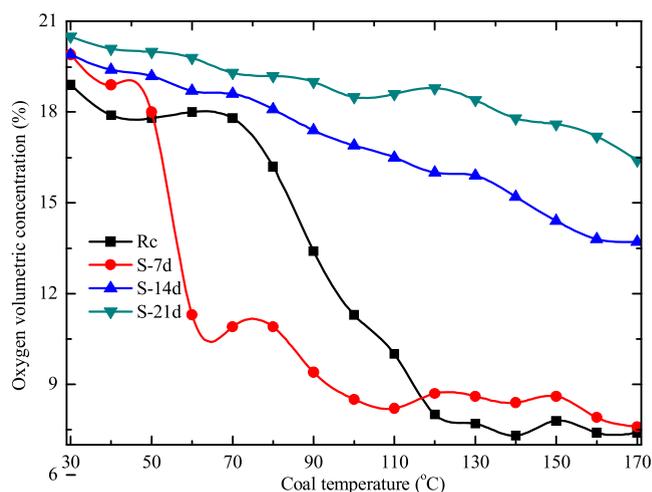


Figure 1. Volume concentration of O₂.

oxygen concentrations were varied. Figure 1 shows the oxygen concentration change law of coal samples that received different soaking treatments and raw coal.

In Figure 1, R_c denotes raw coal, and S-7d, S-14d, and S-21d denote raw coal soaked in water for 7, 14, and 21 days, respectively. Within the range of 30–60 °C, the volume concentration of O₂ of the raw coal was the lowest. In several coal samples soaked in water for different times, the longer the soaking time was, the higher the oxygen concentration became. Within the range of 60–110 °C, the volume concentration of O₂ of the raw coal was lower than those of S-21d and S-14d, and S-7d had the lowest concentration. Within the range of 110–170 °C, the volume concentration of O₂ of S-7d was lower than those of S-21d and S-14d, and raw coal had the lowest concentration. In general, for the coal samples soaked in

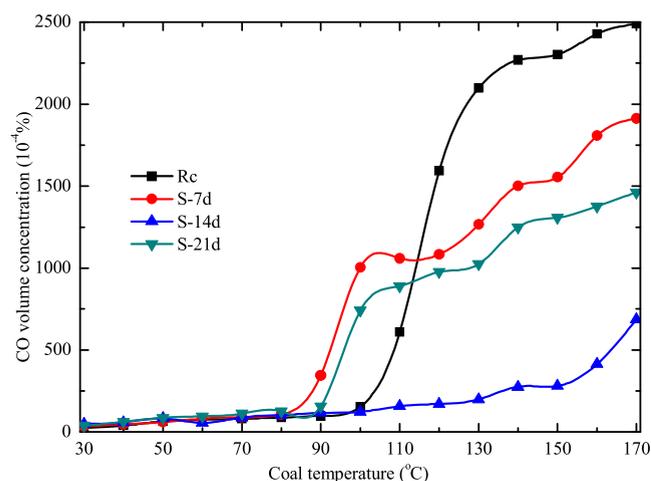


Figure 2. Volume concentration of CO.

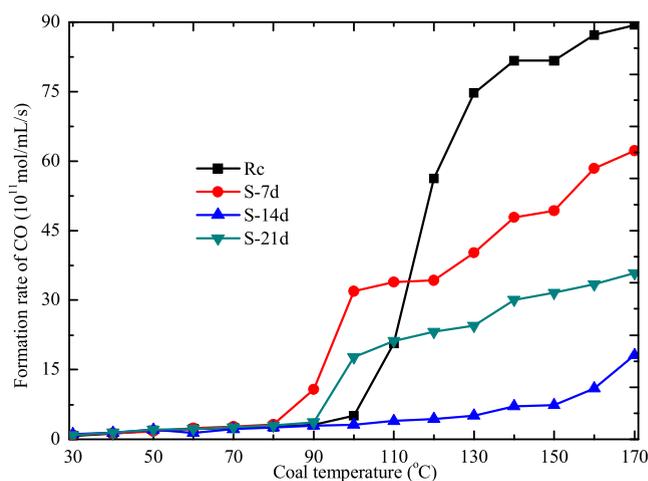


Figure 5. Generation rate of CO.

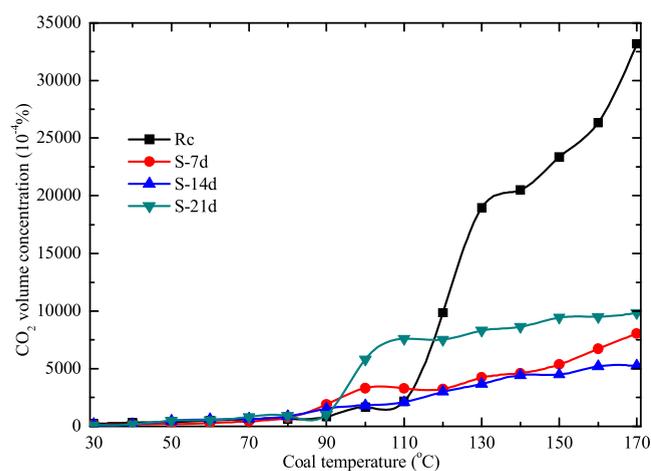
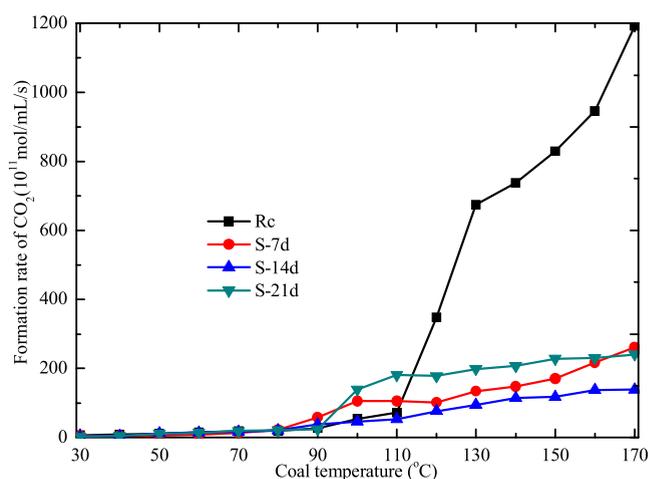
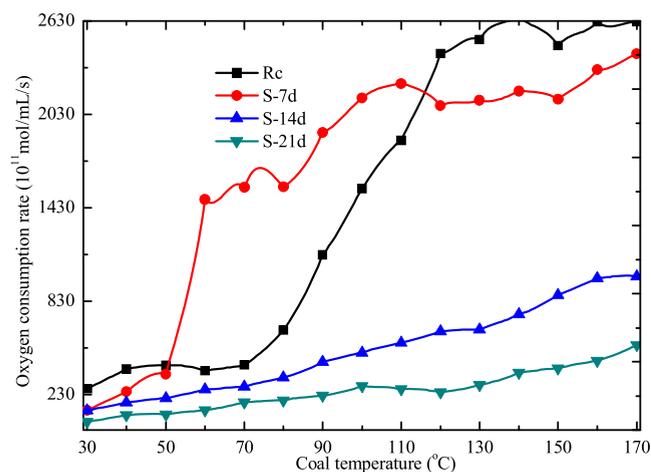
Figure 3. Volume concentration of CO₂.Figure 6. Generation rate of CO₂.

Figure 4. Oxygen consumption rate.

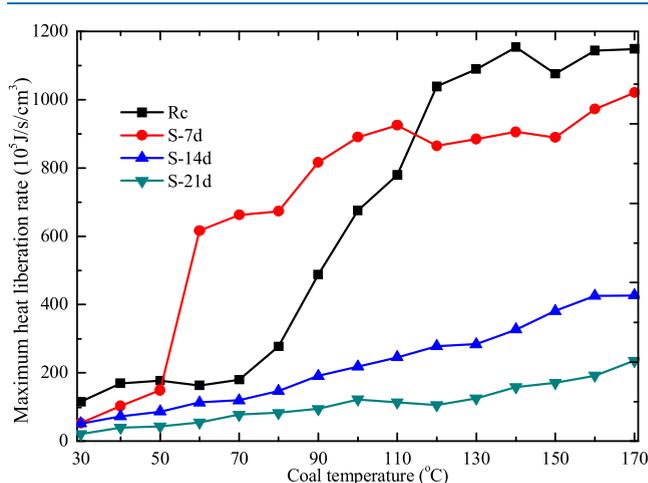


Figure 7. Upper limits of oxidation exothermic intensity.

water, the longer the soaking time was, the higher the oxygen concentration in the oxidation reaction process became.

Figure 2 shows the variation of carbon monoxide with temperature under different soaking conditions. When the temperature was lower than 90 °C, the carbon monoxide concentration was low, and there were no significant differences between different immersion treatments. Within

the range of 90–110 °C, the sample with the highest concentration of CO was S-7d, the second was S-14d, the third was Rc, and the lowest was S-14d. When the temperature was higher than 110 °C, the highest concentration of CO was Rc and the lowest was S-14d. In general, the concentration of CO was the highest for the coal samples soaked in water for 7 days.

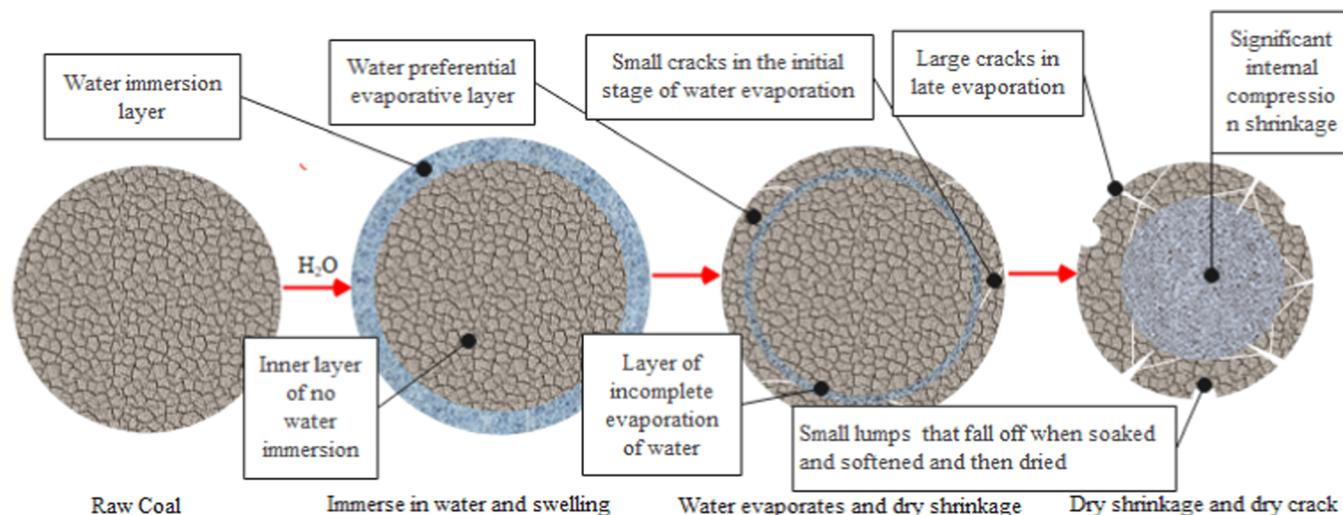


Figure 8. Process of coal soaking, drying, drying shrinkage, and drying cracking.

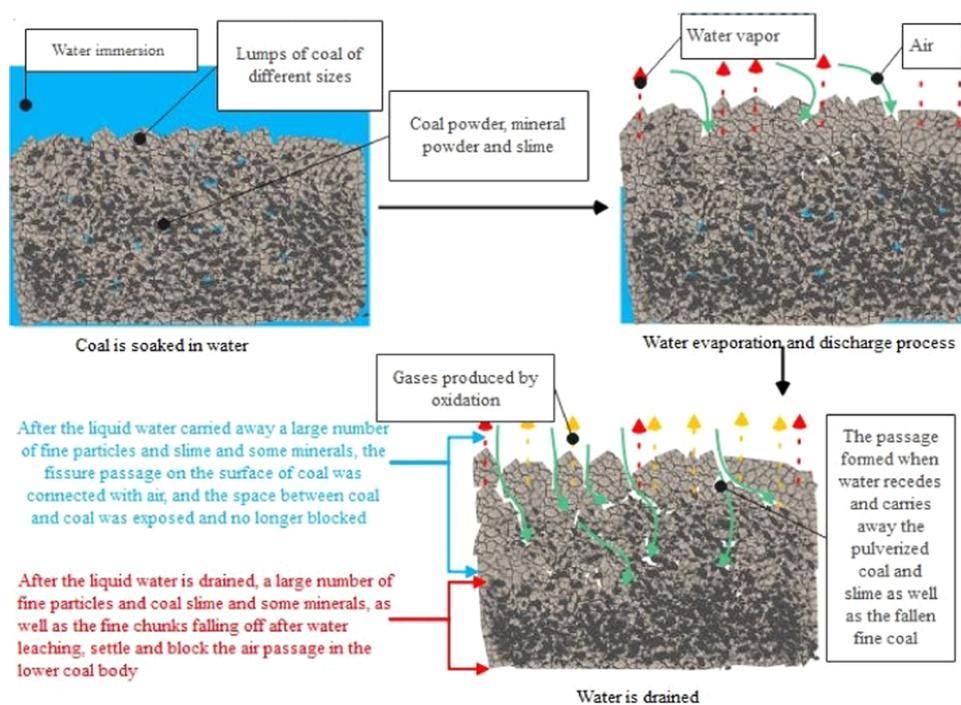


Figure 9. Formation process of air channels after coal dump or goaf is soaked and drained.

Consistent with the variation trend of the CO concentration with temperature, when the temperature of the coal sample was below 90 °C, the CO₂ concentration was low, and there was no significant difference between different treatments, as shown in Figure 3. When the temperature was higher than 110 °C, the CO₂ concentration of R_c was the highest.

2.2. Analysis of the Oxygen Consumption Rate and Generation Rate of CO and CO₂. The variations of the oxygen consumption rates of coal with temperature for the coal samples with different soaking treatments and raw coal are shown in Figure 4. The oxygen consumption rate increased as the temperature increased, and the lower the soaking time was, the higher the oxygen consumption rate became.

When the temperature of the coal sample was lower than 50 °C, the oxygen consumption rate R_c of coal was the highest, followed by that of S-7d, and that of S-21d is the lowest. Thus,

in the low-temperature oxidation stage, the oxygen consumption rate of the immersed coal was lower than that of raw coal due to water immersion. When the temperature of the coal sample was between 50 and 110 °C, the oxygen consumption rate of the coal sample soaked for 7 days was the highest, followed by those of raw coal, the coal soaked for 14 days, and the coal soaked for 21 days. When the coal temperature was higher than 110 °C, the oxygen consumption rate of the raw coal was the highest, which was significantly higher than that of soaked coal. Meanwhile, the longer the soaking time was, the lower the oxygen consumption rate of the coal sample became. When the temperature of the coal sample was higher than 110 °C, the oxidation capacity of the saturated coal sample was significantly lower than that of raw coal, and the longer the soaking time was, the more significantly the oxygen consumption rate was inhibited.

The variation in the oxygen consumption rates of the different coal samples in different temperature stages showed that the influence of the immersion of the coal sample on the coal oxidation process was complex. With the low temperature of 50 °C and the high temperature of 110 °C as the boundaries, the coal sample immersion at low and high temperatures led to the reduction of the coal sample oxidation capacity, and water immersion reduced the coal oxidation capacity. Soaking for 7 days promoted the improvement of the coal's oxidation capacity while soaking for a long time also led to a decrease in the coal's oxidation capacity. Whether soaking could promote or inhibit the oxidation capacity of coal was related to the soaking time or water quantity, and it was also significantly affected by the temperature of the coal sample.

The variation of the CO generation rate with temperature is shown in Figure 5. In the temperature range of 90–110 °C, the highest CO generation rate was for the coal soaked for 7 days. When the coal temperature was higher than 110 °C, the CO generation rate was the highest for raw coal.

The variations in the CO₂ generation rate with temperature are shown in Figure 6. When the temperature was higher than 90 °C, there was a gradual difference between different soaking times. The CO₂ generation rate of the coal soaked for 21 days was much higher than those of the others, and when the temperature was higher than 110 °C, the CO₂ generation rate of raw coal was the highest.

2.3. Analysis of Spontaneous Combustion Oxidative Exothermic Intensity. The variations in the upper limits of the oxidation exothermic intensities of the coal sample are shown in Figure 7. The variations in the upper limits of the oxidation exothermic intensity of different coal soaking times were like the variations in the oxygen consumption rate. When the coal temperature was lower than 50 °C, the upper limit of the oxidation exothermic intensity of R_c was the highest. When the temperature of the coal sample was between 50 and 110 °C, the upper limits of the oxidation exothermic intensity of the coal sample soaked for 7 days was the highest, followed by those of raw coal, coal soaked for 14 days, and coal soaked for 21 days.

The changes in the oxygen consumption rates with soaking time were examined. For coal samples with large particle sizes (the largest particle size was 20 mm, particles with sizes of 10–20 mm accounted for about 12% of the particles, and particles with sizes of 5–10 mm accounted for about 35% of the particles), when the temperature of the coal sample was lower than 50 °C, the intensity of oxygen oxidation reaction with the coal was low due to the large amount of water in the outer layer of the soaked coal. The oxygen consumption rate of immersed coal was lower than that of raw coal. When the coal temperature increased to between 50 and 110 °C, the oxygen consumption rate and heat release rate increased significantly and rapidly. However, the oxygen consumption rate and heat release rate of the coal samples soaked in water for 7 days were greater than those of raw coal, while those soaked in water for 14 and 21 days were lower than those of raw coal. When the coal sample temperature was increased to above 110 °C, the oxygen consumption rate of raw coal was the highest and was significantly higher than that of soaked coal. In this temperature stage, for the soaked coal, soaked for 7 to 21 days, the longer the immersion time, the lower the oxygen consumption rate and heat release rate. With the increase in temperature, the difference in the oxygen consumption rates

and heat release rates between the soaked coal and raw coal became larger and larger.

2.4. Discussion. In the low-temperature stage, the oxidation and heat release rates of the soaked coal were significantly inhibited by external moisture. When the temperature was lower than 50 °C, although the number of functional groups in the coal could be significantly increased because the functional groups could directly participate in oxidation, the above phenomenon occurred because the pore system of the coal was filled with water and the contact area between oxygen and coal, in which the oxidation reaction occurred, was small.

When coal was immersed in water for a certain period, the water content increased from the outside in, accompanied by significant swelling.¹⁶ When coal was under evaporation conditions, moisture loss via evaporation occurred gradually. As the moisture loss occurred, the coal began to exhibit different degrees of drying shrinkage and volume shrinkage, the surface exhibited many cracks, and the crack width, crack length, and humidity in the sample chamber gradually increased. Eventually, the moisture in the coal and the external environment reached a balance. Drying of the soaked coal produced significant shrinkage and created many cracks, and big internal particles or lump coal were gradually compressed due to drying shrinkage. The collapse of deep internal pores of the coal resulted in a more compact structure, as shown in Figure 8.

In the process of water loss of the immersed coal, many cracks occurred on the surface, the porosity and pore connectivity increased, and the air permeability was enhanced, which is consistent with the results of most of the water-immersion studies using smaller particles (1–3 mm) or even coal powder. However, the smaller particles or powder were studied from the perspective of the microscopic pore structure, revealing the reason for the coal spontaneous combustion risk after flooding increased. However, the actual residual coal in goaf (generally the median size is more than 15 mm) is larger than most of the coal sample particles were available for experimental testing. After soaking, the pore volume and connectivity increased as shown in Figure 8. The shrinkage and porosity decreased, enhancing the connectivity and increasing the airflow resistance. This phenomenon cannot be ignored in the production process. Furthermore, this phenomenon will also result in smaller coal granules or powder. In other words, for coal with large particle sizes, the microscopic research conclusions are not sufficient to completely reveal the influence of coal soaking on spontaneous combustion characteristics.

If the particles of waterlogged coal were larger, although the porosity and pore connectivity of shallow surfaces would increase after soaking, it would possibly increase the risk of spontaneous combustion. However, the internal coal body would shrink and become more compact, reducing the permeability, and thereby reducing the risk of spontaneous combustion. Therefore, for the abandoned coal or coal pile in goaf, attention should be paid not only to the influence of water immersion on the microstructure but also to the different influences on the surface and inner layers of the coal during the process of water immersion. Whether the final spontaneous combustion risk is increased or decreased depends on the balance of the two. If internal shrinkage and compaction play a dominant role, the risk of coal spontaneous combustion after soaking is reduced. If external wet swelling and drying cracking

play a dominant role, the risk of coal spontaneous combustion is enhanced by soaking.

The effect of soaking on the functional groups of coal was the same as that on the pore structure. Although the functional groups and activities of soaked coal will be enhanced and the risk of spontaneous combustion will increase, these results are obtained by powder testing and reveal the microscopic nature rather than the macroscopic nature. For large coal particles, the flooding process is still as shown in Figure 8, so for large coal particles, after a certain immersion time, the functional groups can significantly change. Over a short time, the effect of water on large coal particles is relatively weak, and the functional group changes are small. Even when submerged for a long time. The deep coal body inside the large-particle coal will not be significantly affected, in contrast to powder coal immersed in water. Thus, except for powder and fine coal, a few days or months of immersion would have no significant effect on the functional groups.

When the coal temperature increased to between 50 and 110 °C, the highest oxygen consumption and heat release rates were achieved by the coal sample soaked for 7 days, higher than those of raw coal. The oxygen consumption and heat release rates of raw coal were higher than those immersed in water for 14 and 21 days. Soaking for 7 days could promote the spontaneous combustion of coal, whereas soaking for 14 and 21 days could inhibit the spontaneous combustion of coal.

For the coal sample soaked for 7 days in this experiment, due to the short soaking time, the water mainly dissolved or washed away the powder and slime on the surface of the coal sample. When a large amount of external water evaporated, the surface of the coal sample could be fully exposed to the air. The blocked pores or cracks in the shallow surface were exposed, the contact area between the air and coal increased significantly, the range of pores extending into the deep coal increased, and the oxidation capacity was enhanced.

Slow oxidation of lump or large coal particles mainly occurs in the superficial layer. If the temperature is not high and the time is not long, internal oxidation is difficult, as shown in Figures 4 and 7. The maximum oxygen consumption rate and heat emission intensity variations as the temperature was varied are shown. When the temperature was above 50 °C, the oxygen consumption and heat release rates increased rapidly. When the temperature was higher, the upward trend of the two diminished and tended to become stable. Therefore, for the coal sample soaked for 7 days, when the temperature was higher than 110 °C, due to the rapid oxidation in the early stage, the oxidation capacity in the later stage was reduced, while the oxidation of raw coal in the early stage was relatively slow. The degree of oxidation in the later stage continued to increase significantly and was greater than that of the coal sample soaked for 7 days.

The oxygen consumption and heat generation rates of the coal samples soaked for 14 and 21 days were lower than those of raw coal, and the longer the soaking time was, the lower the oxidation and spontaneous combustion ability became. The main reason was that, although the flooding could improve the effectiveness of the moisture on the coal pore connectivity, it could also promote the oxidation of the functional group content. However, for large coal particles, after a short immersion period of 10 days, the above effects only occurred in the superficial layer and not in the whole coal body. In addition, during the drying process after soaking in water, the outer layer of the large particles or large blocks of coal were

significantly dried and cracked, and the inner layer was dry and compact, which would reduce the flow capacity, oxygen and coal contact area, oxidation capacity, and amount of heat released.

The oxidation and spontaneous combustion capacities of the coal samples soaked in water for 21 days were lower than those of coal samples soaked in water for 14 days. The main reason was that for large-particle coal samples, increasing the soaking time from 14 to 21 days could not significantly improve the total moisture content of the coal. However, it could lead to an increase in the water content and an improvement in the uniformity of the water distribution in the flooded layer. After the moisture outside the coal particles was drained, with the evaporation of internal water, the outer layer underwent a significant drying phenomenon, the permeability was enhanced, and the surface area increased. However, it also led to more significant shrinkage of the internal coal body and a worse internal permeability, which could lead to a reduction in the overall oxidation capacity and heat release.

At the same time, there was another reason. When the coal sample was immersed in water for a certain amount of time, the coal would undergo significant wet swelling, softening, and breakage, causing some fine particles to fall off. In the process of water discharge, the shed fine particles, coal powder, and slime on the surface of the original coal sample would move to the bottom of the coal with the water, resulting in the blockage of many spaces in the lower area of the coal sample, as shown in Figure 9. As a result, there would be difficulties in air intake and air circulation, which would lead to a lack of oxygen in some coal samples and low oxidation, resulting in the reduction of macroscopic oxygen consumption rate and heat release rate of oxidation.

In the process shown in Figure 9, the coal pile was submerged in and then drained of water. Similarly, the permeable residual coal in a goaf was submerged in and then drained of water. After the liquid water carried away many fine particles, slime, and some minerals, the sizes of the fissure passages in the upper part of the drainage area of the coal increased, and the residual coal in the goaf related to air, and the coal was exposed and no longer blocked. The increase in the sizes in the airflow channels, the increase in the number of functional groups of the coal samples soaked in water, and the changes of the pore structures in the coal after soaking resulted in an enhanced oxidation capacity and risk of spontaneous combustion. After the liquid water was drained, many fine particles, coal slime, and some minerals, as well as the fine chunks that fell off after water leaching, settled and blocked the air passages in the lower coal body, resulting in a reduced oxidation capacity and risk of spontaneous combustion.

If more fine particles fall off after immersion and there are more fine coal particles and slime attached to the surface of lump coal, for the coal pile, the airflow channels in the upper and lower parts will change after water immersion and water drainage, which will reduce the area prone to spontaneous combustion. For the abandoned coal in a goaf, this is equivalent to reducing the thickness of the residual coal that can spontaneously ignite. As a result, the oxidation temperature of residual coal cannot be sustained, and the risk of spontaneous combustion is significantly reduced. Therefore, for coal in the actual production process, because the particles are much larger than the particles of coal samples used in the previous microscopic experiments, the microscopic influence of water immersion on the coal spontaneous combustion

characteristics is not applicable for describing the real macroscopic phenomena.

The effect of water on the oxidation and spontaneous combustion characteristics of large coal particles was different from that of water on powder or fine coal. After powder or fine coal was fully immersed in water, pores developed, the number of active groups increased, and the oxidation and spontaneous combustion characteristics were enhanced. However, influenced by the coal particle size, soaking time, and water penetration depth, the increase in the water content in the soaking process had significant differences from outside to inside. The effect of water on the pore structure and active groups of coal was only significant near the surface, and softening, shedding, and transport of the surface fine coal and powder by water occurred. However, most of the deep coal was weakly affected, and the large coal particles underwent significant nonuniform drying shrinkage and cracking in the process of water loss. Consequently, the spontaneous combustion of large coal particles by short-term soaking was mainly inhibited. Soaking could also promote spontaneous combustion under specific soaking times and coal sample temperatures. Accordingly, for the residual coal in goaf and the coal involved in daily production, storage, and transportation with a certain content of large coal particles, the changes of the spontaneous combustion characteristics after being affected by water cannot be predicted by the test results of powder or fine coal alone. The heterogeneity of the water distribution in coal caused by the differences in the coal particle sizes and the change of the pore structures and gas circulation in the whole coal pile or goaf during water evaporation should also be fully considered.

When the fine-particle or powder coal was soaked in water, the number of functional groups increased significantly, the permeability increased, and the risk of spontaneous combustion increased. However, for the coal pile or the residual coal in a goaf dominated by large-particle coal, to find whether the risk of spontaneous combustion was aggravated, we needed to consider the degree of soaking, nonuniform drying shrinkage, and drying cracking after soaking. It was also necessary to consider the influence of water immersion and drainage on the transport and distribution of soluble minerals and organic small molecules, fine powder coal attached to the coal surface, and slime, as well as the resulting influence on the air passage of the coal pile or residual coal in goaf. These macroscopic effects may lead to a reduction in the macroscopic oxidation capacity and spontaneous combustion risk of coal with a higher content of large particles. In the process of studying the influence of water on the spontaneous combustion characteristics of coal, fine powder cannot be used alone for the experimental tests of water immersion and spontaneous combustion characteristics. Because this can only explain the microscopic influence mechanisms, and the particles in the spontaneous combustion of coal in actual production are much larger than the particles of coal samples required for experimental tests (e.g., thermogravimetric analysis, differential scanning calorimetry, gas chromatography, mass spectrometry, and Fourier-transform infrared spectroscopy), it is necessary to pay enough attention to the macroscopic changes in the large-particle coal affected by water. These macroscopic changes may significantly affect the air circulation characteristics of saturated coal and the size of coal–oxygen contact surface.

This paper studies the process, adopts the method of temperature programming to carry out the flooding of coal

spontaneous combustion characteristics and process test, can from the macroscopic angle to reveal the influence of different flooding degree of coal spontaneous combustion rule, but because of immersion time is limited, and, in this study with lignite as experiment material, only the results only effective for short-term immersion lignite. Follow-up study, therefore, need for different kinds of coal research on coal spontaneous combustion characteristics change after immersion, and adding different kinds of coal, coal under the conditions of different soaking time and moisture distribution of pore structure research, at the same time increase the functional test, from the macroscopic and microscopic phase together to reveal the influence law of water immersion of coal spontaneous combustion characteristics. Only in this way can the research conclusion have a more extensive application value.

In the study, the temperature-programmed method was adopted to test the characteristics and process of the spontaneous combustion of immersed lignite. It can reveal the influence of different immersion degrees on coal spontaneous combustion from a macroscopic angle. However, due to the limited soaking time and the fact that only lignite is used as the experimental material in this study, the research results are only effective for short-term soaking lignite. Therefore, in the follow-up study, it is necessary to carry out research on the variation rules of coal spontaneous combustion characteristics after soaking in water for different types of coal. In addition, the pore structure and water distribution law of coal under different types of coal and different soaking times are studied, and the functional group test is added to reveal the influence of soaking water on coal spontaneous combustion characteristics from the macro and micro stages. Only in this way can the research conclusion have a more extensive application value.

3. CONCLUSIONS

An experimental study on the spontaneous combustion of soaked crushed coal after water immersion was carried out using a temperature-programmed method to clarify the macroscopic influence of water immersion on coal spontaneous combustion characteristics. The oxygen consumption, gas generation, and oxidative heat release rates of raw coal with particle sizes less than 20 mm and coal samples with different soaking times were analyzed. The main conclusions are as follows:

- (1) After compression crushing, the spontaneous combustion characteristics of coal samples with particle sizes less than 20 mm changed after immersion. The differences were mainly in the characteristics of different temperature stages. When the temperature was lower than 50 °C, the oxygen consumption and heat release rates were lower than those of raw coal. Due to water immersion, the surface water of the coal was relatively high, fully blocking coal's contact with air, and the evaporation of water eliminated considerable heat. Hence, when the temperature of the coal was lower than 50 °C, water immersion inhibited the low-temperature oxidation and spontaneous combustion of coal.
- (2) In the coal temperature range of 50–110 °C, for the large coal particles soaked in water for 7 days, water could not significantly enter the coal body, and more structures and functional groups in the coal body could not be significantly changed. After the loss of a small

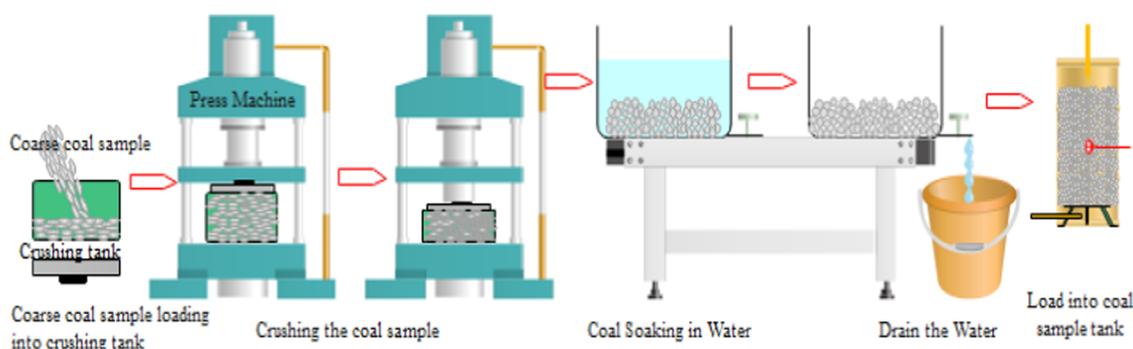


Figure 10. Coal sample preparation process.

Table 1. Particle Size Compositions of the Samples

number	R_c	S-7d	S-14d	S-21d
particle size range	mass scale (%)			
20–10–mm	12.3	10.4	13.9	11.7
10–5 mm	31.5	32.6	31.9	33.8
5–2.5 mm	17.2	18	17.7	17.4
2.5–1.0 mm	19.1	20.4	19.6	20
1.0–0.5 mm	8.8	8.8	8.8	8.4
0.5–0.25 mm	5.8	5.6	6.4	6
0.25–0.18 mm	1.4	1.5	1.3	1.5
0.18–0.15 mm	0.8	1	0.2	0.5
0.152–0.1 mm	1	0.5	0.1	0.3
<0.1 mm	2	1	0.2	0.5

Table 2. Coal Sample Processing 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	1554	0.41
soaked for 7 days (S-7d)	1404	20.9	13.3	1641	0.43
soaked for 14 days (S-14d)	1378	21.4	14.1	1680	0.46
soaked for 21 days (S-21d)	1433	22.2	15.5	1743	0.46

amount of water on the surface, the permeability of air was enhanced, and the surface area for the oxidation reaction increased. Consequently, the oxygen consumption rate and heat release rate of coal after 7 days of soaking in water were significantly higher than those of raw coal. In this temperature range, the oxygen consumption rates, and heat release rates of the coal samples soaked for 14 and 21 days were lower than those of raw coal.

- (3) When the coal temperature was 110–170 °C, the maximum soaking time was 21 days, and the water had a limited penetration depth in the coal, so its influence on

the coal body was limited to a certain depth on the surface. The depth and moisture content of water entering the coal could not change significantly as the soaking time was prolonged to 21 days. However, the distribution was more uniform in the shallow coal where water entered, leading to a significant increase in the nonuniformity of the water content and distribution in the deep and shallow coal. In the process of water evaporation, the nonuniform drying shrinkage and drying cracking of the coal body were more significant, the internal drying shrinkage was more significant, and the air permeability was worse. Thus, extending the soaking time within 21 days reduced the overall oxidation and heat release capacity of coal.

4. MATERIALS AND METHODS

4.1. Materials. The samples of lignite in this experiment were obtained from the Fengshuigou Coal Mine of the Pingzhuang Coal Company in Inner Mongolia, China. The process of coal sample crushing preparation is the same as refs 7 and 16, and crushing and soaking are shown in Figure 10, the water used for soaking is distilled water.

The particle size composition is shown in Table 1. And the canning parameters of coal samples are shown in Table 2, the mass (m), volume (V), height (H), porosity (n), and moisture content (M_c) of each coal sample in the copper coal tank are shown in Table 2.

4.2. Temperature-Programmed Experimental Method. The temperature-programmed experimental system shown in Figure 11 was established. The heating rate of the temperature-programmed furnace during the experiment was 1 °C/min. Gas samples were collected and analyzed by gas chromatography as the temperature was varied under an air atmosphere from 30 °C to the maximum temperature of the coal samples at intervals of 10 °C to determine the

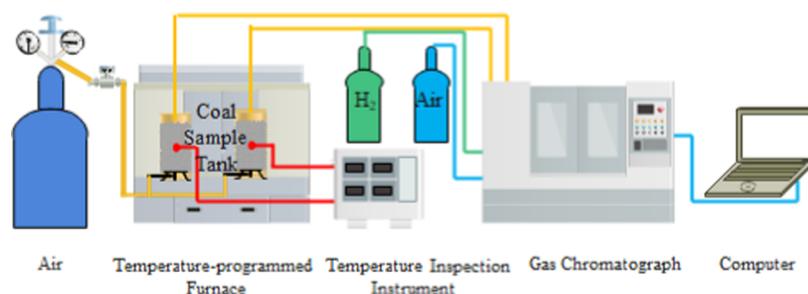


Figure 11. Temperature-programmed experimental system.

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.

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Notes

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REFERENCES

- (1) Onifade, M.; Genc, B. Spontaneous combustion of coals and coal-shales. *Int. J. Min. Sci. Technol.* **2018**, *28*, 933–940.
- (2) Fan, Y. J.; Zhao, Y. Y.; Hu, X. M.; Wu, M. Y. A novel fire prevention and control plastogel to inhibit spontaneous combustion of coal: Its characteristics and engineering applications. *Fuel* **2020**, *263*, No. 116693.
- (3) Shao, J.; Zosky, G. R.; Hall, G. L.; Wheeler, A. J.; Dharmage, S.; Melody, S.; Dalton, M.; Foong, R. E.; O'Sullivan, T.; Williamson, G. J.; et al. Early life exposure to coal mine fire smoke emissions and altered lung function in young children. *Respirology* **2020**, *25*, 198–205.
- (4) Guo, J.; Wen, H.; Zheng, X. Z.; Liu, Y.; Cheng, X. J. A method for evaluating the spontaneous combustion of coal by monitoring various gases. *Process Saf. Environ.* **2019**, *126*, 223–231.
- (5) Hao, Y.; Xie, T. P. Coupling Treatment of Gas Drainage and Nitrogen Injection for Fire Prevention in the First Stratified Mining goaf: Theoretical Calculation and Field Experiment. *Combust. Sci. Technol.* **2021**, *126*, No. 1895774.

- (6) Hu, G. Z.; Xu, J. L.; Ren, T.; Gu, C. X.; Qin, W.; Wang, Z. W. Adjacent seam pressure-relief gas drainage technique based on ground movement for initial mining phase of longwall face. *Int. J. Rock Mech. Min. Sci.* **2015**, *77*, 237–245.

- (7) Dong, Z. W.; Sun, L. F.; Jia, T. G.; Guo, S. L.; Geng, W. L.; Yu, W. H.; Pi, Z. K.; Li, G. L.; Luo, C. Rapid contrastive experimental study on the adiabatic spontaneous combustion period of loose lignite. *ACS Omega* **2021**, *6*, 34989–35001.

- (8) Xu, Y. L.; Bu, Y. C.; Liu, Z. J.; Lv, Z. G.; Chen, M. L.; Wang, L. Y. Effect of the reignition characteristics on long-flame coal by oxidation and water immersion. *Environ. Sci. Pollut. Res.* **2021**, *28*, 57348–57360.

- (9) Shi, Q.; Qin, B.; Bi, Q.; Qu, B. An experimental study on the effect of igneous intrusions on chemical structure and combustion characteristics of coal in Daxing Mine, China. *Fuel* **2018**, *226*, 307–315.

- (10) Roy, D.; Singh, G.; Seo, Y. Coal mine fire effects on carcinogenicity and non-carcinogenicity human health risks. *Environ. Pollut.* **2019**, *254*, No. 113091.

- (11) Lin, S.; Liu, Z.; Qian, J.; Li, X. Comparison on the explosivity of coal dust and of its explosion solid residues to assess the severity of re-explosion. *Fuel* **2019**, *251*, 438–446.

- (12) Xu, Q.; Yang, S.; Yang, W.; Tang, Z.; Hu, X.; Song, W.; Zhou, B. Microstructure of crushed coal with different metamorphic degrees and its low-temperature oxidation. *Process Saf. Environ. Prot.* **2020**, *140*, 330–338.

- (13) Wang, G.; Xie, J.; Xue, S.; Wang, H. Laboratory study on low temperature coal spontaneous combustion in the air of reduced oxygen and low methane concentration. *Teh. Vjesn.* **2015**, *22*, 1319–1325.

- (14) Song, S. *Experimental Study on the Effect of Water Immersion on Structures and Spontaneous Combustion Characteristics of Coal*; China University of Mining and Technology, 2019.

- (15) Kan, L. *Experimental Study on the Promotion of Low-Temperature Oxidation of Low-Rank Coal by Water Immersion Process*; China University of Mining and Technology, 2019.

- (16) Dong, Z. W.; Yu, W. H.; Jia, T. G.; Guo, S. L.; Geng, W. L.; Peng, B. Experimental Study on the Variation of Surface Widths of Lignite Desiccation Cracks during Low-Temperature Drying. *ACS Omega* **2021**, *6*, 19409–19418.

- (17) Wen, G.; Yang, S.; Liu, Y.; Wu, W.; Sun, D.; Wang, K. Influence of water soaking on swelling and micro characteristics of coal. *Energy Sci. Eng.* **2020**, *8*, 50–60.

- (18) Fry, R.; Day, S.; Sakurovs, R. Moisture-induced swelling of coal. *Int. J. Coal Prep. Util.* **2009**, *29*, 298–316.

- (19) Zheng, K. Y.; Yang, Y. L.; Miao, G. D.; Li, P. R. Influencing Mechanism of Water Soaking Process on Spontaneous Combustion Characteristics of Goaf Residual Coal. *J. Combust. Sci. Technol.* **2020**, *27*, 665–674.

- (20) Qin, B. T.; Song, S.; Qi, X. Y.; Zhong, X. X.; Liu, C. Effect of soaking process on spontaneous combustion characteristics of long-flame coal. *J. China Coal Soc.* **2018**, *43*, 1350–1357.

- (21) Li, F.; An, S. G.; Xing, Z. Q. Experimental study on pore structure and spontaneous combustion characteristics of submerged coal. *Coal Sci. Technol.* **2019**, *47*, 208–212.

- (22) He, X. Q.; Liu, X. F.; Song, D. Z.; Nie, B. S. Effect of microstructure on electrical property of coal surface. *Appl. Surf. Sci.* **2019**, *483*, 713–720.

- (23) Qin, Y. P.; Song, Y. P.; Liu, W.; Wei, J.; Lv, Q. L. Assessment of low-temperature oxidation characteristics of coal based on standard oxygen consumption rate. *Process Saf. Environ.* **2020**, *135*, 342–349.

- (24) Lu, Y. Laboratory study on the rising temperature of spontaneous combustion in coal stockpiles and a paste foam suppression technique. *Energy Fuels* **2017**, *31*, 7290–7298.

- (25) Qiao, L.; Deng, C. B.; Zhang, X.; Wang, X. F.; Dai, F. W. Effect of soaking on coal oxidation activation energy and thermal effect. *J. China Coal Soc.* **2018**, *43*, 2518–2524.

- (26) Qin, B. T.; Song, S.; Qi, X. Y. Effect of soaking process on spontaneous combustion characteristics of long-flame coal. *J. China Coal Soc.* **2018**, *43*, 1350–1357.
- (27) Deng, C. B.; Qiao, L.; Wang, X. F.; Dai, F. W.; Zhang, X. Spontaneous combustion characteristics and infrared analysis of soaked lignite. *China Saf. Sci. J.* **2018**, *28*, 105–110.
- (28) Yuan, J.; Zhu, H. Q.; Zhao, J. L.; Zhu, X. M.; Wang, H. R. The effect of soaking in increasingly alkaline aqueous solutions on the spontaneous combustion characteristics of bituminous coal. *Fire Mater.* **2021**, *35*, No. 3034.
- (29) Lu, W.; Sun, X. L.; Gao, L. Y.; Hu, X. M.; Song, H. Z.; Kong, B. Study on the characteristics and mechanism of DL-malic acid in inhibiting spontaneous combustion of lignite and bituminous coal. *Fuel* **2022**, *308*, No. 122012.
- (30) Song, Y. W.; Yang, S. Q.; Song, W. X.; Zhang, Z. C.; Yang, K.; Jiang, X. Y.; Zhou, Q. C.; Zhang, D. P. Adsorption Characteristics of Soaked Air-dried Coal and Reaction Characteristics of Free Radical Functional Groups in CH₄-containing Oxidizing Atmosphere. *Combust. Sci. Technol.* **2020**, *44*, No. 1806253.
- (31) Zhai, X. W.; Ge, H.; Wang, T. Y.; Shu, C. M.; Li, J. Effect of water immersion on active functional groups and characteristic temperatures of bituminous coal. *Energy* **2020**, *205*, No. 118076.
- (32) Yong, L. X.; Yun, C. B.; Meng, L. C.; Lan, Y. W. Effect of Water-immersion and Air-drying Period on Spontaneous Combustion Characteristics for Long-flame Coal. *Combust. Sci. Technol.* **2020**, No. 1788007.
- (33) Guo, J.; Wen, H.; Zheng, X. Z.; Liu, Y.; Cheng, X. J. A method for evaluating the spontaneous combustion of coal by monitoring various gases. *Process Saf. Environ. Prot.* **2019**, *126*, 223–231.
- (34) He, X. Q.; Liu, X. F.; Nie, B. S.; Song, D. Z. FTIR and Raman spectroscopy characterization of functional groups in various rank coals. *Fuel* **2017**, *206*, 555–563.
- (35) Onifade, M.; Genc, B.; Rupprecht, S. Spontaneous combustion liability between coal seams: A thermogravimetric study. *Int. J. Min. Sci. Technol.* **2020**, *30*, 691–698.
- (36) Lu, W.; Li, J. H.; Li, J. L.; He, Q. L.; Hao, W. S.; Li, Z. H. Oxidative kinetic characteristics of dried soaked coal and its related spontaneous combustion mechanism. *Fuel* **2021**, *305*, No. 121626.
- (37) Onifade, M.; Genc, B. Spontaneous combustion liability of coal and coal-shale: a review of prediction methods. *Int. J. Coal Sci. Technol.* **2019**, *6*, 151–168.
- (38) Song, S.; Qin, B. T.; Xin, H. H.; Qin, X. W.; Chen, K. Exploring effect of water immersion on the structure and low-temperature oxidation of coal: A case study of Shendong long flame coal, China. *Fuel* **2018**, *234*, 732–737.
- (39) Wang, K.; Liu, X. R.; Deng, J.; Zhang, Y. N.; Jiang, S. R. Effects of pre-oxidation temperature on coal secondary spontaneous combustion. *J. Therm. Anal. Calorim.* **2019**, *138*, 1363–1370.
- (40) Xiaowei, Z.; Wang, B.; Wang, K.; Obracaj, D. Study on the influence of water immersion on the characteristic parameters of spontaneous combustion oxidation of Low-Rank Bituminous coal. *Combust. Sci. Technol.* **2019**, *191*, 1101–1122.
- (41) Guo, J.; Wen, H.; Zheng, X.; Liu, Y.; Cheng, X. A method for evaluating the spontaneous combustion of coal by monitoring various gases. *Process Saf. Environ. Prot.* **2019**, *126*, 223–231.
- (42) Xi, X.; Shi, Q.; Jiang, S.; Zhang, W.; Wang, K.; Wu, Z. Y. Study on the effect of ionic liquids on coal spontaneous combustion characteristic by microstructure and thermodynamic. *Process Saf. Environ. Prot.* **2020**, *140*, 190–198.
- (43) Xu, Q.; Yang, S.; Hu, X.; Song, W.; Cai, J.; Zhou, B. Low-temperature oxidation of free radicals and functional groups in coal during the extraction of coalbed methane. *Fuel* **2019**, *239*, 429–436.
- (44) Zhang, Y.; Wang, J.; Wu, J.; Xue, S.; Li, Z.; Chang, L. Modes and kinetics of CO₂ and CO production from low-temperature oxidation of coal. *Int. J. Coal Geol.* **2015**, *140*, 1–8.
- (45) Wu, Y. G.; Zhang, Y. L.; Wang, J.; Zhang, X. Y.; Wang, J. F.; Zhou, C. S. Study on the Effect of Extraneous Moisture on the Spontaneous Combustion of Coal and Its Mechanism of Action. *Energies* **2020**, *13*, No. 1969.
- (46) Sun, L. L.; Zhang, Y. B.; Wang, Y.; Liu, Q. Q. Study on the Reoxidation Characteristics of Soaked and Air-Dried Coal. *J. Energy Resour. Technol.* **2019**, *141*, No. 022203.
- (47) Huang, Z. A.; Li, J. Y.; Gao, Y. K.; Shao, Z. L.; Zhang, Y. H. Thermal Behavior and Microscopic Characteristics of Water-soaked Coal Spontaneous Combustion. *Combust. Sci. Technol.* **2020**, No. 1777993.